

## COALESCENCE PHENOMENON AT THE INTERFACE BETWEEN IMMISCIBLE FLUIDS

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*The paper is concerned with experimental investigation of the water drop coalescence phenomenon at the interface between water – air and water – oil, respectively. Differences between these interfaces are indicated by representing the time evolution of the coalescence phenomenon and associated processes - drop resting time on the interface, bouncing time, partial coalescence, coalescence total time, time evolution of the vortex ring formed in the underlying liquid. Increasing viscosity in the layer above water suppresses the partial coalescence and decreases the coalescence total time.*

**Keywords:** coalescence, immiscible fluids, interface, vortex ring.

### 1. Introduction

Coalescence is defined as the mixing phenomenon of two identical or miscible fluids placed in contact. It employs physical and chemical principles, along with dynamics principles of fluid interfaces breakup. Drop coalescence studies are important in understanding interfacial processes from nature and industry. Along with emulsions or foams stability (food, pharmaceuticals and cosmetics industry), it plays a key role in liquid/liquid separation (petroleum industry), sprays, efficiency of separators or mixing vessels, optimization of internal combustion processes and manipulation, transport or mixing processes in microfluidic devices [1][2].

Studying drop – interface coalescence, along with drop - drop coalescence is of interest in oil - water separation in emulsions, with important applications in petroleum industry [3]. One of the main obstacles in oil recovery is that extracted crude oil contains a considerable amount of water, which must be separated. This separation occurs at ground level and the water must be processed to remove benzene, toluene and other pollutants. Coalescence dynamics is complex. There has been an active research regarding drop – interface coalescence, but up to date

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there is no comprehensive study to provide a solid benchmark for the new generation of two - phase separators.

In 1881, Lord Rayleigh observed raindrops remaining on a dust - free water surface for some seconds before coalescing with the underlying water, temporary noncoalescence. The phenomenon of drops floating on the same liquid surface was observed in the case of water drops coalescing with water free surface, due to partial coalescence. Charles and Mason, in 1960, published their investigations regarding drop coalescence with interfaces, explaining the stages and defining the partial coalescence [4]. The phenomenon of secondary drops that succeed the primary drop in the coalescence process was described as partial coalescence. The daughter drop formed may either bounce or coalescence.

The partial coalescence is distributed both before and after bouncing, as a transitional regime between bouncing and coalescence. This can be qualitatively explained by the competing effects between the viscosity and surface tension [5] [6]. For partial coalescence, gravity is known to be as important as surface tension. If Ohnesorge number is high (high viscosity), a mechanism enhances the emptying of the drop, resulting in a premature total coalescence [7].

Control and reproducibility of the droplets are very important in nowadays technology. Obtaining single small droplets in motors industry is important in injection processes; efficiency of combustion is increased for smaller droplets. Partial coalescence is a suitable way to achieve it [2].

Couder et al [8] have shown that a drop is able to bounce infinitely on the surface of a liquid pool without coalescing. This effect is basically obtained when the liquid bath is vertically shaken. The droplet moves horizontally due to its interaction with the wave it produces on the liquid surface.

Drop coalescence with a planar interface of two immiscible fluids and the vortex ring associated offers a unique opportunity to investigate flow dynamics in the presence of a fluid interface. The present study is focused on drops driven by gravity to interact with a fluid lying beneath an initially flat interface, the coalescence phenomenon of a single drop on the interface between two immiscible fluids (water – oil and water - air interface, respectively).

## 2. Methodology

The experimental setup is composed of a glass cell with two layers of Newtonian immiscible fluids that create an interface. A needle is connected to a syringe pump. By injecting dyed fluid of the more dense fluid, at a constant flow rate, through the needle in the above lighter layer, a drop is created (Fig. 1). The drop formed falls down, settles onto the interface, rests the time necessary for thin film drainage of the surrounding fluid, then merge with the liquid beneath. The drop collapses in the water bulk, the drop's water sinks slowly and a vortex ring

can be observed in the underneath water layer. Extremely useful is the use of high - speed digital cameras to capture the phenomenon. Videos over the dynamics of this phenomenon were acquired using two fast CMOS camera (Photron SA1 at 250 fps and Phantom Miro at 3200 fps) with microscopic magnification.

The rest or residence time is the time necessary for the drop to squeeze the thin underlying film so the Van der Waals forces are able to generate a hole, which expands (decrease of the interfacial area and interfacial free energy).

