

Binner, Eleanor and Robinson, John P. and Kingman, S.W. and Lester, Edward and Azzopardi, Barry J. and Dimitrakis, Georgios and Briggs, John (2013) Separation of oil/water emulsions in continuous flow using microwave heating. Energy and Fuels, 27 (6). pp. 3173-3178. ISSN 1520-5029

Access from the University of Nottingham repository:

http://eprints.nottingham.ac.uk/31948/1/130514%20Separation%20of%20oil%20water %20emulsions%20in%20continuous%20flow%20using%20microwave%20heating_for %20archiving.pdf

Copyright and reuse:

The Nottingham ePrints service makes this work by researchers of the University of Nottingham available open access under the following conditions.

- Copyright and all moral rights to the version of the paper presented here belong to the individual author(s) and/or other copyright owners.
- To the extent reasonable and practicable the material made available in Nottingham ePrints has been checked for eligibility before being made available.
- Copies of full items can be used for personal research or study, educational, or notfor-profit purposes without prior permission or charge provided that the authors, title and full bibliographic details are credited, a hyperlink and/or URL is given for the original metadata page and the content is not changed in any way.
- · Quotations or similar reproductions must be sufficiently acknowledged.

Please see our full end user licence at: http://eprints.nottingham.ac.uk/end_user_agreement.pdf

A note on versions:

The version presented here may differ from the published version or from the version of record. If you wish to cite this item you are advised to consult the publisher's version. Please see the repository url above for details on accessing the published version and note that access may require a subscription.

For more information, please contact eprints@nottingham.ac.uk

Separation of oil/water emulsions in continuous flow using microwave heating

Eleanor R. Binner, John P. Robinson*, Sam W. Kingman, Ed H. Lester, Barry J. Azzopardi, Georgios Dimitrakis, John Briggs

Process & Environmental Research Division, Faculty of Engineering, University of Nottingham

Abstract

This work studies a continuous flow microwave system to enhance gravity settling of water-inoil emulsions. Settling times were found to be dependent upon the applied power, flowrate and energy input. Power and energy input are linked to liquid flowrate within the flow system used in this study, so a key objective of this work was to understand the effect of turbulence on the heating and separation of the flowing emulsion. At high flowrates (9 - 12 L/min) it was found that turbulence dominates, with settling times largely independent of energy input. At lower flowrates (1 - 6 L/min), when turbulence was decreased, it was found that the settling time decreased as the power density was increased. Settling times have the potential to be less than half that of untreated emulsions, and can be reduced further if turbulence can be minimised between the microwave heating zone and the settling zone in the process equipment.

Introduction

The demulsification of stable oil/water emulsions is a key process in the removal of water from crude oil. These emulsions are formed during the production of oil downstream from the well. Water is present with the oil, particularly when water injection is used to enhance the production rate. On leaving the well the mixture, at high pressure, is let down through a choke valve so that it can be further processed at moderate pressures. The pressure reduction valve creates significant shear within the fluid and it is this pressure drop that causes the emulsions to form. The emulsions can contain more than 20% water [1], which must be separated before further downstream transport and processing can commence. As the world's oil production comes from increasingly heavy oil reserves, these emulsions are becoming more stable and therefore costly to separate [2].

Demulsification is based on the density difference between oil and water. Droplets of oil rise through water to the top whereas droplets of water fall through oil to the bottom. The rise and fall of droplets in a continuous medium can be quantified according to Stokes' Law, which is given by Equation 1:

$$u_{s} = \frac{D_{1}^{2}g(\rho_{1} - \rho_{2})}{18\mu_{2}}$$
(1)

where u_s is the terminal settling velocity of the droplet (m/s), D_1 the droplet diameter (m), g the acceleration due to gravity(m/s²), ρ_1 the droplet density (kg/m³), ρ_2 the density of the continuous medium (kg/m³), and μ_2 the viscosity of the continuous medium (kg/m.s). The separation of the two phases is governed by the settling velocity, u_s . If u_s is small then a large residence time is required for the separation to occur, hence separation vessels need to be very large. Conversely if u_s is large then the separation occurs in a short time and separation equipment can be relatively small.

