THE INFLUENCE OF SURFACTANTS ON IMPACTING DROPS

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ABSTRACT In many practical applications, the coating of solids is obtained by spraying complex liquids such as surfactant or/and polymer solutions on the surface. Upon collision with the solid, the droplets become highly deformed in the shape of a more or less flat wetting film in a few milliseconds, and then, they relax towards equilibrium. This very strong and short deformation causes the droplets to so much depart from equilibrium that the transient values of the physical properties of the liquid itself can be drastically different from the equilibrium ones. In the case of polymer solutions, elastic stresses arise from the elongation of the polymer chains, and in the case of surfactant solutions, there is no longer equilibrium between the adsorbed surfactants at the surface and those in the film core. The consequences are severe for the quality of the coating which is damaged by the excessive occurrence of splashes, of detaching satellite droplets, or of thickness irregularities. To analyze these effects, we have realized impacts of drops on disk-like targets of same size as the diameter of the impacting drop. Upon collision with these small targets, the drop is transformed into a thin lamella with free surfaces bounded by a thicker toroidal rim giving rise to splashes in a way which depends on the nature of the additives and their concentration in solution. The high and fast deformation of the drop liquid with the above mentioned transient change of the physical properties similarly occurs on a small target that it does on a large plate, but the viscous interaction of the liquid with the solid surface is now suppressed. The physical situation is therefore simpler in the former, which enabled us to calculate the drop dynamics and to carefully analyze the influence of the dynamic physical properties of the surface-active solutions in a pure view. A major practical conclusion of our studies is that under very similar impact conditions, surfactants can either stabilize the lamella or destabilize it depending on their nature and their concentration in solution.

Keywords: Droplet impact, Surface-active solutions, Dynamic surface tension, Small target

1. INTRODUCTION

Most liquids in spraying processes are surfactant or polymeric solutions at different degrees. To know how drops from these liquids behave in impact processes is of utmost importance for aerosol coating, inkjet printing, agricultural spray and for any technologies where liquid droplets are impacting a substrate.

The collision of a drop with a plane unbounded solid substrate results in a very fast liquid lamella growth and a subsequent relatively slow retraction. In the case of a drop of a surface-active solution impacting a planar solid substrate of low surface energy [1-2], it was observed that surfactant additives do not influence the growth-stage, whereas they can very significantly slow the lamella retraction [3]. During the stage, the liquid in the drop experiences a high rate of bulk and surface deformations, and the subsequent drop retraction is driven by the ‘response’ of the liquid to these extreme circumstances. Generally speaking, a ‘fresh’ surface is formed very fast in comparison with the kinetics of surface adsorption of the surfactant; therefore the process is controlled by a dynamic surface tension effect. Then, the retraction stage is influenced by the ability of the surfactants to restore the equilibrium surface concentration decreasing the dynamic surface tension towards its equilibrium value.

The purpose of the present work is the understanding of the surfactant-related mechanisms that drive the lamella dynamics as well as its disintegration. To achieve this task, we have used a small target instead of a large plate. The reason for this particular experimental configuration is to remove the viscous friction caused by the drag exerted by the solid plate on the drop liquid, and to analyze the role of the surfactants in the impact process in a pure view.

2. EXPERIMENTAL

2.1 Materials

The tested liquids were aqueous solutions of dioctyl sulfo succinate sodium salt (DOS) and of trisiloxane oxypropylene polyoxyethylene (Silwett L77). DOS was supplied by Aeros Organics and Silwett L77 by Crompton Europe S.A. Milli-Q distilled water at room temperature was used as solvent in all cases. DOS molecules have two long chains and as their concentrations increase in solution, they organize as vesicles [4], while Silwett L77 molecules which have a very large polar head with only one chain form more or less spherical micelles at low concentration and then lamellar aggregates of different kinds as the concentration increases [5].