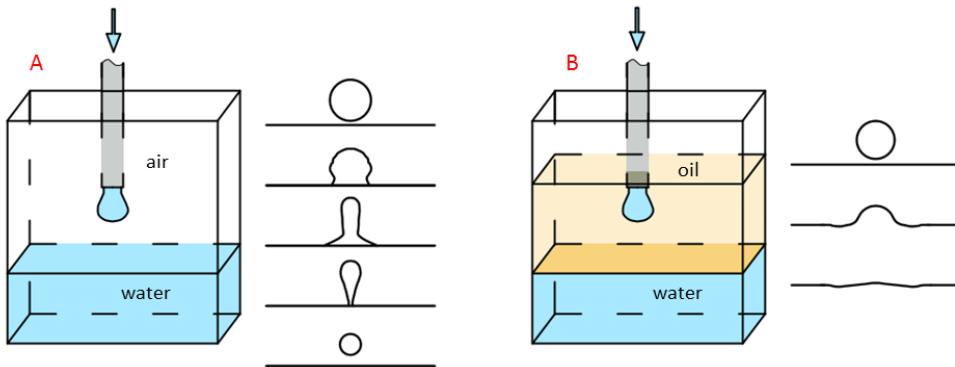


Fig. 1. Experimental method and sketch of coalescence: (A) for water drop on water free surface (evidencing first droplet of partial coalescence); (B) for water drop on the interface between water and oil (evidencing total coalescence).

This is the main step in time evolution of the phenomenon. Film squeezing and drainage has been intensively studied. Van der Waals forces, non - hydrodynamic forces, are required at short - range interaction to rupture the flattened thin film [9]. Hydrodynamic forces are driving forces to approach the drop and the interface together. The hole generation (not necessarily in the center of the film [10][11]) is the beginning of the coalescence phenomenon. After the film rupture, it retracts in all directions from the rupture point. This retraction is dominated by surface tension forces due to its very small thickness.

Further investigations regarding time evolution will consider the time prior to generation of the hole,  $t=0$ . The hypothesis that before rupture the drop was essentially motionless above the interface has to be made [12].

Two dimensionless numbers were used to quantify the experiments:

- Bond number ( $Bo$ ) - square ratio between the capillary time and the gravity time:

$$Bo = \frac{(\rho_1 - \rho_0)gR^2}{\sigma_1}$$

- Ohnesorge number ( $Oh$ ) - the relative importance of viscous stresses and surface tension:

$$Oh = \frac{\eta_i}{\rho_i R \sigma},$$

where  $\rho_i = 1000 \text{ kg/m}^3$  is the water density,  $\eta_i = 10^{-3} \text{ Pas}$  is the water viscosity,  $\sigma [\text{N/m}]$  is the interfacial/surface tension ( $\sigma_{\text{water-air}} = 0.07 \text{ N/m}$  and  $\sigma_{\text{water-oil}} = 0.037 \text{ N/m}$ ),  $R [\text{m}]$  is the radius of the water drop (measured for the pictures) and  $\rho_0 [\text{kg/m}^3]$  is the density of the above layer ( $\rho_{\text{oil}} = 910 \text{ kg/m}^3$  for organic oil and  $\rho_{\text{air}} = 1.2 \text{ kg/m}^3$  for air). Viscosities were measured using Anton Paar Physica MCR301 rheometer with cone - plate geometry and interfacial tensions were quantified using pendant drop method.

### 3. Results

In the experiments presented, the Bond number ( $Bo$ ) is small: 1.4 for water – air interface and 1.7 for water - oil. Ohnesorge number ( $Oh$ ) is and 10.5, respectively.

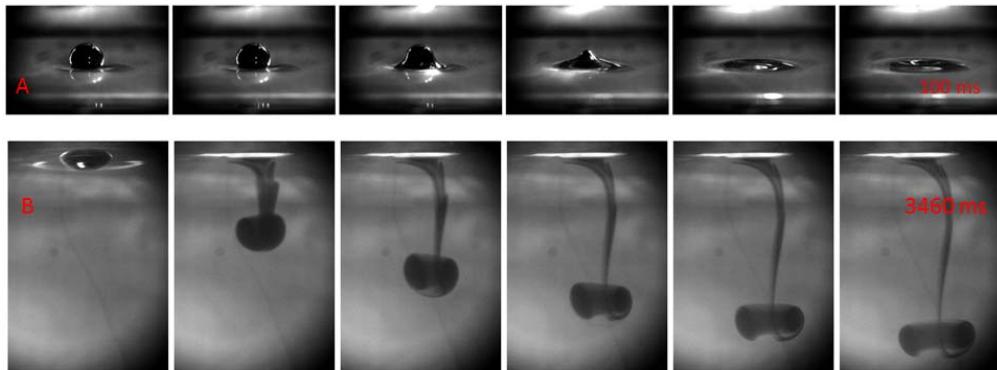


Fig. 2: (A) Total coalescence of a water drop with the interface between water and organic oil (each two frames are separated by 20 ms); (B) Vortex ring generated (each two frames are separated by 728 ms).

