An unexpected flow regime found via selective withdrawal

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Abstract: *This paper explores various flow regimes that can be easily achieved through selective withdrawal. By varying the distance between the pipette and the interface, we observed that increasing the distance leads to an increase in droplet frequency and a decrease in droplet diameter. The study also highlights the transition to a different regime characterized by a corrugated liquid column, which we refer to as a Brâncuși column due to its striking resemblance to Constantin Brâncuși's sculpture, The Infinity Column. This phenomenon occurs when the distance between the pipette and the interface is reduced, acting as an upper boundary for droplet formation. When the viscous-capillary time scale, which governs droplet breakup, matches the convective time scale of the continuous phase, the transition to a corrugated fluid column takes place. Interestingly, the characteristic wavelength of these columns remains constant regardless of the distance between the pipette and the interface but increases with the extraction flow rate. The study also explores the selective withdrawal of water droplets standing at the interface between two immiscible liquids. Complete extraction or partial coalescence with the heavier liquid are observed during the extraction process of the droplet. The present exploratory study may prove beneficial for various droplet production processes which involve selective withdrawal as a basic method.*

Key Words: *selective withdrawal, droplet generation, immiscible liquids, interfacial instability*

1. INTRODUCTION

Fluids are extremely susceptible to shape modifications under stresses such as pressure or shear. Their ability to deform and adapt their shape according to various boundary conditions represents a fundamental aspect of fluid behavior.

In certain cases, these deformations can become sufficiently pronounced and lead to morphological transitions. A well-known example is selective withdrawal, where shear stresses can give rise to fluid threads [1].

When two liquids are placed into a reservoir, the two will settle into an overlayed configuration and a stable interface will be formed separating the two phases. If a capillary connected to a withdrawing pump is brought near the interface, the latter will destabilize and a thin jet of denser fluid will be entrained into the tube [2].

The fundamental physics of selective withdrawal depends on the importance of viscous stresses and interfacial tension [3].

Variations of the tube's position relative to the separating interface can influence the withdrawal process and several distinct flow regimes have been observed, from a thin liquid thread to a droplet generation regime and finally the formation of column-like structures [4]. A previous study has investigated the droplet's diameter and generation frequency in a similar configuration which can serve as a method for producing simple or double emulsions [5-7].

Previous studies have shown that a minimum withdrawal flow rate must be imposed for the entrainment of both liquids into the capillary regardless of the distance between its tip and the separating interface [8].

