Fuel 150 (2015) 288–297

Contents lists available at ScienceDirect

Fuel

journal homepage: www.elsevier.com/locate/fuel

Self-sustaining smouldering combustion of coal tar for the remediation of contaminated sand: Two-dimensional experiments and computational simulations

Tanzeer Hasan [a,b], Jason I. Gerhard [a,*], Rory Hadden [a,c], Guillermo Rein [d]

[a] Department of Civil and Environmental Engineering, University of Western Ontario, London, Canada
[b] Department of Geology, University of Dhaka, Dhaka, Bangladesh[1]
[c] Institute for Infrastructure and Environment, University of Edinburgh, Edinburgh, United Kingdom[1]
[d] Department of Mechanical Engineering, Imperial College London, London, United Kingdom

ARTICLE INFO

Article history:
Received 1 September 2014
Received in revised form 2 February 2015
Accepted 5 February 2015
Available online 15 February 2015

Keywords:
Smouldering
Remediation
Multiple dimensions
Modelling
Thermal severity

ABSTRACT

This study presents the development and validation of a computational model which simulates the propagation of a smouldering front through a porous medium against unique experiments in coal tar and sand. The model couples a multiphase flow solver in porous media with a perimeter expansion module based on Huygens principle to predict the spread. A suite of two-dimensional experiments using coal tar-contaminated sand were conducted to explore the time-dependent vertical and lateral smouldering front propagation rates and final extent of remediation as a function of air injection rate. A thermal severity analysis revealed, for the first time, the temperature–time relationship indicative of coal tar combustion. The model, calibrated to the base case experiment, then correctly predicts the remaining experiments. This work provides further confidence in a model for predicting smouldering, which eventually is expected to be useful for designing soil remediation schemes for a novel technology based upon smouldering destruction of organic contaminants in soil.

1. Introduction

Fuels such as gasoline, diesel, crude oil and coal tar, are deposited in large amounts in the subsurface at tens to hundreds of thousands of sites due to poor historical disposal practices and ongoing accidental releases [1]. The presence of such Non-Aqueous Phase Liquids (NAPLs) in the subsurface pose human health and environmental hazards because of their toxicity and persistence. Despite the availability of several approaches, many NAPL remediation efforts fail to meet clean-up objectives with respect to the amount of contaminant removed, the time required, and/or the cost. Coal tar and heavy hydrocarbons remain among the most challenging to remediate: their long chain, complex, and multicomponent chemical structures resist transformation and breakdown by thermal, biological and chemical approaches [2]. When cleanup of these sites is required, excavation and disposal to a hazardous waste landfill or incinerator is typical, representing costly and unsustainable options.

STAR (Self-sustaining Treatment for Active Remediation) is a remediation technology that exploits the energy content of liquid organic contaminants, such as coal tar and heavy hydrocarbons, to achieve their in situ destruction by smouldering combustion. Smouldering is a flameless form of combustion, which propagates as an exothermic reaction wave driven by heterogeneous oxidation reactions (i.e., gas phase oxygen and condensed phase fuel) [3,4]. In general, smouldering combustion occurs within a solid porous medium which can be a permeable aggregate of particles, grains, or fibres where the fuel is either a combustible component of the porous matrix or a separate substance embedded in it [4]. The reaction can be self-sustaining in the presence of sufficient fuel, sufficient oxygen and limited heat losses [5,6].

Numerous porous solids can sustain a smouldering reaction, including polyurethane foam, coal, tobacco, dust, peat and wood [4]. The smouldering of polyurethane foam, in particular, has been extensively studied in the field of fire safety science (e.g. [7–9]). The majority of smouldering research has been conducted using one-dimensional (1D) experiments. These studies examined the effect of such parameters as air flux, buoyancy and moisture

* Corresponding author at: Department of Civil and Environmental Engineering, The University of Western Ontario, Spencer Engineering Building, Rm. 3029, London, Ontario N6A 5B9, Canada. Tel.: +1 (519) 661 4154; fax: +1 (519) 661 3942.

E-mail addresses: thasan7@du.ac.bd (T. Hasan), jgerhard@uwo.ca (J.I. Gerhard), r.hadden@ed.ac.uk (R. Hadden), g.rein@imperial.ac.uk (G. Rein).

[1] Present address.

http://dx.doi.org/10.1016/j.fuel.2015.02.014
0016-2361/

content on the propagation rate and temperature of the smouldering reaction [6,7,10,11]. In a 1D scenario, it is convenient to define the propagation direction of a smoulder reaction relative to the flow direction of the oxidizer. In forward smouldering, the reaction propagates in the same direction as the oxidizer flow, while in opposed smouldering the reaction propagates in the opposite direction [3,4]. This has important implications for the heat and mass transfer during smouldering and consequently the reaction rate and temperature.

In many real systems, this simplification is no longer valid and smouldering propagation cannot be considered 1D. There are few multi-dimensional smouldering studies available in the literature but the process is expected to be influenced by many factors including ignition source geometry, fuel geometry and the influence of buoyancy [3]. Ohlemiller [3] summarised the available studies which included cellulose-based fuels (e.g., cardboard, shredded tobacco) examined under different fuel geometrics and with either buoyancy-induced air flows through the porous matrix or forced air flow across the free surface. These conditions were studied because they represent typical smouldering scenarios in a fire safety context. These studies revealed, for example, that the opposed smoulder velocity in cellulosic insulation was significant but weakly related to air flow rate applied across the top of the sample; meanwhile the velocity of the forward smouldering reaction on the surface was high and decreased with depth into the sample [12]. To the authors' knowledge, there are no publications on the spread of smouldering in two dimensions with a forced airflow applied through the porous medium instead of across the free surface.

