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Development and evaluation of a continuous microwave processing system for hydrocarbon removal from solids

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HIGHLIGHTS

- A continuous microwave processing system for treatment of contaminated soils is demonstrated.
- Degree of organic removal was assessed as a function of power and energy input.
- Applying the power in a single stage was the most energy efficient.
- Hydrocarbon removal is higher using two stage heating but required more energy.

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ABSTRACT

A continuous conveyor-belt processing concept using microwave heating was developed and evaluated. Four hydrocarbon-contaminated soils were used as model feedstocks, and the degree of organic removal was assessed against the power and energy input to the process. The findings of this study at scale (150 kg/h) are in direct agreement with data obtained in batch laboratory scale experiments, and show that microwave heating processes are fundamentally scalable. It is shown that there is a trade-off between the efficiency of organic removal and the power distribution, and applying the power in a single stage was found to be 20–30% more energy efficient but the overall degree of organic removal is limited to 60%. 75% removal was possible using two processing steps in series, but the organic removal is ultimately limited by the amount of power that can be safely and reliably delivered to the process material. The concept presented in this work is feasible when 75% organic removal is sufficient, and could form a viable industrial-scale process based on the findings of this study.

1. Introduction

Many papers are published each year reporting promising microwave experiments in a plethora of applications, and yet the conversion of these to industrial scale microwave processes is low [1]. A lack of data from pilot and demonstration systems often prevents successful scale-up because the technical risks have not been fully evaluated and mitigated. In recent years, a number of pilot-scale and commercial prototype systems have been developed for applications that exploit the selective and volumetric heating benefits offered by microwave heating [2–4]. These processes are continuous, and have been developed via multidisciplinary collaboration between experts in solids handling, process engineering and electromagnetics. There are numerous engineering challenges that need to be overcome in order to successfully implement the scale-up of microwave heating processes, in part because the 'engineering science' within this field is not fully established. One such challenge relates to electric field breakdown, or arcing, which can result in very high localised temperatures and damage to the microwave and materials handling systems. Arcing occurs in gases as they can only sustain an electric field of relatively low intensity compared to liquids and solids. For example, the breakdown voltage in air is 33 kV/cm at ambient conditions, compared to 30,000 kV/cm for water [5]. The breakdown voltage reduces significantly at higher temperatures and when droplets/-particulates are present within the gas [6]. The physics of arcing is well understood, but the mitigation of this effect in continuous processing systems often presents the final barrier in moving from prototype to full scale commercial implementation.

This study reports the development and evaluation of a continuous, pilot-scale microwave processing system for the removal of hydrocarbons from solid materials, and highlights the key challenges in operating at scale and the mitigating steps that can be

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employed. There are numerous applications to which this process can be applied, however the specific focus of this study is on using hydrocarbon contaminated soils as model feedstocks for process evaluation.

The UK government has estimated that around 300,000 hectares, or 2% of the UK land mass can be classed as contaminated. This contamination can take the form of heavy metals, asbestos, radionuclides and organic compounds [7]. Sites deemed to have unacceptable soil contaminant concentrations can undergo risk management, involving removal of the source, interruption of exposure pathways (containment) or management of receptor behaviour, or some combination of the three [8]. In situ remediation options often involve soil vapour extraction (SVE) [9], sometimes allied to heating methods such as six phase soil heating or steam injection to increase volatilisation, or target less volatile contaminants [10]. More aggressively, a range of oxidising agents, such as ozone, persulphates or peroxides may be injected into the subsurface to treat both soil and groundwater [11]. Biodegradation methods, particularly for hydrocarbon contamination, can also be used for in situ treatment [12]. Ex situ methods include mechanical and thermal treatment. Mechanical processing usually involves soil washing [13]. Thermal desorption may also be used for some of the most grossly contaminated sites, such as former gasworks, and involves heating the soil to between 100 and 800 °C to volatilise heavy organics [14]. This is generally confined to large, centralised processing facilities and may be unsuitable for the remediation of smaller-scale sites.

