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**THE EFFECT OF VISCOSITY RATIO ON THE HYDRODYNAMICS OF SEPARATION
FROM AN OIL-COATED PARTICLE**

Sasan Mehrabian

Department of Mechanical & Industrial
Engineering
University of Toronto

Nima Abbaspour

Department of Mechanical & Industrial
Engineering
University of Toronto

Markus Bussmann

Department of Mechanical & Industrial
Engineering
University of Toronto

Edgar Acosta

Department of Chemical Engineering & Applied
Chemistry
University of Toronto

ABSTRACT

Separating oil from solid particles is of great importance in many industrial processes including the extraction of bitumen from oil sands, and the remediation of oil spills. The usual approach is to separate the oil from the solid by introducing another liquid (e.g. water). Separation is often assisted by fluid mixing, and chemical addition. Yet while oil-water-particle separation has been well studied from a chemical standpoint, little research has taken into account the effect of hydrodynamics on separation. In this work, the separation of oil from a single oil-coated spherical particle falling through an aqueous solution was evaluated as a function of viscosity ratio. Solvents were used to modify the viscosity of the oil. The experiments were recorded using a high-speed camera and post-processed using the MATLAB image-processing toolbox. A CFD model has also been developed to study this phenomenon.

The results indicate that when viscous forces are strong enough, the oil film deforms, flows to the back of the sphere, and forms a tail that eventually breaks up into a series of droplets due to a capillary wave instability. When the viscosity ratio is small (i.e. the oil is less viscous than the solution), a thin tail forms quickly, the growth rate of the instability is high, and hence the tail breaks very quickly into smaller droplets. When the viscosity ratio is high (i.e. the oil is more viscous), more time is required for the deformation/separation to initiate, and the tail is thicker and breaks more slowly into larger droplets. It was observed that when the viscosity ratio is close to 1, the rate of separation is increased and maximum separation is achieved.

INTRODUCTION

There are many applications that involve the separation of oil from solid particles in a viscous fluid undergoing shear, including bitumen extraction from oil sands [1-3], the remediation of oil spills [4, 5], and remediation of oil-based drill cuttings [6]. Such oil/particle separation depends on the processing method, and on the physicochemical properties of the system, such as the viscosities and interfacial tension. Oil/particle separation has been studied from thermodynamic and chemical standpoints [7-9], but few studies have considered the effect of hydrodynamics in a fundamental way. The objective of this study is to examine the separation of oil from a solid particle by examining an oil-coated sphere in a well-defined shear field.

When an oil-coated particle is subjected to a uniform aqueous flow, viscous, gravitational and surface tension forces may all play an important role. Viscous and gravitational forces will shear and deform the oil that is coating the particle. Surface tension, on the other hand, attempts to maintain the oil on the particle and stabilize its shape. In this study, the separation of oil from an oil-coated sphere falling through an aqueous solution is examined experimentally and numerically as a function of the viscosity ratio. The Reynolds number $Re = 2\rho_c UR_0/\mu_c$ based on the terminal velocity and diameter of the oil-coated sphere is small, so that the effects of particle inertia are negligible. Fig. 1 illustrates the configuration. ρ_c and μ_c are the density and the viscosity of the aqueous solution respectively, ρ_d and μ_d are the density and the viscosity of the oil respectively, R_0 is the sphere radius, h is the initially oil film

thickness, and U is the velocity of the falling oil-coated particle.