Smouldering of organic liquids embedded in an inert porous medium has been studied in relatively few contexts. Fire flooding involves igniting a combustion reaction in deep wells to produce heat and gas waves for secondary oil recovery [13]. In contrast, STAR initiates smouldering to consume the organic liquids (NAPLs) and thereby remediate the sand [14]. The process is expected to be energy and cost efficient because, within the right parametric bounds, smouldering is self-sustaining; in other words, once initiated with a short, local energy input for ignition, the process requires no additional external energy to continue propagating. Switzer et al. [15] demonstrated STAR across a range of sands and NAPLs with 1D bench-top experiments. Pironi et al. [16] used column experiments to investigate the sensitivity of the STAR process to key parameters for coal tar and crude oil. Overall, those studies demonstrated that smouldering destruction is self-sustaining for a wide range of relatively non-volatile NAPLs in silts and sands when their concentrations in sands are above a minimum threshold (10,000–15,000 mg/kg at the bench scale). Even sands containing highly volatile NAPLs, which could not otherwise support a self-sustaining smouldering reaction, can be treated with STAR by injecting vegetable oil as a supplementary fuel [17]. These studies further demonstrated that, the sand through which the reaction passed exhibits non-detect concentrations of hydrocarbons, and the forward smouldering propagation rate linearly increased with the injected air flow rate. Switzer et al. [18] illustrated that STAR performed equally well when scaled up 1000 times in a 3 m^3 test. Field pilot testing for in situ and ex situ applications of STAR are currently underway.

Numerical modelling is an important part of the process for designing and optimising remediation systems. Numerical models of smouldering combustion are mainly 1D to reproduce the phenomenon observed in experimental forward or opposed propagation modes [5,6,8,19–24]. However, two-dimensional (2D) models have been developed [25,26] as well as one considering three dimensions [27]. These models aim to solve the heat and mass transfer processes that occur in a porous media as well as the chemical reactions and species generation and consumption. In all cases, simplifying assumptions are used to reduce the complexities associated with this multiphysics problem. Even with these simplifications, multi-physics numerical models of smouldering are computationally intense even for simulations at small (e.g., ~10 cm) scales [8]. As such, they are unsuitable for the field scale (~10 m to 100 m) domains required to explore smouldering as a site remediation technology.

MacPhee et al. [28] developed a novel, phenomenological approach to modelling smouldering with particular application to NAPLs in porous media. The ISSM (In Situ Smouldering Model) coupled a multiphase (air, NAPL) flow in porous media model and a geometric combustion front expansion model. This approach allows prediction of the movement of the propagating smouldering front as a function of sand permeability, contaminant concentration, and air flux, with little computational demand. The low computational cost means that the model is suitable for application at the field scale even accounting for heterogeneous porous media. A disadvantage is that it requires calibration based on smouldering experiments for each fuel and sand type. The ISSM was calibrated against one-dimensional smouldering experiments for coal tar and crude oil in coarse sand [28]. Numerical testing of the ISSM verified that the model was robust under a variety of expected applications, numerically stable and computationally efficient [28]. MacPhee et al. [28] provided some demonstration 2D simulations, but acknowledged those results were speculative due to an absence of experimental data for validating 2D smouldering combustion predictions.

The main objective of this study was to compare the ISSM to 2D NAPL smouldering experiments and thereby increase confidence in its predictions of smouldering propagation in multidimensional scenarios. Eight experiments were conducted to evaluate the 2D smouldering behaviour of coal tar in sand for different air injection rates. A key difference between this study and previous smouldering research is the use of forced air injection that produces a multidimensional air flow field within the porous medium; this configuration was chosen as it is similar to how STAR is applied in the field. The experiments were quantified in terms of the vertical and lateral rate of smouldering front propagation as well as the overall extent of remediation. In addition, a thermal severity analysis was conducted to identify the critical temperature-residence time combinations that correspond to remediation of coal tar-contaminated sand. The ISSM was calibrated against one of the experiments for lateral combustion front propagation phenomena not previously observed. The calibrated model was then employed for independent simulations of the remaining experiments.

2. Methods

2.1. Experiments

Two-dimensional smouldering propagation experiments were carried out in a steel box 370 mm long × 300 mm high × 205 mm deep (Fig. 1). NAPL-contaminated sand was prepared by mixing commercially available quartz sand (Number 12, Bell & MacKenzie Co., mean grain diameter = 0.88 mm, coefficient of uniformity = 1.6) with commercial grade fresh coal tar (Alfa Aesar, density = 1180 kg/m^3 at 20 °C). All of the experiments were conducted using a coal tar concentration of 71,000 mg/kg sand (NAPL saturation ~ 25%), which corresponds to typical field values and matches experiments previously conducted in 1D [16]. It is noted that, for smouldering studies, fresh coal tar is an appropriate surrogate for the weathered coal tar typically found at contaminated sites because both are dominated by high molecular weight compounds [29] that are involved in the exothermic (combustion) reactions occurring above 500 °C [30].