The main difference between DOS and Silwett L77 in solution at same concentration (in cac unit) is the following: DOS is a ‘fast’ surfactant, which means that the solution surface tension relaxes relatively rapidly to its equilibrium value if a ‘fresh’ surface is formed. On the contrary, Silwett L77 is a very effective, however a ‘slow’ surfactant, its surface tension relaxing relatively slowly toward equilibrium. Figure 1 illustrates these properties.
2.2 Experimental procedure

Drops were slowly generated at the tip of a capillary connected to a syringe pump in 30 – 60 seconds. This time is large enough to ensure equilibrium between the surface-active molecules in the bulk and at the surface. Drops detached from the capillary under the action of gravity, fell from a height equal to 65cm, and reached the target at velocities \( v_i \) of order of 3.4 m/s. The drop diameters \( d_i \) were in the range of 2.5 - 2.8 mm.

Under typical conditions, \( d_i=2.7 \text{ mm}, \mu=1\text{mPa}\cdot\text{s}, \) and \( \gamma_w=72\text{mN/m} \) (\( \gamma_w \) is the water surface tension). The time scale of the process is \( t_*=d_i/v_i=0.794 \text{ ms} \), the impact Reynolds number \( \text{Re}=ho v_i d_i/\mu \) is 9180, and the impact Weber number \( \text{We}=ho v_i^2 d_i/\gamma \) is 433.

The carefully polished planar end surface of a stainless steel cylinder (\( \Phi = 3.90\pm0.05 \text{ mm} \)) with a slightly blunt edge (\( = 0.1 \text{ mm} \)) was used as a target. The cylinder axis was aligned with the drop trajectory. Before each impact, the target surface was processed by a jet of compressed nitrogen to remove the liquid remained there from previous impacts.

We used a video recording of top views of the drop impact by means of a high speed camera equipped with a strobe lightening. The grabbing frequency was 1000 frames per second and the exposure time 1 \( \mu \text{s} \). Output frames had a resolution of 1024×512 pixels with 256 grey levels.

3. RESULTS

A few examples of top view observations of impacts of surfactant solutions drops are presented in figures 3-4. Images of water drops are also given for reference. The solutions concentrations were \( 1\times\text{cac} \) in figure 3, and \( 10\times\text{cac} \) in figure 4. For all the solutions, the drops form a thin liquid lamella with a relatively thick toroidal rim like the water ones [6].
Results show that the surface-active additives can modify the impact of water drops on a small target at least in three ways.

(1) The first observable modification is a more intensive finger formation and a slight increase of the stability of the liquid fingers in comparison with the case of pure water. The most stable fingers are obtained with DOS drops. Secondary jets ejected from the rim look longer than in the case of pure water, they do not quickly disintegrate, but they transform into liquid filaments, forming a spider-like structure, like for polymer solutions [7]. However, in the case of surfactants the reason for this aspect is different from the one for polymers. The increase of the filament lifetime is not here related to inner elastic stresses in the filament as it occurred with polymer solutions, but it is caused by the decrease of the surface tension, a high value of the surface tension favouring filament disintegration.

The second modification is connected with the influence of the surfactants on the lamella spread factor. Figure 5 shows that for DOS solution at C=1×cac, and for Silwett L77 solutions at C=1 and 10×cac, the spread factor \( \beta_m \) satisfactorily coincides with the one for pure water. In particular the maximum spread factors, \( \beta_m \) for water and \( \beta_m \) for these solutions can be estimated as \( \beta_m \approx 5.0 \). It means that the impacts are controlled by the dynamic surface tension, which is practically equal to the water surface tension for these solutions. Besides, figure 5 shows that the spread factor of the DOS solution at C=10×cac significantly exceeds the water spread factor as \( \tau > 2 \), its maximum size being \( h \approx 6.0 \). Besides, the lifetime of the lamella becomes noticeably larger than the one of water under same impact conditions.

The third modification of the lamella caused by the presence of surfactants was rather unexpected: holes spontaneously nucleated in the internal part of the lamellas of Silwett L77 solutions at \( t \approx 3 \) ms for the dilute solution and at \( t \approx 2 \) ms at 10×cac.