Experimentally, the water drop may sit on the interface when reaching it for tens of seconds (in the case of oil - water interface the oil thin film between the interface and the drop has to be drained before beginning of coalescence) or coalescence starts immediately (in the case of water free surface, due to very low viscosity of air). This residence time represents the time necessary for the thin film of oil to be drained between the water drop and the oil – water interface.

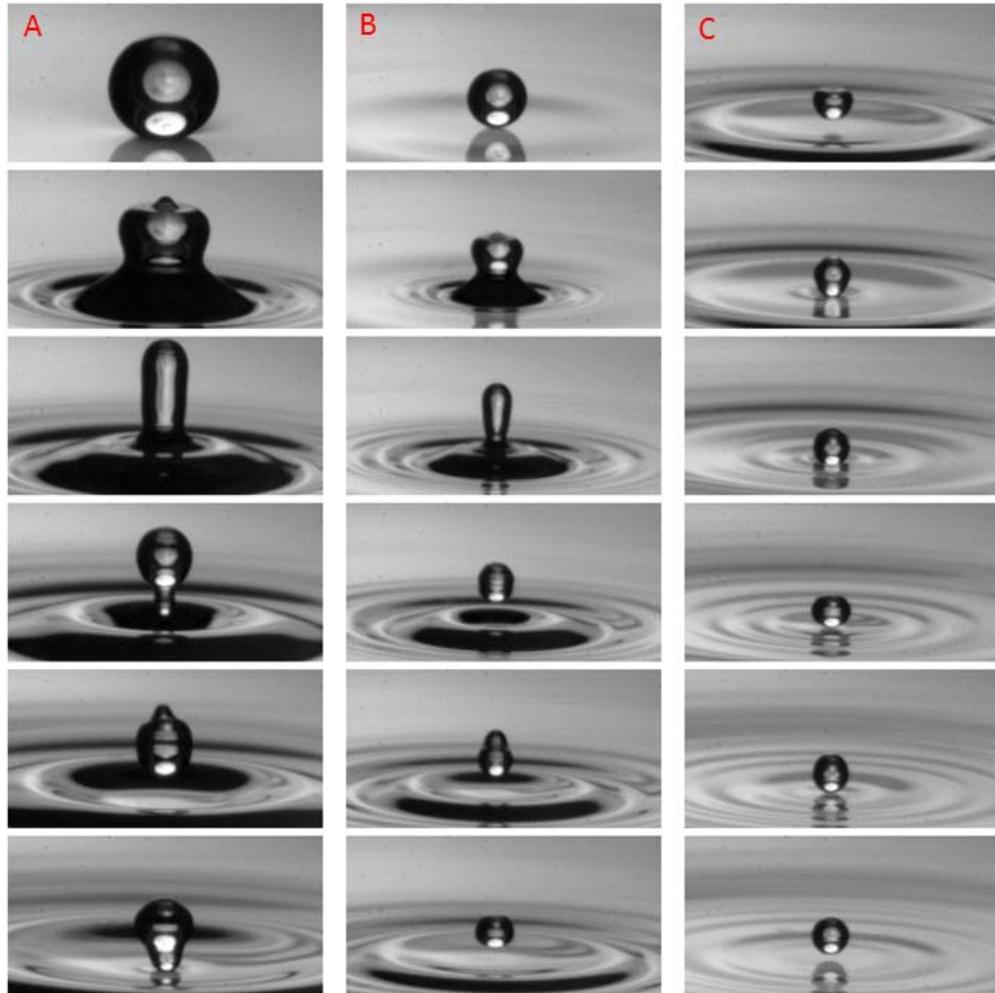


Fig. 3: (A) Coalescence of a water drop with water free surface; each two frames are separated by 1.25ms. (B) Formation of the second daughter drop; each two frames are separated by 1.25ms. (C) Bouncing of the second drop; each two frames are separated by 0.625ms.

Drainage of the film is governed largely by the balance between gravity, which produces a high pressure in the film centre, and viscous force resisting the flow in the thin film [13].

Coalescence of water drops at the interface between water and organic oil is total and rapid (80 ms) (Fig. 2A). When it is drained in the underlying water phase, the drop fluid sinks slowly and a vortex ring with an elongated structure can be observed if the drop is dyed. The evolution in time of this vortex in the water phase is slower (Fig. 2B). At early stages of the coalescence phenomenon,

vorticity is generated to maintain zero viscous shear stress at the interface. When the curved drop surface is in contact with the interface and the drop fluid is collapsing, a curved streamline is responsible for the development of this vortex ring [14].

