

Contents lists available at ScienceDirect

Chemical Engineering Journal



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

Remediation of oil-contaminated drill cuttings using continuous microwave heating

J.P. Robinson^{a,*}, S.W. Kingman^a, C.E. Snape^a, R. Barranco^a, H. Shang^a, M.S.A. Bradley^b, S.M. Bradshaw^c

^a School of Chemical and Environmental Engineering, University of Nottingham, University Park, Nottingham NG7 2RD, UK

^b Wolfson Centre for Bulk Solids Handling, University of Greenwich, UK

^c Department of Process Engineering, University of Stellenbosch, South Africa

ARTICLE INFO

Article history: Received 18 December 2007 Received in revised form 29 April 2009 Accepted 8 May 2009

Keywords: Oil-contaminated drill cuttings Microwave heating Environmental Remediation

ABSTRACT

This paper outlines a novel, continuous pilot-scale microwave treatment process for the remediation of oil-contaminated drill cuttings from North Sea drilling activities. The underlying scientific methodology is highlighted, and the development of the continuous processing concept is discussed. Continuous tests were carried out at throughputs up to 250 kg/h using microwave hardware capable of delivering up to 15 kW of microwave power at 2.45 GHz. It is shown that the cuttings can be remediated to below the offshore discharge threshold of 1% oil on cuttings, and that clean-up can occur to below 0.1% oil, which is the classification for non-hazardous waste. The trade-off between applied power, throughput and residual oil content is shown for tests carried out continuously over several hours. The energy consumption of the process is also shown in relation to the remediation levels achieved. The results obtained with the continuous pilot-scale system are compared with previous batch laboratory studies.

© 2009 Elsevier B.V. All rights reserved.

1. Introduction

Oil-contaminated drill cuttings arise from drilling activities in the exploration and extraction of oil and natural gas. Drilling fluids known as 'muds' are used to lubricate the drill bit, provide hydraulic power and transport the drill cuttings back to the production platform [1]. The separated cuttings were formerly discharged straight into the sea without further treatment, which lead to the degradation of the marine environment in the vicinity of the platform due to the hydrocarbon contamination. Environmental legislation for the UK now demands that oil levels must be <1% by weight for discharge to take place [2]. In contrast the discharge limits in the Gulf of Mexico are 5% [3]. Drill cuttings samples obtained from the North Sea, and produced using oil-based muds (OBM) can contain 5-15% oil, meaning that treatment is required before disposal. Recent landfill directives [4] and concerns about transporting cuttings to shore mean that an offshore treatment process is desirable, and microwave treatment has been identified as a candidate technology.

1.1. Previous studies of microwave heating

Along with our previous work in this area [5–7] there are several review articles and texts which introduce the concepts of microwave heating, for example [8,9], so the basics will not be discussed in detail here. There are two principal advantages of microwave heating in process engineering applications:

- 1. Selective heating, meaning that energy does not have to be wasted bulk-heating an entire material volume.
- 2. Volumetric heating, meaning that energy can be dissipated instantaneously beyond the surface of a material, overcoming heat transfer limitations.

Selective heating accounts for the energy efficiency of many microwave processes compared with conventional heating. Volumetric heating can result in very fast heating times, leading to compact equipment with high material throughputs and low residence times.

Previous microwave-based studies of drill cuttings treatment were carried out at small scales using single mode or multimode systems [5,7–9]. The main findings of these studies relate to the mechanisms of oil removal from the contaminated cuttings. Microwaves do not heat the oil directly as the oil is essentially transparent at microwave frequencies since it has a dielectric loss factor of 0.002 [6]. Instead, the water within the pores of the cuttings is heated and converted to steam. As the steam escapes it physically entrains the oil, which exists at the surface of the cuttings fragments. Other potential mechanisms have also been identified such as stripping and steam distillation [7], however the entrainment mechanism is the most thermodynamically attractive since energy

^{*} Corresponding author. Tel.: +44 0115 9514092; fax: +44 0115 9514115. *E-mail address:* john.p.robinson@nottingham.ac.uk (J.P. Robinson).