4. DISCUSSION

To analyse the effects of the surface tension changes on the lamella flow, we have modelled the lamella formation as resulting from a steady cylindrical liquid source (figure 6), of diameter \( d_i \), supplying an axisymmetric free liquid film with initial flow rate \( q_s \), velocity \( v_s \), thickness \( h \), liquid surface tension \( \gamma \), and surface concentration of surfactant \( \Gamma_s \) at the point of ejection \( r=r_{me}/d_i/2 \) like in [8]. The ejection parameters are \( v_s=v_i, d_i=d_i, \) and \( \gamma=\gamma_c \).

The lamella behavior results from the continuous competition between the inertial forces that drive the drop liquid outwards (figure 6) whereas the unbalanced capillary forces acting on the lamella surfaces pull back the liquid in a thickening rim at constant velocity relatively to the film (figure 7)

\[
\nu_t = (2 \gamma/\rho h)^{1/2},
\]

where \( \gamma \) is the liquid surface tension, \( \rho \) the liquid density and \( h \) the film thickness. The maximum extension of the lamella, \( r_m \), is obtained when the velocity, \( v \), of the free liquid film and the one of the rim velocity are equal, that is when the local Weber number, \( We=\nu_t^2h/2\gamma \), is equal to 1. With a water drop, it was shown in [8] that \( r_m = \rho q_v/4\pi \gamma_c \) and that the thickness of the lamella close to the rim \( h_{m}=q_s/(2\pi r_m v_s) \).

4.1. Equation of motion

Assuming that the film thickness \( h \) is small enough, in comparison to all other length scales, to use the approximation of flat velocity profile in the film, the kinematic and dynamic variables in the lamella are only function of the radial coordinate \( r: v=v(r), h=h(r) \).

The liquid surface tension, \( \gamma(r) \), changes under the influence of two factors. The first one is related to the residual surfactants initially adsorbed at the drop surface...
before impact, whose concentration drops due to the high rate of deformation experienced by the drop surface, increasing the local surface tension in different parts of the lamella. The second factor is the surfactant flux from the bulk lamella to the surface, which tends to restore the equilibrium surfactant adsorption (figure 8).

The equation of motion follows from the momentum balance for a motionless control volume in the lamella:

$$\frac{d (r p v^2 h)}{dr} = 2 r \frac{d r^2}{dr}$$

(2)

Introducing in (2) the continuity equation

$$q_s = 2 \pi r h v,$$

we obtain:

$$\frac{dv}{dr} = 4 \pi r^2 v r + r q_s dr$$

(4)

Equation (4) represents the balance between the lamella inertia and the so-called Marangoni stresses. It shows that, in a lamella with constant surface tension, the liquid velocity does not change [6,7]. If the surface tension $\gamma$ decreases as $r$ increases, then the liquid velocity $v$ also decreases, and vice versa.

4.2. Surfactant transfer kinetics

Surfactant mass balance is described as

$$\frac{J}{r} \frac{d}{dr} \left( r \Gamma v r \right) = J,$$

(5)

where $\Gamma$ is the surface concentration of the free surfactant molecules (monomers). The left term in (5) describes the surfactant transfer due to the deformation of the surface, and the right one describes the flux of surfactant, $J$, from the bulk to the surface [9].

Assuming that the surfactant transfer is purely diffusive, the flux $J$ is given by [10],

$$J = D \frac{\partial c}{\partial n} \bigg|_{n=0} \sim D \frac{c}{\delta},$$

(6)

where $D$ is the diffusion coefficient, $c$, the bulk concentration of the free surfactant molecules (monomers), $n$, the normal to the surface, $\delta$, the thickness of the diffusion boundary layer. The estimate (6) is based on the statement that due to the high rate of surface dilution, the surface concentration $\Gamma$ and the subsurface bulk concentration $c_{sub}$ are small in comparison with the saturation surface concentration $\Gamma_s$ and the bulk concentration $c$, accordingly [1-3, 11].