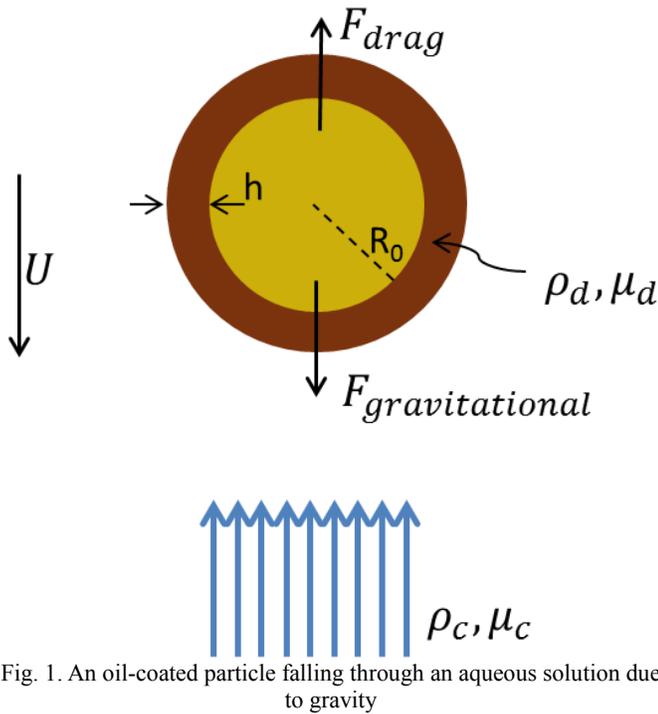


Fig. 1. An oil-coated particle falling through an aqueous solution due to gravity

An analysis of such a solid/liquid/liquid system can be guided by the considerable literature on the deformation and breakup of immiscible liquid droplets in an unbounded media subject to shear [10-15]. At low Reynolds numbers, viscous forces contribute to the deformation and breakup of a droplet, while surface tension tends to oppose deformation. The ratio of these forces is expressed as a Capillary number Ca . If Ca exceeds a critical Capillary number Ca_{crit} that is a function of the viscosity ratio $p = \mu_d/\mu_c$, the droplet will breakup. Grace [13] showed that for $0.1 < p < 1$, Ca_{crit} is a minimum, meaning that in that range of p the shear rate required to deform and breakup a droplet is a minimum.

To the best of our knowledge, the work of Smith and Van de Ven [16] is the only experimental work that has examined the deformation/separation of an oil layer coating a single spherical particle. They investigated the separation due to gravity of an axisymmetric pendant drop attached to a fixed solid sphere within an aqueous solution. Johnson [17] and Choudhuri and Padmavati [18] produced analytical models of the Stokes flow past a fluid-coated sphere, based on the assumption that the film remains of constant thickness. Johnson [17] used a perturbation scheme to solve the problem and showed that the drag on a uniformly coated sphere is lower than the drag on an uncoated sphere when $p < 1/4$. Choudhuri and Padmavati [18] confirmed Johnson's results and determined the optimal film thickness for which the drag is a minimum as a function of p .

The CFD study of Fan et al. [19] is most closely related to the work presented here. A model of an oil-coated particle

immersed in an axisymmetric laminar flow was developed. They defined a Ca_{crit} at which an equilibrium configuration is obtained. The Capillary number is defined as $Ca = \mu_c U/\sigma$, where σ is the interfacial tension. For Ca greater than Ca_{crit} , separation occurred. They also found that at smaller contact angles, oil was more difficult to remove.

In summary, there remains a lack of studies on the hydrodynamics of separation from an oil-coated particle. The work presented here complements the research of Fan et al. [19] and Smith and Van de Ven [16]. We present an experimental study of the separation of oil from an oil-coated sphere in free fall through an immiscible viscous fluid, and complementary numerical results. In Section 2, we introduce the experimental methodology, an image processing technique to quantify our results, and a brief overview on the numerical method. In section 3, results of the effect of viscosity ratio on oil separation are presented.

METHODOLOGY

Experimental Apparatus:

The experimental apparatus consists of a $10 \times 10 \times 45$ cm acrylic tank that contains an aqueous solution (Fig. 2). A square tank was chosen instead of a cylindrical one to minimize optical distortion. Images were recorded with a high speed CCD video recorder at up to 1000 frames per second. Lights were positioned on top of the tank to provide adequate illumination. An electromagnetic device was placed at the top of the tank, to serve as the release mechanism for the oil-coated solid sphere. Each experiment began with the sphere at rest and completely immersed in the aqueous solution. Steel spheres were used, with an average diameter of 4.26 mm (standard deviation of 0.0238 m), and a density of 8302.7 kg/m³ (standard deviation of 123 kg/m³).

Coker feed bitumen was used as the oil phase in this study. Bitumen was diluted with toluene at different concentrations to yield a wide range of viscosity. The bitumen (16.3 wt% saturates, 39.8 wt% aromatics, 28.5 wt% resins, and 14.7 wt% asphaltenes, according to a SARA analysis [28]) was donated by Syncrude Canada, and the toluene was purchased from Caledon Lab Canada. Table 1 lists the viscosity of the various oils used. The aqueous solution contained 2.5% of the common water-soluble polymer Carboxymethyl Cellulose (CMC), purchased from Sigma-Aldrich Canada. A CMC solution was used to control the viscosity ratios and to reduce the terminal velocity of the oil-coated sphere. The density of the CMC solution was measured to be about $\rho_c = 1$ gr/cm³.