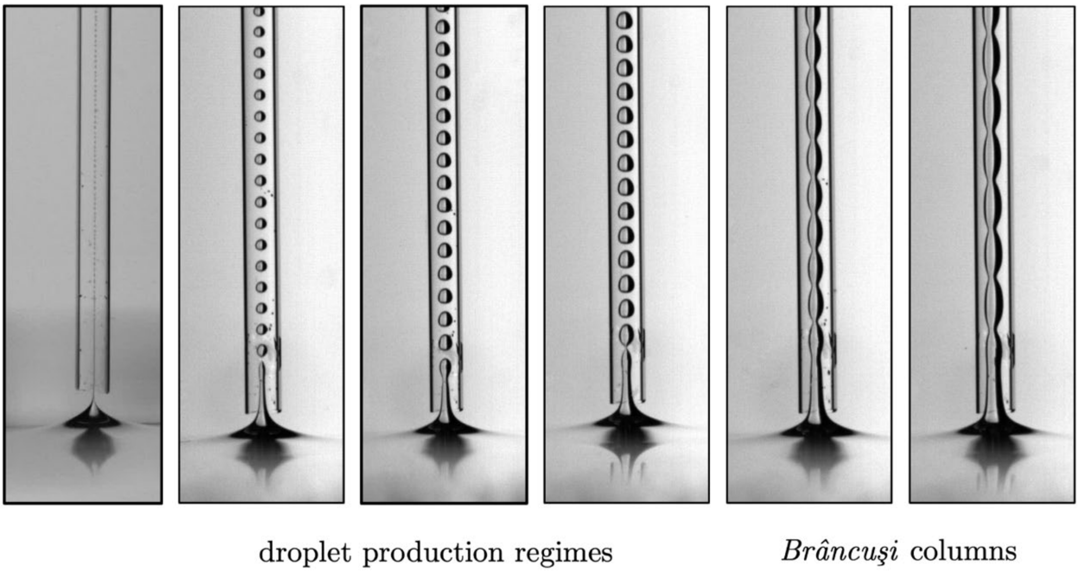

Fig. 1 – Distinct flow regimes in selective withdrawal obtained by lowering the capillary tip and imposing a constant withdrawal flow rate $Q = 5$ ml/min (the outer diameter of capillary tube was considered as the reference scale for measurements).

The latter influences the degree of liquid entrainment [9]. Selective withdrawal can also be observed in the case of miscible or stratified liquids, where surfaces of constant density can be considered apparent interfaces [10]. Numerical investigations tackling the selective withdrawal of viscoelastic liquids have shown that the interface is more deformed than the Newtonian case and that the phenomenon can serve as a method of steady-state elongational viscosity determination [11].

Selective withdrawal is a common method of controlling the quality of water released from stratified reservoirs [12]. It can also be used in transferring crude oil from large caverns as a safer withdrawal method for the environment [13]. The production of micrometer-size droplets represents a problem of interest in many domains, such as pharmaceutics and biotechnology [14]. Additionally, selective withdrawal can act as a mechanism for coating microparticles which is paramount in pharmaceutics and biotechnology [15].

In this paper, we extend previous studies through a detailed investigation of droplet formation as a function of variations in the position of a capillary, and the formation of peculiar fluids structures previously unreported in the literature, one of which shows a spectacular resemblance to one of the most famous sculptures by Constantin Brâncuși (see Fig. 1). We examine the effects on droplet size and frequency, as well as the associated flow spectra, while simultaneously identifying the limitations of the method when applied to droplets located near the interface.

2. EXPERIMENTAL DETAILS

The experimental setup described in Fig. 2 consists of a glass vessel containing two immiscible liquids, a mechanical mechanism for the positioning of the Pasteur pipette, and a syringe pump connected to the pipette that extracts the liquids at a constant flow rate. Experiments were performed using standard Pasteur pipettes with an inner diameter $D_i = 0.97$ mm at the tip and the immiscible liquids used were distilled water as the heavier fluid and a layer of sunflower oil on top.

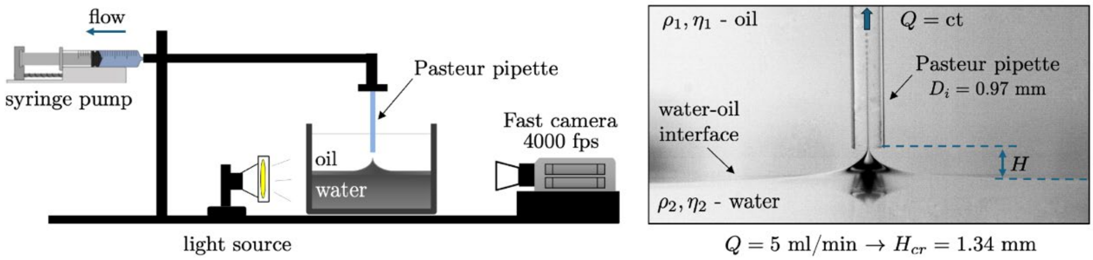

Fig. 2 – Schematic representation of the experimental setup and a typical example of selective withdrawal

The Pasteur pipette is lowered into the sunflower oil layer and placed in the vicinity of the separating interface. An extraction flow rate is imposed and at the beginning only oil is withdrawn. By means of the mechanical mechanism, the pipette is further lowered until both fluids are extracted. When the flow regime settles, the high-speed camera records the phenomenon at 4000 fps. The recorded frames are then processed via ImageJ software. The control parameters are the flow rate of the extraction Q and the position of the pipette tip relative to the flat surface H. For each imposed flow rate the distance between the interface and the tip is modified in order to capture each regime.