The magnitudes of $\delta$ and $c$ in (6) depend on the surfactant total concentration $C$. For dilute solutions, with $c=C \leq 1 \times cac$, the thickness $\delta$ grows with $r$ because of surfactant depletion in the solution in the vicinity of the surface induced by the consumption of surfactants by the diluting surface. In this case, $\delta$ scales as $r Pe^{-1/2}$, where $Pe=v r / D$ is the Péclet number [12].

For concentrated solutions with $C>1 \times cac$, the dilution of the solution near the surface is compensated by the disintegration of aggregates, where most surfactant molecules are concentrated. In this case, the limiting step to surfactant transport is the direct diffusive transfer of surfactant molecules from the disintegrating aggregates to the almost pure surface through a layer of constant thickness, $\delta$, which is of the order of the mean distance between two neighbour aggregates (figure 8). Since the concentration of free surfactant molecules in the neighbourhood of the aggregates is equal to $1 \times cac$, the difference between $c$ and $c_{sub}$ is also of the order of $1 \times cac$.

The flux $J$ can, therefore, be estimated as $J=\delta D cac / \delta$, where $\delta$ is now equal to the mean distance between two neighbour aggregates.

The leading role of the aggregates disintegration in concentrated solutions is confirmed by the increase of the lamella size for DOS at $10^{3} \times cac$ which is not as large as at $1 \times cac$ (figures 4,5) while the bulk concentrations of free surfactant monomers, $c=1 \times cac$, are the same in both solutions.

Below, we only consider a theoretical model for highly concentrated solutions in which surfactant effects are more significant. In this case, $\delta=const$ and the combination $Dcac / \delta$ is everywhere constant. A model for dilute solutions could be similarly considered assuming that $\delta=r Pe^{-1/2}$.

4.3. Constitutive equations

Integrating equation (5) with $J=\delta D cac / \delta$ and using the boundary conditions $r=r_s$, $v=v_s$, $\Gamma=\Gamma_s$, we obtain the surfactant distribution in the lamella surface:

$$\Gamma (r) v r = \Gamma_s v_s r_s^2 + D cac \frac{r^2 - r_s^2}{2 \delta}$$

(7)

Locally, the surface concentration $\Gamma$ is related to $\gamma$ by the asymptotic form of the Langmuir-Frumkin equation of state

$$\gamma(r) = \gamma_{sat} - RT T (r),$$

with $T \Gamma_s << 1$

(8)

where $R$ is the gas constant and $T$ the temperature.

Introducing (8) in (7), we obtain the dependence of $\gamma$ with $r$ as

$$\gamma (r) = \gamma_{sat} - (\gamma_{sat} - \gamma_s) \frac{v_s r_s}{v r} \frac{D cac \frac{r^2 - r_s^2}{2 \delta} RT}{vr}$$

(9)

where $v=v(r)$ is the velocity distribution in the lamella.

In this work, we are mainly interested by the influence of the surfactants and dynamic surface tension effect on the maximum size of the lamella. To simplify the physical problem, we assume that due to the fast deformation of the...
lamella surface the local surface tension $\gamma$ is far from equilibrium $1-\gamma X^*<1$, and that $J$ is the main restoring flux, neglecting by residual surfactant. Due to the last approximation we can suppose for simplicity $r_0=0$.

By substituting (9) into (4) with $X^*$ we obtain the equation of the sheet motion in dimensionless form as

$$\left(1-V^2\right)^{2} \frac{dV}{dX} = -\frac{FX}{V}$$

(10)

where $X=r/r_w$, $V=v/v_s$, $r_w=\rho_l\sqrt{4\pi\gamma}/\gamma_w$ is the maximum radius of steady water lamella [8] and $F=RTD\times cac_F=RT\delta/2\gamma_w$. The boundary condition is $V=1$ at $X=0$.

The analytical solution of (10) is obtained as

$$X = \frac{\sqrt{2V^2}}{F} \ln V.$$

(12)

The dimensionless form of the distribution of surface tension $\Sigma=\gamma X^*$ in the lamella is

$$\Sigma=1-FX/V.$$

(13)

The distribution of the local Weber number in the lamella $We_{sp}m^2/\pi\gamma_s$ is determined if both $V=V(X)$ and $\Sigma=\Sigma(X)$ are known. The condition $We=1$ that defines the position $X_m$ of the free rim of the steady lamella follows from (1). It writes:

$$We = \frac{V_m}{\Sigma_m X_m} = 1$$

(14)

where index “m” indicates the values near the free rim.