The interfacial tension between all of the oils presented in Table 1 and the CMC solution is a very similar 27.63 ± 1.39 mN/m and was measured using the Axisymmetric Drop Shape Analysis (ADSA) method [28, 29]. This method is widely used due to its simplicity and accuracy. The samples were processed by "inverted sessile drop" method, where the interfacial tension was determined by a combination of surface tension and gravity effects. Surface tension tends to make the drop spherical whereas gravity tends to elongate the drop. The

results are comparable with the results of [20] where they used Heptol instead of toluene as their solvent.

A Carri-Med Rheometer with a 4-centimeter cone and plate geometry, with a cone angle of 2 degrees, was used to measure the viscosity of both the oil phase and the CMC solution. CMC solutions are non-Newtonian and follow the power law equation ($\tau = K\gamma^n$). A thorough investigation of the rheological behavior of CMC solutions is in [21, 22]. To characterize the viscosity ratio p , we require a single apparent viscosity for the CMC solution because of its shear thinning behavior. A comprehensive review of spheres falling through non-Newtonian fluids is presented in [24-26]. If the average shear rate on a sphere falling through a continuous medium is $U/2R_0$ [24], the apparent viscosity of a power-law fluid is given by the following equation:

$$\mu_c = K\left(\frac{U}{2R_0}\right)^{n-1}$$

where K is the consistency index and n is the power law index. K and n were determined to be $2.65 \text{ Pa}\cdot\text{s}^n$ and 0.7 , respectively. Consequently, $p = \mu_d/\mu_c$ depends on the terminal velocity of the oil-coated sphere, which can vary depending on the initial oil film thickness and the oil viscosity.

Table 1. Physical properties of the oils examined.

	Components	μ_d (Pa.s)	ρ_d (g/cm ³)
Oil 1	Bitumen	48.45	1
Oil 2	90.91 wt% Bitumen + 9.09 wt% Toluene	1.82	0.986
Oil 3	83.33 wt% Bitumen + 16.67 wt% Toluene	0.43	0.975
Oil 4	76.92 wt% Bitumen + 23.08 wt% Toluene	0.10	0.965

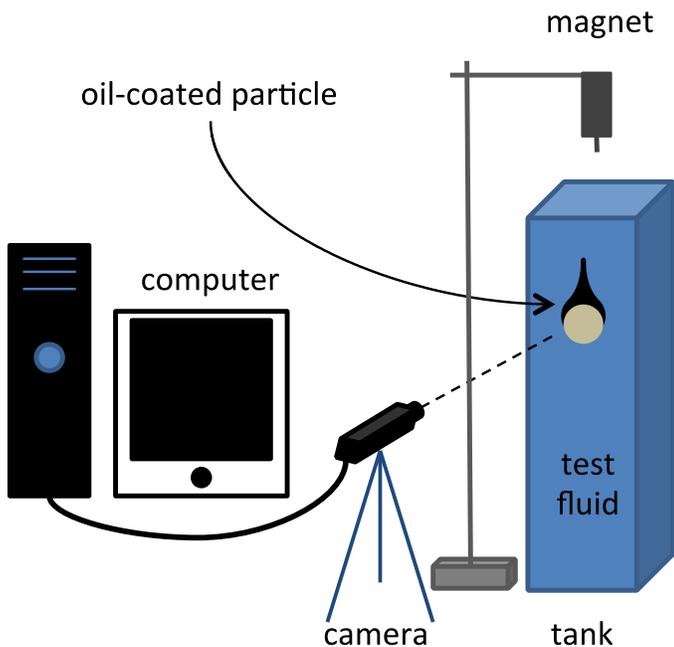


Figure 2. Schematic of the experimental apparatus.

Experimental Method:

Experiments were carried out to measure the amount of oil

separated during the fall. For each experiment, a sphere was initially placed on a weigh boat containing oil, all placed within a digital balance (accuracy of $\pm 10^{-4}g$) to obtain the mass of the sphere plus oil. The weigh boat was then agitated until a uniform layer of oil covered the sphere. The coated sphere was then picked up by an electromagnet and gently immersed into the aqueous solution at the top of the tank. The remaining oil in the weigh boat was weighed to determine the mass of oil on the sphere.