The material properties of the working liquids, viscosity and density, are $\eta_w = 1$ mPa s and $\rho_w = 998$ kg/m^3 for water, $\eta_o = 55$ mPa s and $\rho_o = 920$ kg/m^3 for sunflower oil, with an interfacial tension $\gamma_{w-o} = 25$ mN/m. The properties were determined via standard procedures. The densities of the two liquids were determined via the mass-per-volume method. The viscosities were measured using an Anton Paar Physica rheometer with a cone-plate geometry. The pendant drop technique was employed to estimate the interfacial tension between the two immiscible liquids.

3. UNSTABLE FLOW REGIMES LEADING TO DROPLET FORMATION

As the distance between the pipette and the separation surface of the two immiscible liquids is varied, at a constant extraction flow rate the morphology of the fluid filaments entrained by viscous stresses undergoes significant changes.

When the Pasteur pipette is sufficiently close to the interface, the fluid in the lower layer is extracted in the form of a thin thread which breaks up into droplets due to the interfacial tension (see Fig. 1). The entrained liquid thread exhibits instability and breaks into droplets via Rayleigh – Plateau instability. The instability is caused by the interfacial tension which drives the system towards a state which minimizes surface area. Droplets grow, pinch off, and are transported by the surrounding fluid. The regime is periodic and of great practical importance.

As the distance between the pipette and the interface decreases, the volume of the entrained liquid increases which induces an increase in droplet diameter. Fig. 3-a shows that, for a constant flow rate, there is a decreasing trend in droplet diameter as the distance between the pipette and the interface increases. An analysis of the experimental data on droplet sizes highlights a maximum diameter of 0.7 mm at a pipette–interface distance of $H = 0.72$ mm and a minimum diameter of 0.08 mm at $H = 1.22$ mm. Similarly, Fig. 3-b shows that the droplet generation frequency decreases as the pipette tip approaches the interface.

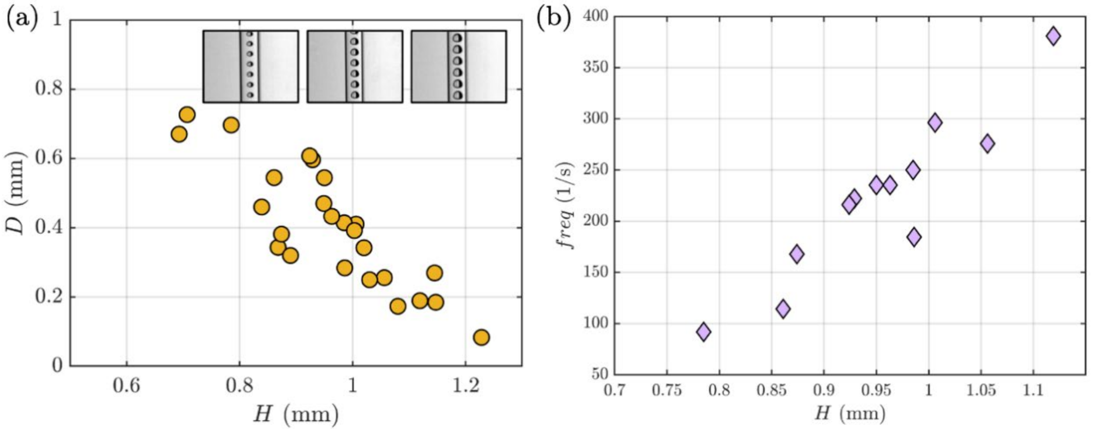

Fig. 3 – Droplet diameter (a) and generation frequency (b) as a function of the distance between the capillary tip and the separating flat interface. Measurements were performed at a constant flow rate Q = 5 ml/min

4. AN UPPER BOUNDARY FOR DROPLET FORMATION

As the capillary tube is brought closer to the interface, because the volume of the entrained fluid increases, the fluid thread that previously separated into large diameter droplets becomes a jet which behaves in an unexpected way. In regimes that display droplet formation, the time scale associated with droplet detachment, which is given by the visco-capillary time scale $t_{vc} = \eta_o r/\gamma_{wo}$, with r as the radius of the fluid thread, is smaller than the convective time scale associated to the continuous phase. As the tip is lowered, the system may reach a state where both time scales are of the same order. This gives rise to a column-like liquid structure, similar to a corrugated jet (see Figure 1 and Figure 4). We call this peculiar fluid structure a *Brâncuși column* after its striking resemblance to the artist's iconic sculpture, The Infinity Column (see Fig. 4-a).