^{1385-8947/\$ –} see front matter 0 2009 Elsevier B.V. All rights reserved. doi:10.1016/j.cej.2009.05.008

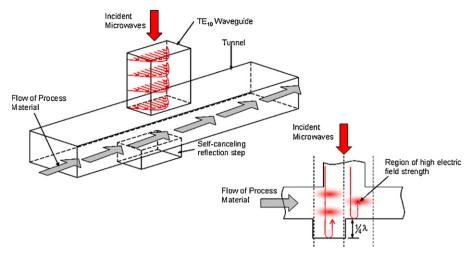


Fig. 1. Concept of a transverse E-field applicator.

only needs to be supplied to the water, and not to the surrounding rock fragments or oil.

To maintain the thermodynamic advantage of the microwave process, the water phase must be converted to steam before significant heat transfer takes place to the surrounding rock. To achieve this the heating rate $(\Delta T/\Delta t)$ must be as high as possible, and the heating rate can be equated with the power density as shown by Eq. (1) [5]:

$$\frac{\Delta T}{\Delta t} = \frac{Pd}{\rho C_p} \tag{1}$$

In moving from laboratory tests to a continuous treatment system, it is imperative that the power density is maximised to allow the rapid conversion of water to steam without significant heat loss to the surroundings. The requirement for a high power density leads to a corresponding requirement for a high-strength electric field, which in turn dictates the type of microwave cavity which is used.

1.2. Microwave cavities for continuous processing

Despite the high power densities obtainable, single mode cavities cannot be used for the vast majority of industrial applications because of their relatively small volume and the inherent nonuniformity of the electric field. A more useful concept for industrial applications which still allows high power densities is the transverse travelling wave applicator, which is an applicator falling into the category of broadband non-resonant devices. An example of such a system is a transverse E-field applicator with a self-cancelling reflection step, and this is shown in Fig. 1.

The process material is conveyed through a tunnel of rectangular cross-section, and the microwaves are fed orthogonally to the direction of the feed. A reflection step is used at the base of the cavity to offset the reflection, producing three overlapping regions of high power density. The variation in power density is largely overcome by the overlap of the three hot spots, so process material will experience a region of high power density irrespective of its vertical position. The orthogonal feed of the waveguide means that the power density is uniform across the width of the cavity also. Transverse E-field applicators yield a high power density and a power density distribution which is relatively even across both the width and height of the cavity.

This is the first study that reports a continuous microwave treatment system for the removal of oil from drill cuttings.

2. Experimental and materials

The cavity used for this work is shown in Fig. 2.

The applicator was designed so that it could be assembled around a rectangular-troughed conveyor belt. It was constructed from aluminium in two sections, which when assembled around the conveyor formed the cavity in the centre, with a choking section on either side. The purpose of the chokes is to stop microwaves propagating out of the cavity into the surrounding environment. The cavity was integrated into a pilot-scale testing facility as shown in Fig. 3.

The continuous pilot-scale system consists of a 5–30 kW variable power microwave generator, which delivers microwaves at 2.45 GHz to the cavity via several sections of WR430 waveguide and an automatic E–H tuner. The tuner works by varying the geometry in the E and H planes to match the impedance of the microwaves with that of the cavity, with an algorithm used to vary the geometry so as to minimise the reflected power. Any reflected power is absorbed in the circulator, which uses a cold water load, and this protects the magnetron and power supply from excessive returned microwave energy. A stream of cold nitrogen was introduced at 5 l/min down the waveguide entrance to the cavity, which was done to provide a positive pressure and prevent oil and water vapours from passing through the waveguide to the microwave generator.