When the electromagnet is turned off, the oil-coated sphere begins to fall, shear stresses begin to act on the oil-aqueous solution interface, a tail (filament) forms behind the particle, which eventually breaks up into a series of mother (larger) and satellite (smaller) drops due to capillary wave instabilities (Fig. 3). The formation of the tail (separation of oil) depends on several factors, including the initial oil film thickness, viscosity ratio, and the interfacial tension. Because of the small density difference between the two liquid phases, the droplets remain still for a sufficient time to image them. High-resolution images (3456×2592) of the droplets were obtained every two centimeters along the tank for post-processing. Using an image processing technique in MATLAB, we calculated the radii and volume of each drop using a circle fitting function. The amount of oil separated was calculated by summing the volume of all droplets.

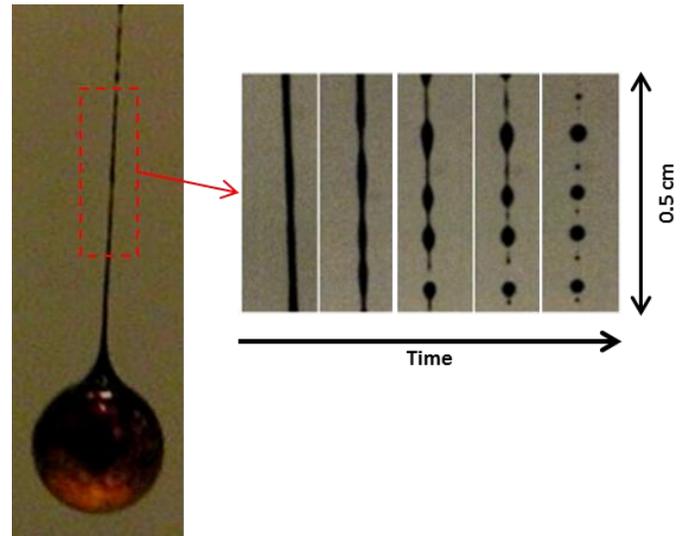


Figure 3. Breakup of the tail into droplets because of capillary wave instabilities.

Numerical Method

A Computational Fluid Dynamics (CFD) model has been developed to solve the Navier-Stokes equations for an oil-coated sphere falling through a continuous medium. The governing equations are:

$$\begin{aligned} \nabla \cdot \vec{V} &= 0 \\ \frac{\partial \rho \vec{V}}{\partial t} + \nabla \cdot (\rho \vec{V} \vec{V}) &= -\nabla P + \nabla \vec{\tau} + \rho \vec{g} + \vec{F}_{it} \end{aligned}$$

where ρ is density, \vec{V} is velocity, P is pressure, t is time, g is the gravitational acceleration, \vec{F}_{it} is the interfacial tension force, and $\vec{\tau}$ is the shear stress tensor, which for Newtonian fluids is:

$$\vec{\tau} = \mu(\nabla \cdot \vec{V} + (\nabla \cdot \vec{V})^T)$$

where μ is the dynamic viscosity. The volume-of-fluid (VOF) method is used to track the interface between the oil and the continuous medium [19, 23]. The interface between the water and oil is indicated by a scalar function f , defined as:

$$f = \begin{cases} 1 & \text{within oil} \\ 0 & \text{within water} \end{cases}$$

where f is advected with the flow according to:

$$\frac{\partial f}{\partial t} + (\nabla \cdot \vec{V})f = 0$$

The equations are solved in the spherical coordinate system at a specific Reynolds number, with the sphere fixed at the centre of the domain. The flow is assumed axisymmetric in the ϕ direction; thus the domain is two-dimensional (r, θ).