In the case of water drop coalescence with water free surface, daughter droplets are formed in air, leading to partial coalescence, as detailed in Fig. 3. Up to 5 daughter droplets can be observed for a  $2\text{ mm}$  diameter water drop. Formation of the first daughter drop takes place in  $6\text{ ms}$  and the merge of the entire droplet water in about  $270\text{ ms}$ .

An interesting feature of the coalescence cascade is the large rebound height of the smaller droplet on the interface after bouncing. Droplets also experience large deformations during bouncing. The rebound height is observed from the images as the distance between the interface and the bottom limit of the drop shape at maximum bouncing height. The rebound height is increasing with decreasing daughter droplet sizes [15]. When the process of coalescence starts, surface waves can be observed. These are capillary waves generated at the bottom of the drop after the thin film rupture. The waves are generated by the receding interface below the drop [4][7]. A part of these waves climbs over the drop and converges at the top of the drop. The convergence of capillary waves on the top of the droplet has a crucial importance on the partial coalescence process. These waves travel along the droplet and are responsible for the delay in the vertical collapse. They, also, have an impact on the rate of daughter/mother droplet radius. Thus, formation of daughter droplets can be predicted [8]. Replacing air with oil in the experiments suppresses these capillary waves and, also, the partial coalescence.

#### 4. Conclusions

The main objective of the study was to indicate the differences between water – oil and water – air interfaces for single water drop coalescence with the water bulk in the presence of a fluid interface.

An experimental work has been made in order to study the above layer viscosity influence in coalescence of drops with liquid interfaces. Increasing viscosity in the above layer (oil instead of water) suppresses partial coalescence and delays the coalescence phenomenon total time. Figure 4 exemplifies the differences between these two interfaces: total coalescence time for water – oil interface is about  $80\text{ ms}$  and  $270\text{ ms}$  for water – air interface, respectively.

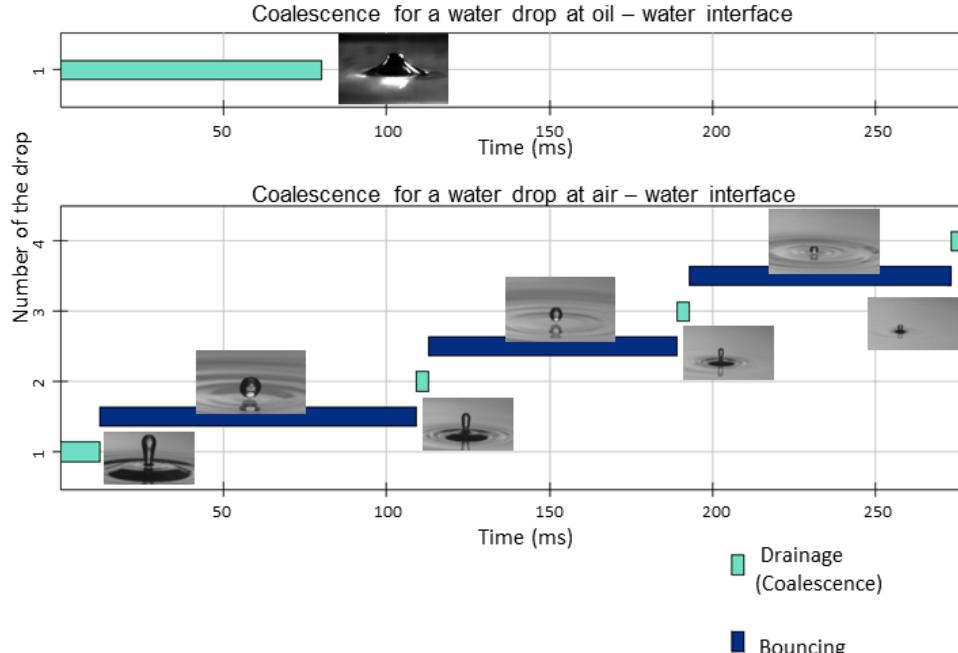


Fig. 4: Comparison between coalescence total time for oil – water and air – water interfaces.

Partial coalescence and the mechanism of daughter drops formation was, also, experimentally investigated and quantified in the case of water free surfaces. Bouncing water drops on water free surfaces were captured (Fig. 3).

Further investigations need to be made in order to establish the viscosity impact in the coalescence dynamics of drops with interfaces, phenomenon governed by a balance between gravity, viscosity, surface tension and inertia.

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