



Enhancing evaporative mass transfer and steam stripping using microwave heating



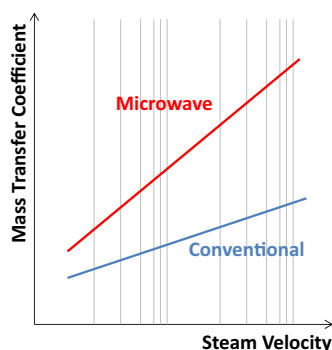
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HIGHLIGHTS

- Hydrocarbon removal is exclusively related to steam velocity during microwave heating.
- Stripping is the dominant mechanism, with the stripping media generated in-situ.
- Enhanced mass transfer coefficients with microwave heating compared to conventional processes.
- In-situ steam generation overcomes the channelling issues that limit conventional stripping processes.

GRAPHICAL ABSTRACT



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ABSTRACT

The effect of microwave heating on evaporative mass transfer of hydrocarbons was investigated for a number of contaminated solid materials. The rate of oil removal could be rationalised by the velocity of steam that was created by selective heating of water within the solid. A single correlation was found to fit 45 independent experiments across 10 separate variables, and the correlation was consistent with the physics of evaporative mass transfer. It is shown for the first time that steam stripping is the dominant mechanism that governs hydrocarbon removal during microwave processing. It was further discovered that mass transfer is enhanced due to microwave heating when compared to conventional stripping processes, with this improvement in efficiency due to the ability of the microwave process to overcome the channelling effects that limit conventional mass transfer processes.

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1. Introduction

Hydrocarbon-containing soils pose a threat to health and to the environment. They arise from legacy industrial activity, where hydrocarbon containing wastes were disposed of with minimal treatment. These are typically found on industrial complexes, fuel storage and coal-gas sites and also within harbour sediments. In these cases there is a need to remediate the legacy hydrocarbon contamination via in-situ or ex-situ treatment processes. More

recent industrial processes that produce hydrocarbon contaminated solids occur within the oil & gas and steel industries, with modern practice and legislation requiring that these wastes are effectively treated to remove the hydrocarbon phase prior to their disposal or reuse. In most cases, whether for legacy sites or current industrial practice, the technologies used to separate the contaminant hydrocarbons are based on thermal desorption or gas stripping. Electrical heating and air stripping have been explored for in situ remediation of contaminated soil (Lord, 1998; Buettner and Daily, 1995), and thermal desorption of oil-contaminated drill cuttings via a heated screw conveyor is widely practised within the oil industry (Hui et al., 2005). Raising the temperature of the solid

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Nomenclature

a	specific interfacial area (m^2/m^3)	u	velocity (m/s)
A	cross-sectional area (m^2)	V	bed volume (m^3)
c	gas phase concentration (mol/m^3)	W	water mass (kg)
d_p	particle diameter (m)	ε	porosity
D_a	diffusion coefficient (m^2/s)	μ	viscosity (Pa·s)
J	hydrocarbon flux ($\text{mol}/\text{m}^2\cdot\text{s}$)	ρ	density (kg/m^3)
k_g	mass transfer coefficient (m/s)	Pe	Peclet number
\dot{N}	hydrocarbon molar flowrate (mol/s)	Re	Reynolds number
t	time (s)	Sc	Schmidt number
		Sh	Sherwood number

increases desorption (Missen et al., 1999) due to an increase in the vapour pressure of the hydrocarbons relative to the partial pressure within the contacting gas.