The drill cuttings are fed from a feed hopper into a twin shaft mixer, where dry material could be introduced to control the moisture content of the feed to the microwave cavity. The mixer deposits the cuttings onto a conveyor belt, which was made from woven Nomex fibres, and formed into a trough to contain the process material. A heated nitrogen stream at 501/min was introduced at the material feed to act as a sweep gas, and also to provide an

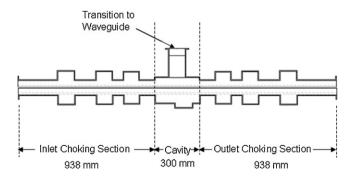


Fig. 2. Transverse E-field microwave applicator with self-cancelling reflection step.

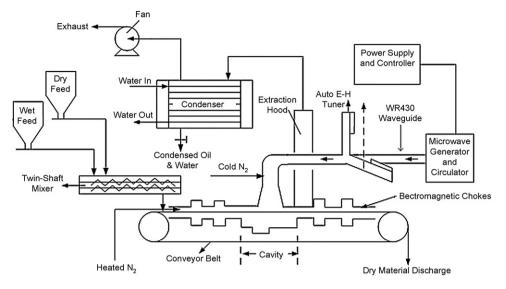


Fig. 3. Schematic of the pilot-scale apparatus for the continuous treatment of contaminated drill cuttings.

inert atmosphere within the cavity to prevent combustion of the oil vapour.

The top section of the cavity and chokes contains perforations which permit the withdrawal of evolved vapours whilst containing the microwave field. An extraction hood covers the perforated sections and the end of the choking section, and the vapours are drawn through a condenser to recover the oil and water. The dry drill cuttings are discharged at the end of the conveyor belt, and collected for analysis and disposal.

The oil and moisture contents of the treated and untreated drill cuttings were measured, and used to determine the degree of treatment. Water contents were measured using the Dean and Stark method (ASTM D-95), which involves reflux distillation with toluene and separation of the water phase. The oil content measurement was carried out using solvent extraction, which is a technique for extracting organics from solid samples with liquid solvents. The organics were extracted using dichloromethane (DCM) at elevated temperatures and pressures to increase the efficiency of the extraction process. Increased temperature accelerates the extraction kinetics, while elevated pressure keeps the solvent below its boiling point, thus enabling safe and rapid extractions. The hydrocarbon content of the organic phase was determined using gas chromatogram techniques.

The drill cuttings were obtained from southern North Sea drilling operations, and contained 10% oil and 10% water with shalebased rock fragments. Dry clay was added to reduce the oil and

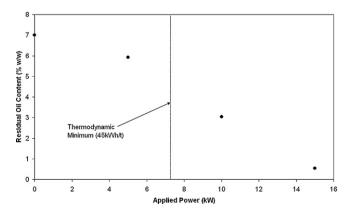


Fig. 4. Residual oil content plotted against applied power for a constant throughput of 160 kg/h.

water content of the cuttings to 7%, which altered the consistency of the cuttings from a slurry to a more permeable, agglomerated granular material.

The continuous treatment tests were carried out over a period of several hours, with three samples of treated material taken for analysis for each set of experimental conditions. The recovered oil was collected at the end of the complete test sequence. Experiments were carried out using an applied power of 5–15 kW, and cuttings throughputs of 100–200 kg/h. Based on the volume of high intensity electric field the space velocity in this case varied between 0.14 and $0.30 \, \text{s}^{-1}$.

3. Results and discussion

3.1. Effects of microwave power

Experiments were performed at power levels of 5, 10 and 15 kW with the material throughput fixed at 160 kg/h. In all the tests the reflected power achieved a stable value between 0.5 and 0.7 kW. In all experiments the average temperature within the bed of treated cuttings was measured at 95-105 °C at the outlet. This observation verifies that only water is heated, and that the subsequent oil removal occurs due to the formation of steam as previously described in [7]. The bulk residual oil contents of the processed cuttings were measured, and these results are shown in Fig. 4.