RESULTS AND DISCUSSION

Experimental Results

We present qualitative observations and quantitative results of the separation of oil from a falling sphere as a function of the viscosity ratio p . Fig. 4 illustrates typical experiments that show the deformation and separation characteristics of an oil-coated sphere with respect to time. The top sequence is of oil 4 with an initial film thickness of $\varepsilon = h/R_0 = 0.077$ and $p = 0.08$. Once the electromagnet is turned off and the oil-coated sphere starts to fall, the oil immediately begins to deform and move to the back of the sphere. Initially a stable and continuous tail forms, that becomes thinner and unstable as the sphere falls. The bottom sequence is of oil 1, $\varepsilon = 0.186$ and $p = 39.22$. This is a thicker and much more viscous film, and longer time is required for fluid shear to begin to deform and separate the oil. At about 2 seconds, the oil forms a cusp that eventually stretches and forms a thick tail. Although some separation was observed at this initial film thickness, no separation was observed when the initial film thickness was lower than $\varepsilon = 0.131$ at $p = 39.22$. This finding is in agreement with Fan et al. [19], who showed that increasing the initial oil film thickness could facilitate separation because a larger oil film increases the area subjected to shear. In addition, the rate at which the tail breaks up due to a capillary-wave instability is much lower for oil 1 than oil 4, and also larger droplets are observed after the breakup of the tail for the more viscous oil.

Fig. 5 illustrates the rate at which oil separates as a function of time for different p . The initial film thickness was constant at $\varepsilon = 0.077$. Depending on the film viscosity the velocity of the oil-coated sphere can change, hence; the Reynolds number and the Capillary number can be different for every system. The Reynolds number varies from 0.42 to 0.69 and the Capillary number varies from 2.31 to 3.04. In the case of pure bitumen (oil 1, $p = 39.22$), no oil separates. This implies that more time might be required for the shear stresses to overcome the interfacial tension forces, which might be seen in a larger tank. For oil 2 ($p = 1.44$), separation starts after a

delay (about 1.5 seconds). Once separation starts, the slope of the curve is steep and does not decrease until the sphere reaches the bottom of the tank. Here, a very thick tail forms, which results in large droplets. For oils 3 ($p = 0.36$) and 4 ($p = 0.08$), separation starts almost immediately and maximum delay does not exceed 0.3 seconds. As p decreases from 0.36 to 0.08, the thickness of the tail also decreases, which results in less oil separation.

It is interesting to point that similar fractions of oils 2 and 4 are separated by the time the spheres reach the bottom of the tank, despite having very different viscosity ratios. The mechanisms of separation are quite different, as will be discussed in the next section. Fig. 5 also shows that maximum separation is achieved with oil 3, where $0.1 < p < 1$, which corresponds to the results of Grace (Figure 19 of reference [13]), that show that the minimum shear required for droplet breakup also occurs when $0.1 < p < 1$.

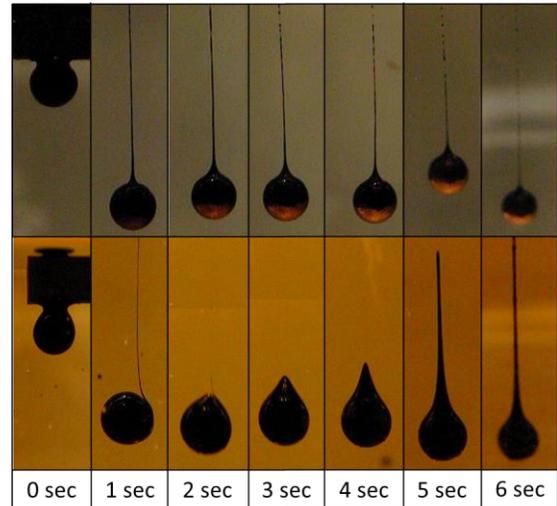


Figure 4. Typical deformation and separation of oil coating a sphere: (top) $\varepsilon = 0.077, p = 0.08$ (bottom) $\varepsilon = 0.186, p = 39.22$.

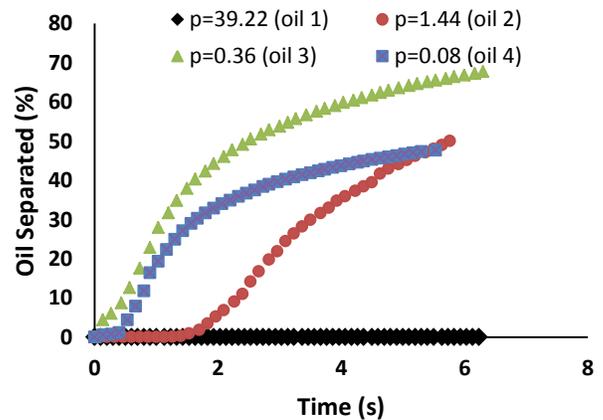


Figure 5. Percentage of oil separated for different values of p , $\varepsilon = 0.077$.