Equation 1 shows that u_s can be manipulated by changing the properties of the emulsion system. The densities of liquids are largely independent of changes in temperature, varying by only 6% over a 100°C range for water [3], hence the two main governing parameters are the viscosity and droplet diameter. Increased settling velocities will result from an increase in droplet diameter and a reduction in the viscosity of the continuous medium. The viscosity of a substance can be reduced by increasing its temperature, typically in an exponential fashion. Droplet size can be increased by a process known as coalescence, where smaller droplets join together to produce larger ones. The probability of coalescence is a function of the droplet size distribution, number of droplets and dispersion density and the surface properties of the interface between the droplet and the continuous medium, namely the interfacial tension.

Improving the separation of water-in-oil emulsions – enhancing settling velocity

Gravity separation is a slow process requiring large residence times, and there are several ways to speed up the process. Heat treatment reduces both the interfacial tension of the oil/water and viscosity of the oil [4]. Centrifugal separation effectively increases g and hence u_s in Equation 1. Coalescence can be promoted by using chemical additives, which lower interfacial tension, or by applying a physical force to move the droplets together, which can be achieved using ultrasound or an electrostatic force [5]. There are several disadvantages with these methods:

- 1. Heating the whole process stream requires a significant amount of energy
- 2. Centrifugation has high operating and maintenance costs
- 3. Chemical demulsifiers can add a significant cost and environmental impact
- 4. Electrostatic Coalescers add significant cost, and are often limited to low water concentrations

One technique that has received attention from previous researchers is microwave heating. There are two main advantages of microwave heating over conventional heating of emulsions:

(i) The water phase of the emulsion can be heated selectively, thus reducing the overall energy requirements

(ii) Microwave heating is volumetric, meaning that the entire dispersed water phase is heated instantaneously, without the limitations of conductive and convective heat transfer.

The extent to which a material will undergo microwave heating is quantified according to Equation 2:

$$\mathbf{P}_{\rm d} = 2\pi \mathbf{f} \varepsilon_0 \varepsilon'' |\mathbf{E}|^2 \tag{2}$$

where P_d is the power density (W/m³), i.e. the power dissipated within a given volume of material, *f* the frequency of the applied electromagnetic wave (Hz), ε_0 is the permittivity of free space (8.85×10⁻¹² F/m), ε'' is the dielectric loss factor and *E* the electric field strength (V/m). The dielectric loss factor is a physical property of the material to be heated, and can be used to quantify the degree to which a material dissipates stored microwave energy as heat; materials with a low (<0.05) value of ε'' can be considered to be effectively microwave transparent since the power dissipated will be very low according to Equation 2. Substances with a higher loss factor are good microwave transparent compared with water, with a dielectric loss of two orders of magnitude smaller [6]. It has been shown that, with the exception of emulsions with very small water droplets (<256 nm), microwave heating will result in selective heating of the water when an oil water emulsion is exposed to a microwave field [5].

Many workers have shown that microwave heating is able to enhance the separation of oil/water emulsions. Fang et al. [7] reported that microwave treatment of 30/70 and 50/50 water-in-motor

4

oil emulsions enhanced gravity settling, and that increasing treatment time decreased settling time up to the point where heating caused turbulence. Fang et al. [8] and Xia et al. [9] also found that gravity settling was enhanced with microwave treatment, using zeta potential to characterise their emulsions. Fortuny et al. [10] investigated the effects of salinity, pH and water content on the microwave demulsification of crude oil emulsions. They concluded that an increase in microwave energy input enhanced demulsification. They also reported samples that exhibited both high pH and salt content were too stable for reasonable demulsification results to be achieved. Lemos et al. [2] extended the work to look at the addition of ionic liquids and also compared conventional heating with microwave heating. They found that demulsification was improved by microwave heating when compared with conventional heating, and conjectured that the ability of the ionic liquids to absorb microwaves and position themselves at interfaces may contribute toward driving the microwave heating toward the interfacial film around the droplets. Dielectric properties of the ionic liquids, which would be required to assess the validity of this conclusion, were not reported. Kovaleva et al. [11] compared the effects of radio frequency, microwave heating and conventional heating on demulsification of water in oil emulsions. The microwave and radio frequency experiments they conducted yielded better results than conventional heating, and this was attributed to volumetric heating. A mathematical model assuming spherical droplets was developed to predict the temperature profiles in the emulsion during electromagnetic heating. In the case of microwave heating, the model suggested that the distribution of the thermal field inside the water droplet was spatially uneven, and the authors suggested that this may cause local ruptures of the armour envelope surrounding the droplet, leading to a multitude of small droplets being formed from the single large droplet. These studies [2, 8-11] demonstrate the ability of microwave treatment to enhance demulsification. However,