Microwave heating has been studied as an alternative desorption technique for a number of applications, and it has been shown that this approach can be more energy efficient than conventional systems due to the mechanism through which microwave energy desorbs the hydrocarbon phase. The hydrocarbon and water phases coexist on the surface and within the pore structure of the solid (Robinson et al., 2008). During microwave processing it is only the water that absorbs microwaves, with steam generated in-situ from within the capillaries and pores of the solid material. Previous studies have highlighted gains in efficiency with this approach for applications including contaminated drill cuttings, contaminated soils and oil sands. A number of studies have speculated that entrainment, steam stripping or steam distillation are possible mechanisms that explain empirical hydrocarbon removal results (Robinson et al., 2010, 2012, 2014; Buttress et al., 2016; Shang et al., 2006; Di et al., 2000), but none have offered conclusive evidence to date. There is also ambiguity within the literature, with studies using terminology around steam stripping and steam distillation interchangeably, not recognising that they are based upon different physical principles. Steam distillation occurs when the boiling point of the hydrocarbon phase is reduced due to the presence of an immiscible phase, in this case water. With steam distillation the hydrocarbon is boiled, whereas with steam stripping the hydrocarbon is removed by evaporation into the stripping gas media. The aim of this study is to establish the extent to which steam-stripping occurs during microwave remediation of hydrocarbon-contaminated solids by measuring mass transfer coefficients for hydrocarbon removal and comparing with existing mass transfer correlations.

Previous workers have studied the rate of mass transfer of hydrocarbons from soils to a gas stripping medium and derived empirical models relating mass transfer coefficient with the velocity of the stripping medium (Wilkins et al., 1995; Yoon et al., 2002; van der Ham and Brouwers, 1998). They studied the effect of introducing gas from an external source into a partially saturated contaminated soil media. Wilkins et al. (1995) noticed that the residual water content and water distribution affected the mechanism of hydrocarbon mass transfer into the stripping gas, which could either be governed by diffusive or convective transport. In this scenario, hydrocarbon droplets may become trapped between aqueous phases in small pores, restricting movement into the gas phase. This phenomenon is known as channelling, where the stripping medium bypasses the trapped organic phase, and is a common disadvantage of gas stripping in conventional fixed bed systems. Yoon et al. (2002) corroborated the findings of Wilkins et al. (1995) by showing that hydrocarbon mass transfer can become diffusion rate-limited, even at high gas velocities. van der Ham and Brouwers (1998) studied non-equilibrium mass

transfer into a steam stripping medium, and found that the interfacial area between steam and the hydrocarbon phase may decrease during treatment of the contaminated soil. In all cases, hydrocarbon removal was confirmed to take place via an evaporative mass transfer process, with the bulk gas temperature being below the end-point of the hydrocarbon phase.

Mass transfer coefficients in fixed beds can be linked to the velocity of the stripping gas in a generalised correlation (Comiti et al., 2000):

$$Sh = bRe^c Sc^{0.33} \quad (1)$$

where Sh is the Sherwood number, Re is the Reynolds number, Sc is the Schmidt number, b and c are constants determined empirically. The Sherwood and Reynolds Numbers are defined as:

$$Sh = \frac{k_g d_p}{D_a} \quad (2)$$

$$Re = \frac{\rho u d_p}{\mu} \quad (3)$$

k_g is the gas phase mass transfer coefficient, d_p is the characteristic length (usually defined as the mean particle diameter), D_a is the diffusion coefficient of the contaminant in the stripping medium, ρ is the density of the stripping medium, u is the velocity of the stripping medium and μ is the viscosity of the stripping medium.

The mass transfer coefficient can be estimated from Fick's law (Sherwood et al., 1975):

$$J = k_g(c_i - c_b) \quad (4)$$

where J is the net molar flux of the species desorbed from the fixed bed into the gas phase, c_b is the concentration of the desorbed species in the bulk gas phase, and c_i is the concentration of the desorbed species at the interface with the stripping gas, which is related to its vapour pressure. The partial pressure at the interface is assumed to equal the vapour pressure at the stripping temperature. Partial pressure values are subsequently converted to gas phase concentration (mol/m^3) to yield k_g values (m/s) when the molar flux is known or measured. Within fixed beds it is often not possible to measure molar flux due to uncertainties in the interfacial area of the solid particles within the bed, which typically exhibit a large size distribution and poorly-defined interstitial regions. In previous studies (Wilkins et al., 1995; Yoon et al., 2002; van der Ham and Brouwers, 1998), the relationship between the mass transfer coefficient and gas velocity in fixed beds of soil has been presented in a modified version of Eq. (1) as follows;

$$Sh_0 = bPe^c \quad (5)$$

where Sh_0 is the modified Sherwood number and Pe is the Peclet number.

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