Numerical Results

A slightly different system was considered numerically. Most importantly, the results are for pure water (aqueous solution), and a particle with diameter and density of $100 \mu\text{m}$ and 2900 kg/m^3 , respectively. Different oil viscosities were examined that provided us with a wide range of viscosity ratios, from 0.01 to 10. At the terminal velocity the Reynolds number ranged from 0.9 to 1.5, and the Capillary number ranged from 0.6 to 1.2. Despite the fact that our numerical results are based on fluid properties that are different from our experimental results, they can be qualitatively compared. Hence, in this section we explain the physics taking place in Figure 5 and justify our experimental observations.

Fig. 6 illustrates the film evolution through separation for different viscosity ratios. The results of Fig. 6 are consistent with the experimental results of Fig. 5. It can be seen that at low viscosity ratios ($p = 0.01$), the tail forms very quickly but is also quite thin (last frame for $p = 0.01$); as the viscosity ratio increases, the tail is thicker, but requires a longer time to evolve.

The shear stress (tangential stress) and the tangential velocity at the oil-water interface play important roles in the separation mechanism. At low viscosity ratios (low oil viscosity), the tangential velocity at the interface is high, so there is internal flow within the film. This leads to a relatively quick deformation. As the viscosity ratio increases from small values to 1, the shear stress increases at the interface, enhancing the separation. For $p > 1$, the separation mechanism changes. Although the shear stress increases, the tangential velocity at the interface decreases. This slows the internal flow within the oil film and hence slows the deformation of the interface. The non-dimensional tangential velocity and the non-dimensional tangential stress used in the simulations are as follows:

- non-dimensional tangential velocity $= \frac{V_\theta}{\sin \theta U}$
- non-dimensional tangential stress $= \frac{R_0 \tau_{r\theta}}{\sin \theta \mu_c U}$

where U is the terminal velocity of an oil-coated particle, V_θ is the tangential velocity of the fluid-fluid interface at angle θ , and $\tau_{r\theta}$ is the tangential stress on the fluid-fluid interface at angle θ . Fig. 7 illustrates that an increase in p decreases the non-dimensional tangential velocity on the interface; Fig. 8 illustrates how the non-dimensional tangential stress changes as p increases. It can be seen that both the tangential velocity and tangential stress become constant for $p > 20$.