The phenomenon was observed for a constant extraction rate by modifying the position of the capillary tube relative to the flat interface. Another striking similarity is represented by the ratio between the largest and smallest values of the diameter, as shown in the Fig. 4-a, where in the case of Brâncuși's sculpture the value is 2, while in the case of the regime obtained in the experiment the value is 2.12 ± 0.05.

For a constant extraction rate, the threshold value of the distance between the pipette and the interface at which these peculiar fluid structures appear is approximately 0.9 mm. The variation of the two characteristic dimensions, the intact jet length that does not show corrugations - l_{jet} and the wavelength - λ, is represented in Fig. 4-b. We have found that both characteristic dimensions do not vary significantly with H, with an average wavelength of 1.33 mm and a jet length ranging between 2 and 2.5 mm.

It is useful to examine the dependence of these characteristic dimensions on the extraction flow rate. It can be easily observed in Fig. 5 that as the flow rate increases the wavelength decreases, while the jet length increases. For a constant distance between the pipette's tip and the separating interface, by increasing the flow rate the convective effects develop faster than the interfacial instability and the associated time scale, $t_c \sim Q^{-1}$, becomes smaller than the visco-capillary time. The liquid jet becomes unstable more slowly when increasing the flow rate and the length of the unperturbed jet increases.

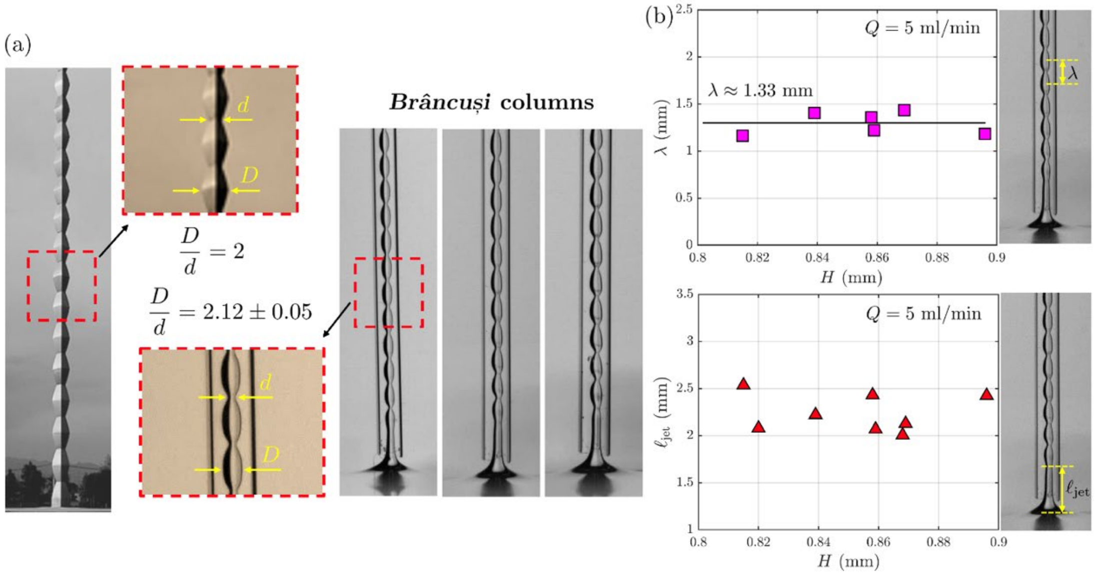

Fig. 4 – (a) The Infinity Column (sculpture by Constantin Brâncuși) vs. several column-like liquid structures obtained via selective withdrawal. (b) Wavelength and jet length as functions of H ($Q = 5$ ml/min)