It can be seen that increasing the applied power results in improved oil removal, with the residual oil level decreasing steadily as the applied power is increased. Given that the reflected power remained relatively unchanged, almost all of the applied power was dissipated within the drill cuttings in the microwave cavity. It is known that the contaminant oil is effectively transparent to microwaves as it has a dielectric loss factor of less than 0.1 at room temperature and a frequency of 2.45 GHz [6]. The effect of increasing the applied power is to increase the power density in the absorbing phases within the material, which is the water that is contained within the pore structure of the rock cuttings. Previous studies showed the remediation mechanism to be the rapid conversion of interstitial water to steam in the first instance, which then entrains the contaminant oil from the surface of the cuttings and into the sweep gas [7]. This is the most thermodynamically attractive mechanism as the microwave energy can be targeted into the water phase alone, rather than heating the entire matrix of rock fragments, oil and water.

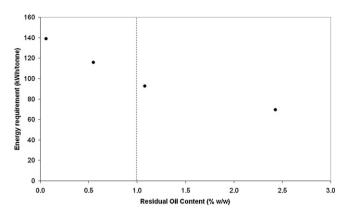


Fig. 5. Energy requirement as a function of the levels of remediation attained using an applied power of 15 kW.

It is possible to calculate the minimum energy required to remove the contaminant oil by assuming that all of the applied microwave energy raises the temperature of the water to 100 °C and subsequently overcomes the latent heat of vaporisation. Based on the latent heat of water of 2250 kJ/kg and an average heat capacity of 4.2 kJ/kg K, the minimum amount of energy required to convert all of the water to steam in a throughput of 160 kg/h can be established, and this figure is 45 kWh per tonne of drill cuttings. An applied power of 7.3 kW is the theoretical minimum at this throughput, and it can be seen in Fig. 4 that power levels below 7.3 kW do not induce significant levels of remediation, whereas at higher powers the residual oil levels are much lower.

The power levels required to reach the 1% environmental discharge threshold are approximately double the thermodynamic minimum, and this is likely to be due to the fact that some of the power is reflected, some goes into superheating the water above 100 °C before steam is formed, and some will inevitably be dissipated within the surrounding rock fragments and oil. Nonetheless, to desorb the oil by conventional heating uses much more energy because temperatures in excess of 250 °C are required, and the entire matrix of rock, oil and water must be heated to this temperature. The minimum energy requirements for a conventional heating process using the same feedstock as with this work are estimated in Table 1, assuming that all the oil vaporises at 250 °C.

On a theoretical basis the conventional thermal desorption process requires over three times as much energy as the microwave treatment process, at 163 kWh per tonne. A comparison can also be drawn with drying processes. Keey [10] reports that industrial convection dryers typically consume 1 kWh per kg of water removed, which would equate to 70 kWh per tonne to remove the water from the drill cuttings used in this work. Fig. 5 shows that the microwave process can remove the majority of both the water and oil using less than 100 kWh per tonne, which is indicative of the inherent efficiency improvements possible using microwave technology over conventional heating processes.

3.2. Effect of throughput

The throughput of drill cuttings was varied by changing the speed of the conveyor belt. The applied power was kept constant at 15 kW, and the throughput varied between 110 and 220 kg/h. This corresponded to belt speeds of 10–20 mm/s and residence times within the electric field of 5–10 s. The bulk residual oil content was determined for each set of conditions, and these results are shown in Fig. 6.

Decreasing the material throughput at constant power leads to a linear decrease in residual oil content, and hence improves the overall cuttings treatment process. Decreasing the throughput increases