they used commercially available microwave systems that operate at constant temperature, and therefore the incident (and absorbed) power varied between runs, making assessment of the effect of different parameters difficult. Furthermore, the temperature in these systems is measured at the sample surface and so may not be representative of the bulk temperature. Tan et al. [12] report the use of chemical demulsifiers along with batch microwave heating. They found that that the use of emulsifiers along with microwave heating significantly improved separation over microwave treatment alone. They also observed that the demulsification efficiency decreased with decreasing water content in the emulsion, and this was attributed to the decrease in microwave absorbing material in the microwave cavity. This problem could be overcome however in a system with an optimised microwave cavity, which would enable a high degree of absorbed power. Guzmán-Lucero et al. [13] found that ultra-heavy oil emulsions could not be 'broken' by microwave treatment alone, but that microwaves could be used in conjunction with a chemical demulsifier to aid the separation process.

While these studies have demonstrated the ability of microwaves to enhance gravity separation of oil/water emulsions, they were all batch experiments. Before larger scale microwave treatment systems are developed it is necessary to evaluate a continuous microwave system to assess the impact of processing conditions on separation of the emulsion. Continuous flow can results in liquid turbulence, which adds a further variable to the process and makes the separation of oil and water more difficult. Nour et al. [14] report the continuous microwave heating of water-in-oil emulsions. As expected, their results show that longer residence times in a multimode cavity lead to shorter settling times. However, the absorbed power was not measured and pumping rates not reported, therefore it is not possible to carry out an energy balance or interpret the degree of turbulence within their system. The objectives of this study are to understand how microwave power, liquid flowrate and energy input effect the separation of oil/water emulsions of different composition, and to understand the opportunities and limitations for an industrial process.

Methodology

The crude oil used in this study was Azeri crude supplied by BP. The assay and physical properties can be downloaded from the BP website [15].

Emulsion formation

A known amount of distilled water (10 or 20%) was added to the Azeri crude resulting in an emulsion with a total volume of 3 L. Fluorescein was also added (2ppm in water) to improve the tracking of the separation interface during data analysis. After this procedure, each mixture was homogenised for 2 minutes using a double stirrer at 600 rpm.

A high standard of emulsion reproducibility was confirmed by utilizing optical microscopy techniques. The procedure involved capturing images of newly homogenized emulsions using a digital microscope, and analysing the size of water droplets in the crude phase using a bespoke image analysis program based on a Kontron KS400 version 3.1 System. An example of the emulsion droplet size distribution and corresponding image is shown in Figure 1.



Figure 1. Size distribution of water droplets formed during emulsification. Inset image shows the water droplets after thresholding from the background oil using image analysis.

Microwave Heating & Interface tracking

Once prepared the emulsions were immediately pumped through the continuous flow microwave treatment system into a 600 mL settling beaker, as shown in Figure 1. The emulsion was pumped through a 20 mm diameter microwave transparent pipe at flow rates from 1 - 12 L/min. The microwave system featured a TE_{01n} single mode cavity and a Sairem 0-15 kW microwave generator operating at 2.45GHz. An automatic E-H tuner was used to match the impedance of the generator to the transmission line in order to minimise the reflected power. The absorbed power was recorded throughout the experiment through logging the data output port of the generator using a specifically created program based on Labview. Once in the settling beaker, the emulsion was filmed using a Sony HDR-FX7 camcorder. An image analysis method was used to

examine the camcorder video, still by still to track the oil-water interface with time. The reproducibility of the demulsification tests using microwave heating was evaluated through the repetition of tests at fixed conditions. One sample within each set of the separation experiments was repeated at least twice. The maximum fluctuation in the separation time results, for a given set of experimental conditions, was found to be <10%.



Figure 2 – Schematic of experimental set-up

Results and Discussion

The settling behaviour of the emulsions with time is shown in Figure 3 - Figure 5.