5. THE SELECTIVE WITHDRAWAL OF DROPLETS PLACED ON A FLUID INTERFACE

We also report on the selective withdrawal of water droplets that were placed at the interface between two immiscible liquids where two distinct behaviors are observed. The first behavior, illustrated in Fig. 6-a, is characterized by the complete extraction of the droplet, which in this case does not interact with the water–oil interface. In this context, flow regimes like those presented in section 3.1 were identified. The water droplet, initially placed on the free surface between the two fluids, is extracted through the tube. As the droplet volume decreases, it detaches from the interface, but selective withdrawal continues until the entire droplet is extracted. After detachment, the water droplet approaches the capillary’s tip until a critical distance for the droplet generation is reached. The distance is reduced during the water extraction and the unstable column-like structures are observed until the complete withdrawal of the droplet.

The second type of behavior is characterized by a coalescence event between the water droplet placed on the interface and the water in the underlying layer (see Fig. 6-c). When the fluid layer between the droplet and the interface becomes sufficiently thin, the lower part of the droplet merges with the water bath, while the upper part continues to be extracted. In this case as well, droplet generation regimes are observed in the tube, as described in the previous subsections. Once the droplet is almost completely extracted, while part of its mass coalesces with the underlying water, a small droplet forms at the end of the process, floating in the oil layer and immediately being aspirated through the tube. Due to the coalescence phenomenon, the water droplet merges with the liquid bath and does not approach the pipette’s tip. Only a thin liquid thread and the corresponding droplet regimes can be observed. As the liquid is extracted through the capillary, simultaneously draining to the underlying bath, the droplet's size is reduced until it becomes small enough to be completely absorbed by the sink flow.

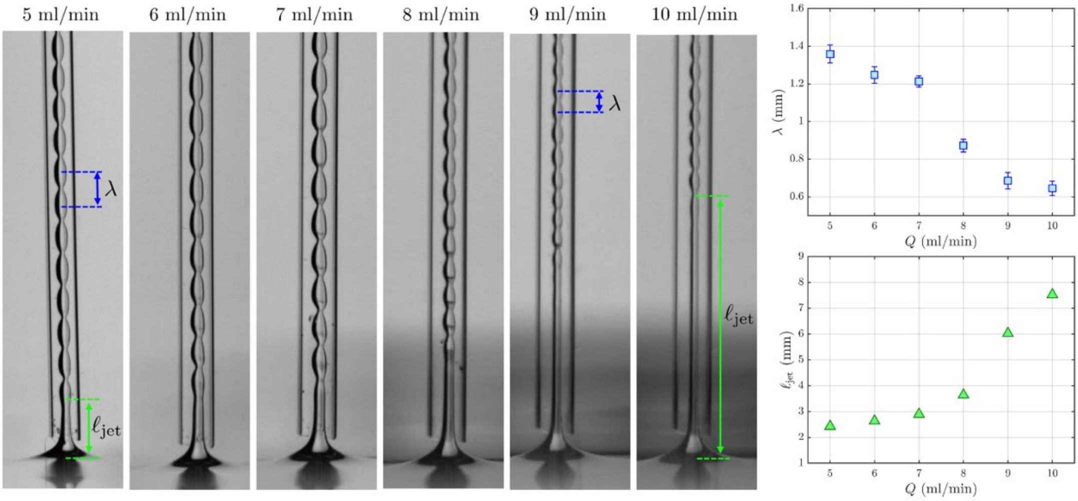

Fig. 5 – Variation of the wavelength and the jet length as a function of the extraction rate Q

The graph in Fig. 6-b shows a linear increase in the distance between the pipette and the interface as a function of the droplet's diameter. In other words, the larger the droplet volume, the greater the distance between the pipette and the interface required for extraction. The trend line through the experimentally determined points is obtained by linear regression. The previously identified equation can be used to predict the critical distance at which selective withdrawal occurs when the interface is flat, in the absence of a droplet. This distance corresponds exactly to the pipette tip position relative to a free interface, marking the onset of viscous entrainment (see the green box in Fig. 6-b).