Microwave heating has previously been shown to remove organic contaminants from soils [15], and may be a viable alternative to conventional thermal desorption processes. Microwaves heat selectively and volumetrically. This unconventional heating mechanism gives rise to a range of potential benefits in the processing of contaminated soils: heating rates can be three orders of magnitude higher than conventional processes [6], leading to smaller (potentially portable) processing equipment and flexible operating schedules; microwaves target water that is bound within the soil, and this leads to the ability to process soils containing significant amounts of clay minerals [16], which are difficult to process using conventional soil remediation techniques, principally due to their low hydraulic conductivity [17].

Microwave processing of contaminated soils has the potential to offer step change improvements in soil remediation. Previous studies have shown that Poly-Aromatic Hydrocarbons (PAHs) could be efficiently removed from two contaminated soils samples [15] and Horikoshi et al. [18] showed that significant removal of trichloroethylene was possible from a model soil material. The levels of organic removal depend on the electric field intensity generated within the microwave applicator, and the subsequent power density within the heated phase(s) of the soil. High electric field intensities lead to high power densities, and correspond to higher levels of organic removal than low field intensities.

1.1. Underpinning mechanism

Previous work identified that the mechanism of organic removal from soil using microwaves arises from selectively heating the aqueous phase and not directly the organic contaminant phases themselves [2,3,16]. Table 1 shows the dielectric properties of a number of relevant materials. The minerals presented could typically be expected to be found in a soil sample. The dielectric loss (ε'') can be considered a measure of a materials ability to dissipate absorbed microwave energy as heat. The higher the dielectric loss, the better the material will heat in an applied field. Materials with a loss of <0.005 can be considered microwave transparent [6].

So in a mixture of bulk soil, hydrocarbon contaminants and water, it is the water phase that will be heated selectively through the volume of the solid. Providing that sufficient power density is achieved in the soil, rapid microwave heating converts the water in the soil to steam, which then strips the organic contaminants from the soil. This effect has also been observed in microwave heating of contaminated drill cuttings [2,3] and oil sands [16], which are structurally similar to contaminated soils, given they contain mineral matter, oil and water. Previous work reported in [16] has shown that while the bulk temperature of a hydrocarbon contaminated soil remains around 100 °C, PAHs with boiling points 218–400 °C are completely removed and in excess of 90% those with a boiling point above 400 °C are removed. This can be attributed to selective heating of the water phase, generating localised temperatures in the soil structure sufficient to entrain hydrocarbon contaminants into the exhaust vapour. In the work reported in [16], a lab-scale single mode batch system was shown to be effective for extracting naturally occurring hydrocarbons from oil shales. However, this system used only a relatively low power (3 kW) and small samples masses (150 g).

The aim of this study is to build on previous laboratory-scale reports and design and test a pilot scale continuous microwave treatment process using soils as a model feedstock, and asses the feasibility of developing the concept to an industrially relevant scale at 2–3 tonnes per hour. The objectives of this work are:

1. Characterise the electric field distribution within the microwave processing system, and understand the interaction of the soil feedstocks with the electric field
2. Establish the processing conditions for stable process operation, and determine the process efficiency under these conditions
3. Investigate the feasibility of in-series processing to improve process robustness, and understand the impact on process efficiency.

2. Materials and methods

2.1. Soil sampling

Four bulk soil samples were obtained from contaminated industrial sites within the UK. The samples were transported from site in skips in 2-tonne batches, and mixed for 4 h using a Meyer 6002 soil mixer, which can act as both a mixer and a feeding system. After the mixing process, the mixer was set into its feeding position and allowed to discharge 100 kg of material. A sample of homogenised material was taken using the cone and quarter method [19] to 5 kg, following by riffling to 5 g for compositional analysis.

2.2. Sample analysis

The water content and total organic content were the parameters used to characterise the performance of the system under trialled operating conditions. The water content was established using the Dean & Stark method [20], which involved reflux distillation with Toluene followed by the subsequent separation and recording of the immiscible water phase. The organic content was determined by extracting the organic phase from the soil

Table 1
Dielectric properties of materials at 25 °C and 2.45 GHz (adapted from [2]).

Material	Dielectric constant (ε')	Dielectric loss (ε'')
Water	77	13
Fuel oil	2.0	0.002
Quartz	3.8	0.001
Mica	1.6	0.005
Feldspar	2.6	0.002