Figure 3 - Interface height plotted against time for 80/20 oil/water emulsions treated at 6 L/min

Figure 3 shows that the emulsions subjected to microwave heating separate much faster than untreated emulsions as the interface height increases more sharply over the time period studied. For example, after 6000 seconds the emulsion treated at 12 kW has almost reached its maximum degree of separation (fractional interface height = 0.2), whereas the interface of the untreated emulsion attains a fractional height of just over 0.1, meaning that the upper oil phase still contains 50% of the total water in the emulsion. From the data shown in Figure 3, obtained at a flow rate of 6 L/min, it is difficult to distinguish between the separation behaviour of the emulsions treated at different microwave powers. However, differences in settling become more apparent at higher flow rates as shown in Figure 4 and Figure 5.



Figure 4 - Interface height plotted against time for 80/20 oil/water emulsions treated at 9 L/min

At 9 L/min there is still a clear difference between the separation of the untreated emulsion and the emulsions treated at 3-12 kW, as shown in Figure 4. Although the separation times of the treated emulsions are difficult to distinguish, it is apparent that the interface height has not progressed at the same rate as for the lower flow rate tests shown in Figure 3. For example, at 7000 seconds all but one of the treated emulsions in Figure 3 have reached the maximum interface height, whereas in Figure 4 the interface of the treated emulsions remains well below the maximum height. When the flow rate is increased to 12 L/min, as shown in Figure 5, the interfaces are lower still and it is possible to distinguish between the applied power levels of the treated emulsions. The emulsion that separates fastest was heated at 12kW, after which the separation times increase with decreasing power levels. It is also apparent in Figure 5 that the separation of emulsions flowing at 12 L/min takes longer than at 6 L/min and 9 L/min.



Figure 5 - Interface height plotted against time for 80/20 oil/water emulsions treated at 12 L/min

There are two competing physical effects that can explain the sequences of data shown in Figure 3 - Figure 5, which are net energy input and turbulence.

1. Energy Input

The power absorbed by the emulsion in kW (P_a) and the net energy input into the system in kJ/L (ΔQ) are given in Equations 3 and 4 respectively, where P_i is the incident power in kW, P_r is the reflected power in kW and q is the emulsion flow rate in L/min.

$$P_a = P_i - P_r \tag{3}$$

$$\Delta Q = \frac{P_a x 60}{q} \tag{4}$$

An autotuner was used in these experiments, which minimized reflected power (typical values of x were recorded), and therefore P_a . approached P_i in these experiments. It is clear from Equations 3 and 4 that increasing P_i and decreasing q increase the net energy input into the

system. Since only the water phase of the emulsion absorbs microwave energy to any significant degree, the temperature of the water will be highest at the lower flow rates or higher powers. The higher temperature of the water leads to two physical effects that enhance the separation of the emulsions according to Equation 1; (i) a reduction in droplet surface tension, which promotes coalescence to form larger droplets, and (ii) a reduction in the viscosity of the oil at the droplet surface due to heat transfer. The larger droplet diameter, D₁, and the lower oil viscosity, μ_2 , increases the droplet settling velocity and therefore reduces the time taken for the emulsion to separate.

The image analysis technique was used to identify the total separation time, i.e. the time at which the interface of the emulsion reached its maximum height. This data is shown in Figure 6.



Figure 6 – Effect of power and flow rate on the separation of 80/20 Azeri crude emulsions. Dashed lines are contours of constant energy input

The data in Figure 6 show the general trend that increasing the applied power leads to a decrease in the separation time at all three flow rates studied. From Equation 4, the net energy input (ΔQ) can be calculated from absorbed power (P_a) and flow rate (q). In order to attain the same net energy input at higher powers, the flow rate must be decreased. Several of the experiments were carried out at an equivalent net energy input as shown by the contours in Figure 6. This data indicates that increasing the flow rate, at the same energy input, leads to an increase in the separation time of the emulsions. This behaviour can be attributed to turbulence within the system.

2. Turbulence

The flow pattern within the heating zone of the single mode cavity is laminar, as the Reynolds Number is within the range of 1.0-5.0. However, upon exiting the microwave cavity the treated emulsion flows into a separate separation vessel, and the expansion from flow in the 20mm diameter pipe to the vessel causes turbulence. The turbulent flow causes two fundamental effects; (i) extra shear forces are created within the liquid which could cause further emulsification, and (ii) heat transfer from the water droplets into the surrounding oil phase is faster, meaning that the viscosity in the vicinity of the water droplets approaches that of the bulk oil. The separation of untreated emulsions at the different flow rates is shown in Figure 7.