6. CONCLUSIONS

The paper investigates multiple flow regimes that can be easily achieved via selective withdrawal. Depending on the application, the diameter and the frequency of the droplets can be varied by adjusting the distance between the capillary pipette and the interface or by changing the extraction flow rate. As the distance between the pipette and the interface increases, the frequency of droplet generation increases while the diameter of the droplets decreases.

This study also highlights the transition to another regime characterized by a corrugated fluid column, which we denote by a *Brâncuși column* after its striking resemblance to the sculpture The Infinity Column by Constantin Brâncuși. The phenomenon was observed when decreasing the distance between the pipette and the interface as an upper boundary for the droplet formation regime when the viscous-capillary time scale, which characterizes the breakup of a droplet, is of the same order as the convective time scale of the continuous phase. We have also observed that the characteristic wavelength of these columns does not vary with the distance between the pipette and the interface but increases with the extraction flow rate.

During the selective withdrawal of droplets which were placed on the liquid-liquid interface two different behaviors have been observed.

If the droplet keeps its integrity and does not merge with the underlying liquid bath, several types of regimes can be observed (from the formation of thin liquid thread to the generation of smaller droplets and finally the unstable column-like structure) until its complete extraction.

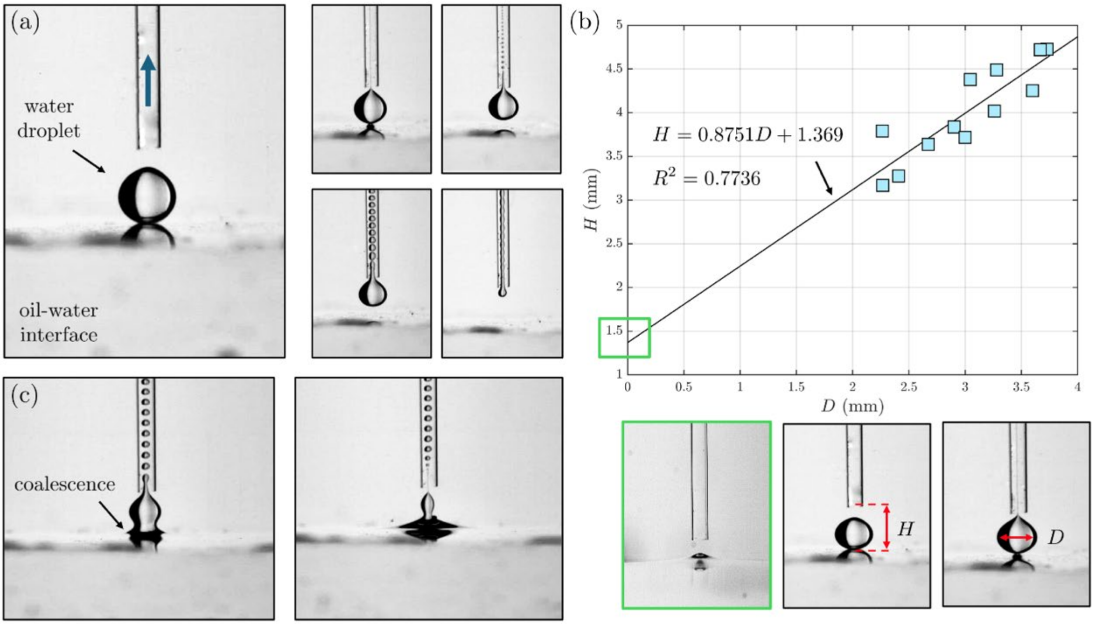

Fig. 6 – (a) Selective withdrawal of a droplet placed on a fluid interface. (b) Measured data for the diameters of extracted droplets as a function of the distance between the pipette and the fluid interface on which the water droplet is placed; solid line – the linear approximation ($R^2 = 0.77$) vs. experimental data. (c) Droplet extraction with a coalescence event

In the case of coalescence, the droplet drains to the water bath while being simultaneously extracted through the capillary. Due to the coalescence event, the distance between the tip and the droplet is not sufficient in order to achieve the unstable column regime.

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