the residence time in the area of the cavity which supports the high intensity electric fields, which allows more of the available electromagnetic energy to be dissipated within the water phase. The environmental discharge threshold can be achieved using 15 kW and process throughputs below 150 kg/h. Throughputs which are higher than this mean that the residence time in the electric field is too low for the creation of sufficient volumes of steam to remove the required levels of contaminant oil. When the process throughput is continually decreased below 150 kg/h the residual oil levels reduce further, and can fall to below 0.1%, which is the current threshold for classification as a hazardous material in the UK. The reflected power levels were again found to be relatively constant across the range of throughputs studied, with values of 0.4-0.7 kW recorded. It is postulated that decreasing the throughput below 100 kg/h at 15 kW will result in an increase in the reflected power as the residence time in the microwave cavity is likely to extend beyond that required to remove the water, meaning that the remaining cuttings effectively become microwave transparent. The electric field distribution is a strong function of the dielectric constant and dielectric loss factor of the process material. When the water is removed the dielectric loss factor decreases to below 0.1, meaning that much of the 15 kW of available microwave power will be transmitted through the material with very little being absorbed [11]. If we compare the data point for 10 kW in Fig. 4, with the extrapolated data in Fig. 6., we can see that in both cases we would achieve a residual oil content of around 3%. This suggests that in this data range at least, energy input is the determining factor, as with two different powers (10 and 15 kW) and different throughputs (and hence residence times) we get the same oil removal for the same energy input.

3.3. Quality of recovered oil

The base-oil in the drilling mud was characterised using gas chromatography. The oil and water recovered from the condenser were collected and the two phases allowed to separate under gravity. Three samples of the oil phase were removed for analysis, and typical results are shown in Fig. 7.

The peaks shown in the chromatogram in Fig. 7a indicate an abundance of C8–C16 hydrocarbons, with small traces of C17–C20 at longer elution times. The chromatogram of the recovered oil indicates the presence of some lighter hydrocarbons than were evident in the original oil, and less of the heavier hydrocarbons. Overall the composition is similar, and is not likely to have a significant impact on the oil properties for recycling within the drilling mud system. Nonetheless the presence of lighter hydrocarbons indicates that some thermal upgrading of the oil has occurred, possibly due to pyrolysis or stream cracking. Analysis of the residual oil in the

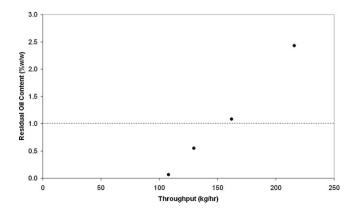


Fig. 6. Residual oil content plotted against throughput for a constant applied power of 15 kW. Dotted line represents environmental discharge threshold of 1% oil on cuttings.

462	
Table	1

Minimum energy	requirements f	for a conventiona	l heating process.
winning chergy	requirements i		I HEALINE DIOLESS.

Substance	Temperature range (°C)	Specific heat capacity (kJ/kgK)	Latent heat of vaporization (kJ/kg)	kg substance per kg cuttings	Energy required	
					(kJ/kg cuttings)	(kWh/tonne)
Rock	20-250	1.5	_	0.86	296.7	82.4
Oil	20-250	2	800	0.07	88.2	24.5
Water	20-100	4.2	2250	0.07	181	50.3
Steam	100-250	2	-	0.07	21	5.8
				Total	586.9	163.0

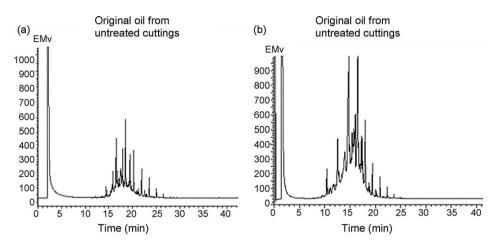


Fig. 7. Gas chromatograms of original oil from untreated drill cuttings and the recovered oil after microwave treatment.

treated drill cuttings was also carried out and this shows a greater abundance of the heavier species, which is shown in Fig. 8. Although there is some evidence of cracking the bulk temperature of the material on the conveyor belt was shown not to exceed 105 °C, therefore any thermal degradation is likely due to localised areas of high electric field strength within the cavity.