Figure 7 – Effect of flow rate on the separation of untreated 80/20 emulsions

It is clear from Figure 7 that increasing the flow rate causes an increase in the separation time for untreated emulsions, as the interface height takes longer to increase. Since there is no heating taking place the data must be caused by the extra turbulence associated with the higher flow rate as the emulsion expands from the pipe to the vessel.

A separate series of experiments was carried out so that the competing effects of energy input and flow rate could be decoupled. Much lower flow rates were used in order to minimise the effects of turbulence, and a matrix of experiments was designed to evaluate the effect of increasing power at the same energy input. The absorbed power (P_a), flow rate (q) and emulsion water content were varied in order to investigate the effect of net energy input (ΔQ , as defined in Equation 3) and Power density (P_d , as defined in Equation 2). The parameters used in these experiments are shown in Table 1.

Emulsion	Absorbed	Flow rate	Net Energy	Power Density
Water Content	Power (kW)	(L/min)	Input (kJ/L)	in Water Phase
(%)				$(x10^8 \text{W/m}^3)$
20	0.6	1.0	36	2.4
20	0.9	1.5	36	3.6
20	1.05	1.75	36	4.2
10	0.3	1.0	18	2.4
10	0.45	1.5	18	3.8
10	0.6	2	18	4.8

Table 1 – Parameters studied in lower flow rate experiments

The data from the lower flow rate experiments are divided according to the energy absorbed by the emulsion. Figure 8 shows the interface height data for 36 kJ/L and Figure 9 shows the corresponding data at 18 kJ/L.



Figure 8 - Interface height plotted against time for 80/20 oil/water emulsions treated with a total energy input of 36 kJ/L



Figure 9 - Interface height plotted against time for 90/10 oil/water emulsions treated with a total energy input of 18 kJ/L

The data shown in Figure 8 and Figure 9 show an opposite trend to the experiments carried out at higher flow rates. In this case the separation time decreases with increasing power (and flow rate) and constant energy input. Given that the turbulence is minimised at these lower flow rates, the explanation for this data lies in the power density within the water phase, and the subsequent role of heat transfer. The power density in each case is shown in Table 1, and it can be related to the dielectric heating rate ($\Delta T/\Delta t$) according to Equation 5:

$$\frac{\Delta T}{\Delta t} = \frac{Pd}{\rho C_{p}}$$
(5)

where P_d is the power density and ρ and C_p are the density and heat capacity of the heated phase respectively. High power densities lead to corresponding high heating rates from Equation 5. The net temperature attained by a water droplet suspended within an oil phase is a function of the rate of energy input (i.e. that due to power density) and the rate of energy loss due to heat transfer. To maximise the effect of microwave heating for emulsion separation it is necessary to maximise the temperature of the water droplets as this leads to:

- (i) A lower droplet surface tension, leading to a higher probability of coalescence
- (ii) A lower localised oil viscosity at the droplet interface.

The results presented in this paper show that increasing power density (and hence dielectric heating rate as given in Equation 5) improves separation. This leads to the conclusion that if the droplets are heated slowly (at low power density), the rate at which energy dissipates from the droplet to the surrounding oil becomes significant relative to the rate of energy input and therefore the temperature attained by the droplet is low. Conversely, at high power densities the rate of energy input can be much higher than the rate of energy dissipation, meaning that much higher droplet temperatures are obtained. It is therefore the selective heating ability of microwaves that lead to improved separation efficiencies when a higher power density is used at the same energy input. Although the bulk emulsion temperature will be the same at a constant energy input, the temperature gradients will be significantly different during and immediately after microwave heating, and it is this phenomenon which leads to the experimentally observed power-density dependence.

Conclusions

This work has confirmed that microwave enhanced gravity settling, which has been well established in laboratory scale batch tests, can be extended to a semi-continuous system in which emulsions could flow continuously through a microwave treatment system into a series of settling vessels. Settling times have the potential to be less than half that of untreated emulsions. The results have established that this is a power density driven process; the higher temperature gradients that can be achieved at higher powers with the same energy inputs promote coalescence and settling velocity. Optimal energy requirements for the system have not been established, but would be less than the heat required to heat the water to 100°C (67 kJ/L for a 80/20 emulsion).