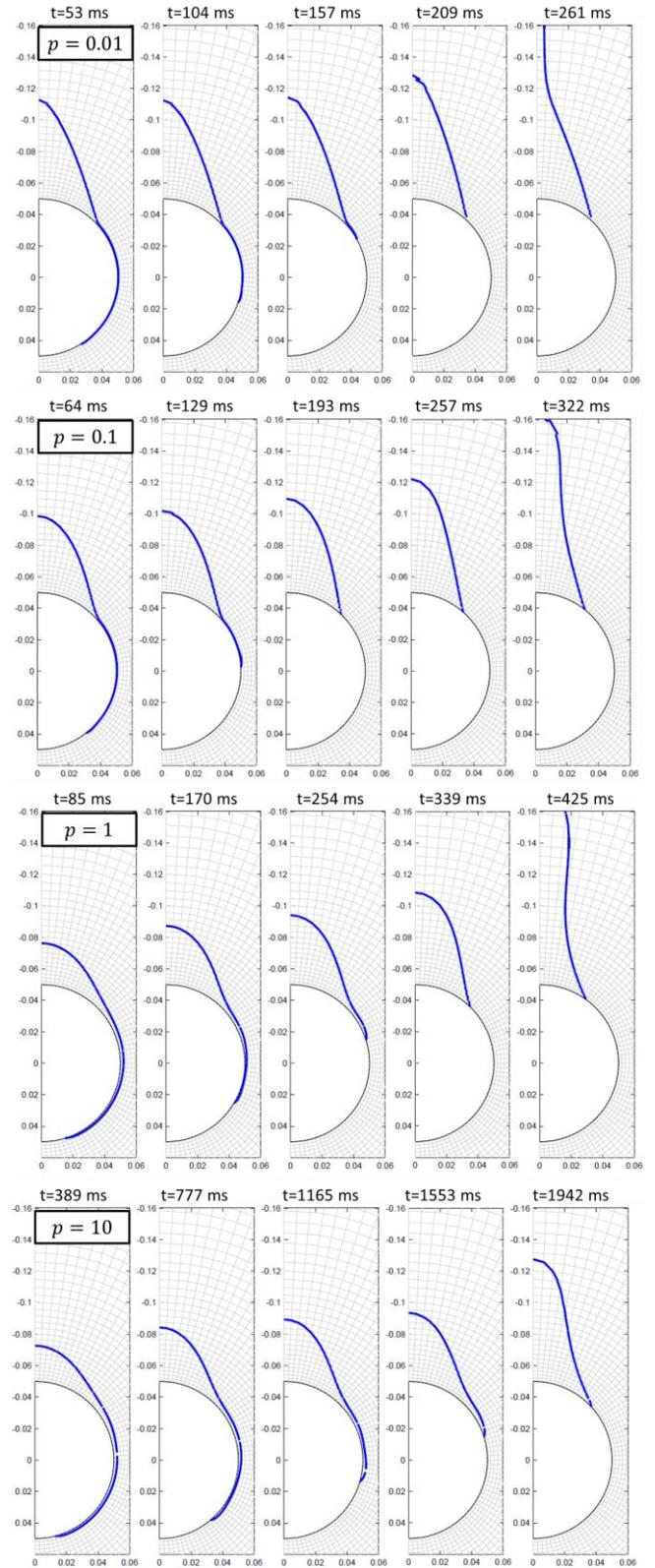


Figure 6. Film evolution for different values of p , $\varepsilon = 0.077$.

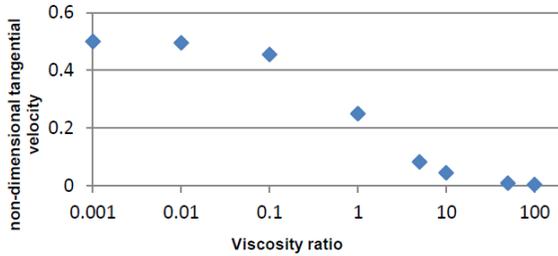


Figure 7. Effect of viscosity ratio on the non-dimensional tangential velocity at the interface.

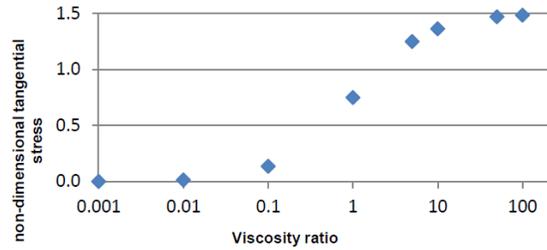


Figure 8. Effect of the viscosity ratio on the non-dimensional tangential stress acting on the interface.

CONCLUSION

Our experimental results show that when p is small, the oil instantly begins to deform and separate, while for large p , more time is required for the oil to separate but with a higher rate. It was seen that maximum separation could be achieved when $0.1 < p < 1$.

The numerical results show that increasing p from small values to 1 increases the shear stresses on the oil-water interface that facilitates separation. As p is increased further the tangential velocity at the interface decreases, which slows the deformation of the interface. Consequently separation takes place with a delay.

To conclude, the mechanism of oil separation from a solid particle can be expressed for three different regimes of viscosity ratio:

- (I) Low p , where the tangential velocity is the dominant factor in the separation process.
- (II) Intermediate p , where both the tangential velocity and shear stress influence the separation. The fact that both of the factors contribute to separation leads to a higher degree of separation.
- (III) High p , where the effect of tangential shear stress on the separation dominates over the tangential velocity.

NOMENCLATURE

h	Initial oil film thickness (m)
K	Consistency index ($Pa \cdot s^n$)
n	Power law index
p	Viscosity ratio (μ_d / μ_c)
R_0	Solid particle radius (m)
U	Terminal velocity (m/s)
V_θ	Tangential velocity at the fluid-fluid interface (m/s)
ε	Dimensionless film thickness (h/R_0)
σ	Interfacial tension (N/m)
μ_d, μ_c	Viscosity of the dispersed phase and aqueous solution, respectively
ρ_d, ρ_c	Density of the dispersed phase and aqueous solution, respectively
$\tau_{r\theta}$	Shear stress at the fluid-fluid interface (Pa)

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