3.4. Overall mass balance

The continuous microwave treatment process is essentially a separation process, with a single feedstock and two product streams. The first product is dry rock fragments which contain residual water and oil, the power and residence time dictate the oil and water content of the dry stream. The second product is an oil/water stream which is condensed and the emulsion separated under gravity. There is very little thermal degradation of the oil and the oil quality does not vary over the studied range of power and

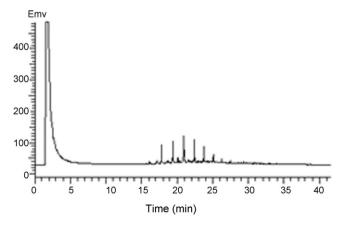


Fig. 8. Chromatogram of the residual oil extracted from treated cuttings.

material throughput. This observation is consistent with the steam stripping and entrainment mechanisms identified in previous work [5–7] as the bulk temperatures are not high enough to cause significant decomposition of the oil. Changes in power and material throughput affect only the efficiency of the separation process, i.e. the amount of oil recovered per unit energy input.

3.5. Energy requirements

The levels of remediation achieved were evaluated against the total microwave energy applied to the system, and this is shown in Fig. 5. Applying more microwave energy leads to an improvement in the remediation process as the residual oil content decreases with increasing energy input. In this case the 1% environmental discharge limit can be achieved using around 100 kWh per tonne of drill cuttings. Significantly cleaner cuttings can be produced at higher energy inputs of the order of 140 kWh per tonne, which correspond to longer residence times within the microwave field or a higher applied microwave power.

The results shown in Figs. 4 and 6 indicate that both higher applied powers and longer residence times in the microwave field are beneficial to the remediation process. The results obtained do not allow the effects of heating rate to be established, since tests would need to be evaluated using different power densities at equivalent energy inputs. For example, it is not possible to determine whether a high power and short residence time is better than a low power and a long residence time. The effect of power density can be evaluated by comparing the results of the continuous testwork with those obtained from batch microwave experiments in single mode and multimode cavities.

3.6. Comparison with batch processing

Batch tests were carried out using the single mode and multimode apparatus described in previous work [5,6]. The key

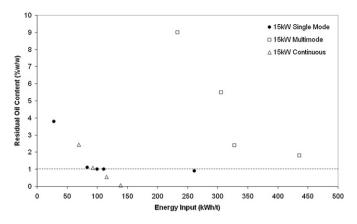


Fig. 9. Variation of residual oil content with energy input for three processing scenarios utilising 15 kW of microwave power.

Table 2

Power densities in multimode, single mode and continuous microwave applicators.

Processing strategy	Multimode	Single mode	Continuous
Applied power (kW)	15	15	15
Throughput (kg/h)	-	-	160
Sample volume (cm ³)	100	100	-
Mean power density (W/m ³)	$4 imes 10^6$	7.5×10^7	1.4×10^8

differences are that the single- and multimode microwave systems are batch, compared with the novel continuous system described in this paper. Multimode cavities, like domestic microwave ovens, have a relatively large volume and low mean power density. Single mode cavities yield a very high peak power density in a small volume, but with a very large power density distribution. In a single mode cavity the power density varies from a peak at the centre to zero at the walls, and it is for this reason that single mode cavities are not used in continuous treatment systems. In all cases the applied power was 15 kW, and the residence time or treatment time was varied. The results are shown in Fig. 9, where the three cavities are compared based on the microwave energy used in each case.

All three microwave cavities are able to induce significant levels of remediation of the contaminated drill cuttings. The cavity which supports the lowest power density is the multimode cavity, and it can be seen from Fig. 9 that the energy requirements are of the order of 450 kWh per tonne to reduce the oil content to 2%. The tests in the multimode cavity showed that the 1% discharge threshold is difficult to achieve, and our previous work also supports this observation [5]. A summary of the experimental conditions used is shown in Table 2, along with the power densities which are supported in each of the three different cavities.