The major limitation of this process is the potential for turbulence at an industrial scale. The minimisation of Reynolds Number must be paramount in the design of any larger scale systems.

AUTHOR INFORMATION

Corresponding Author

*John P. Robinson, Process and Environmental Research Division, Faculty of Engineering, University of Nottingham, NG7 2RD, UK; Tel: +44 115 951 4092; e-mail:

john.p.robinson@nottingham.ac.uk

Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

ACKNOWLEDGMENT

The authors wish to acknowledge BP for funding this work.

REFERENCES

1. Eow, J. S.; Ghadiri, M., Electrostatic enhancement of coalescence of water droplets in oil: A review of the technology. *Chemical Engineering Journal* **2002**, *85* (2-3), 357-368.

2. Lemos, R. C. B.; Da Silva, E. B.; Dos Santos, A.; Guimarães, R. C. L.; Ferreira, B. M. S.; Guarnieri, R. A.; Dariva, C.; Franceschi, E.; Santos, A. F.; Fortuny, M., Demulsification of water-in-crude oil emulsions using ionic liquids and microwave irradiation. *Energy and Fuels* **2010**, *24* (8), 4439-4444.

3. Perry, R. H.; Green, D. W., *Perry's Chemical Engineers Handbook*. Seventh Edition ed.; McGraw Hill: London, 1998.

4. Arnold, K.; Stewart, M., *Surface Production Operations - Design of Oil Handling Systems and Facilities.* 3rd Edition ed.; Elsevier: 1998; Vol. Volume 1.

5. Holtze, C.; Sivaramakrishnan, R.; Antonietti, M.; Tsuwi, J.; Kremer, F.; Kramer, K. D., The microwave absorption of emulsions containing aqueous micro- and nanodroplets: A means to optimize microwave heating. *Journal of Colloid and Interface Science* **2006**, *302* (2), 651-657.

6. Erle, U.; Regier, M.; Persch, C.; Schubert, H., Dielectric properties of emulsions and suspensions: Mixture equations and measurement comparisons. *Journal of Microwave Power* and Electromagnetic Energy **2000**, *35* (3), 185-190.

7. Fang, C. S.; Lai, P. M. C., Microwave heating and separation of water-in-oil emulsions. *Journal of Microwave Power and Electromagnetic Energy* **1995**, *30* (1), 46-57.

8. Fang, C. S.; Lai, P. M. C.; Chang, B. K. L.; Klaila, W. J., Oil recovery and waste reduction by microwave radiation. *Environmental Progress* **1989**, *8* (4), 235-238.

9. (a) Xia, L.; Lu, S.; Cao, G., Demulsification of Emulsions Exploited by Enhanced Oil Recovery System. *Separation Science and Technology* **2003**, *38* (16), 4079-4094; (b) Xia, L. X.; Lu, S. W.; Cao, G. Y., Salt-assisted microwave demulsification. *Chemical Engineering Communications* **2004**, *191* (8), 1053-1063.

10. Fortuny, M.; Oliveira, C. B. Z.; Melo, R. L. F. V.; Nele, M.; Coutinho, R. C. C.; Santos, A. F., Effect of salinity, temperature, water content, and pH on the microwave demulsification of crude oil emulsions. *Energy and Fuels* **2007**, *21* (3), 1358-1364.

11. Kovaleva, L. A.; Minnigalimov, R. Z.; Zinnatullin, R. R., Destruction of water-in-oil emulsions in radio-frequency and microwave electromagnetic fields. *Energy and Fuels* **2011**, *25* (8), 3731-3738.

12. Tan, W.; Yang, X. G.; Tan, X. F., Study on demulsification of crude oil emulsions by microwave chemical method. *Separation Science and Technology* **2007**, *42* (6), 1367-1377.

13. Guzmán-Lucero, D.; Flores, P.; Rojo, T.; Martínez-Palou, R., Ionic liquids as demulsifiers of water-in-crude oil emulsions: Study of the microwave effect. *Energy and Fuels* **2010**, *24* (6), 3610-3615.

14. Nour, A. H.; Yunus, R. M., A continuous microwave heating of water-in-oil emulsions: An experimental study. *Journal of Applied Sciences* **2006**, *6* (8), 1868-1872.

15.Azeri(Ceyhan)Assay.http://www.bp.com/extendedsectiongenericarticle.do?categoryId=9010999&contentId=7020188.