The results obtained with the single mode cavity showed that the 1% discharge threshold could be achieved, and that the remediation process occurs using lower energy inputs than were evident in the multimode cavity. This is because the single mode cavity supports a power density which is an order of magnitude higher than that in the multimode cavity (see Table 2) and this leads to an increased heating rate according to Eq. (1). The microwave power is dissipated within the water phase, causing superheating of the water and rapid conversion into steam. At lower power densities the heating rate of water is proportionately lower, meaning that more heat is lost to the surrounding oil and rock before the water is converted to steam. The energy requirements are therefore higher at low power densities as more heat is lost to the surroundings, leading to a less efficient process. Conversely at high power densities the water is converted to steam very rapidly, with less heat transfer to the surroundings and therefore the process is more efficient.

The average power density supported by the continuous system is 1.4×10^8 W/m³, which is approximately double that supported by the single mode cavity. It can be seen in Fig. 9 that the effect of energy input on oil removal with the two cavities is roughly comparable up to the 1% threshold, but the continuous cavity allows much lower levels of residual oil to be achieved. The single mode treatment is a batch process, and the dielectric constant and loss factor of the drill cuttings decrease with time as water is lost. At low oil levels there is a correspondingly small amount of water within the sample, and its dielectric loss factor is low. This means that it is difficult to concentrate sufficient microwave energy into the sample, and results in a high reflected power towards the end of the batch test. This was observed in practice, with over 80% of the applied power reflected at the end of the single mode tests, which corresponds to less than 3 kW of power absorbed. It is thought that the increasing reflected power during the tests accounts for the inability of the single mode treatment to remediate the drill cuttings to levels significantly below 1% oil.

4. Conclusions

The continuous treatment of contaminated drill cuttings has been demonstrated using a microwave cavity based on a transverse E-field applicator. The removal of the contaminant oil is dependent on the applied microwave power, and the residence time within the microwave cavity. The residual oil levels can be reduced to below the 1% environmental discharge threshold, and under continuous processing conditions can be further reduced to 0.1%. It is shown that the continuous system is capable of higher levels of remediation than equivalent batch processes, and at lower energy inputs.

Acknowledgements

BP, BG Technologies and DEFRA are acknowledged for funding this work programme.

References

- G.C. Okpokwasili, C. Nnubia, Effects of oil spill dispersants and drilling fluids on substrate specificity of marine bacteria, Waste Management 15 (7) (1995) 515–520.
- [2] Oslo and Paris Commission, OSPAR 2000/3, 2000.
- [3] US Environment Protection Agency, NPDES Offshore General Order, February 16th 2002.
- [4] Landfill of waste, Council Directive 99/31/EC, 1999.
- [5] H. Shang, C.E. Snape, S.W. Kingman, J.P. Robinson, Microwave treatment of oil-contaminated North Sea drill cuttings in a high power multimode cavity, Separation & Purification Technology 49 (2006) 84–90.
- [6] H. Shang, C.E. Snape, S.W. Kingman, J.P. Robinson, Treatment of oil contaminated drill cuttings by microwave heating in a high power single mode cavity, Industrial & Engineering Chemistry Research 44 (2005) 6837–6844.
- [7] J.P. Robinson, S.W. Kingman, A. Onobrakpeya, Microwave enhanced stripping of oil contaminated drill cuttings, Journal of Environmental Management 88 (2008) 211–218.
- [8] H. Shang, S.W. Kingman, C.E. Snape, J.P. Robinson, Reactor effects on microwave decontamination of oily wastes in a multimode cavity, Industrial & Engineering Chemistry Research 46 (2007) 4811–4818.
- [9] J.P. Robinson, C.E. Snape, S.W. Kingman, H. Shang, Thermal desorption and pyrolysis of oil contaminated drill cuttings by microwave heating, Journal of Analytical and Applied Pyrolysis 81 (2008) 27–32.
- [10] R.B. Keey, Drying of loose and particulate materials, Taylor & Francis, Washington, 1992.
- [11] R. Meredith, Engineers Handbook of Industrial Microwave Heating, Institute of Electrical Engineers, London, 1998.