4.4. Effect of the surfactant flux

The solution $V(X)$ of (12), and the surface tension distributions (13) are plotted in figures 9 and 10 for different values of the parameter $F$ which measures the effect of the surfactant flux from the bulk to the surface. Only parts of the curves up to the points of return satisfy the boundary conditions, and therefore only these parts are appropriate from a physical point of view. They are drawn in black. These plots show that due to the surfactant flux, the surface tension gradient is negative, which causes a deceleration of the flow in the lamella. This effect increases as $F$ increases.

The common feature to all curves is that a steady flow exists only up to a certain critical point $X_c(F)$ beyond which it becomes impossible. At the critical point, $dV/dX=\Sigma dX/dX=-\infty$. Introducing $dX/dV=0$ in (12), we obtain the values of the critical velocity $V_c$ and of $X_c$ as $V_c=\exp(-1/2)=0.60653$ and $X_c=0.60653/\Sigma$. $V_c$ does not depend on $F$, whereas $X_c$ decreases as $F$ increases. The finite values of the velocity and of the thickness at the critical point mean that the flow continues beyond the critical point $X_c$, but it becomes unsteady or/and it loses its spatial uniformity: $v=v(r,t,X)$, $h=h(r,t,X)$, where $X$ is additional coordinate - the polar angle in cylindrical coordinate system. In other words, $X^*$ is the $F$-dependent position of the boundary between steady uniform and unsteady (or/and non-uniform) flow in the lamella. We do not know now which kind of flow is formed at $X^*>X_c$: regular oscillations, regular spatial waves, chaotic instability, turbulence or anything else? It is only obvious that this temporal or/and spatial instability can have a great influence on the lamella dynamics.

![Fig. 9: Effect of surfactant flux on the radial distribution of velocity $V$ for different values of $F$ (indicated on the corresponding curves). The horizontal dotted line represents the critical level $V=0.60653$. Symbols (+) show the positions of the liquid rim for different values of $F$.](image)

![Fig. 10: Effect of surfactant flux on the radial distribution of the surface tension $\Sigma$ for different values of $F$ (indicated on the corresponding curves).](image)

The maximum size of a steady free lamella $X_m$ is defined by equation (14), which has the solution

$$V_m = \exp\left(-\frac{1-\sqrt{1-4F^2}}{8F}\right)$$

(15)

with $X_m=X_m(V_m)$ defined by relation (12).

The values of $X_m$ and $V_m$ are shown in figure 9 by means of crosses on the corresponding curves. Figure 9 shows that the free lamella rim is located in the steady domain $X_m<X^*$ if $F<0.25$. Obviously, in this case the surfactant flux $J$ enlarges the maximum size of the lamella in this domain since $X_m>1$ (figure 9). At $F=0.25$, $X_m$ reaches a maximum $(X_m)_{\text{max}}=1.213$ and $V_m$ a minimum $(V_m)_{\text{min}}=V=\exp(-1/2)=0.60653$; simultaneously, $X^*$ is equal to $(X_m)_{\text{max}}$, which means that the lamella rim becomes the
boundary of the steady uniform flow if $F=0.25$.

It is possible to show that $We$ decreases in the lamella with radial coordinate $X$, but $We$ cannot reach 1 if $F=0.25$ because equation (15) has no solution in this case. It means that $We>>1$ everywhere in the domain defined by $X<X_m$ and therefore a steady free rim (defined by $We=1$) cannot be located in the domain of steady uniform flow. What will be with free lamella rim at $F>0.25$ is not obvious for us, and the mechanism of the rim formation is unclear in this situation.

Comparing the theoretical results for $X_m$ with our experimental data, one may conclude that the observed increase of the lamella size in the case of DOS, $10\times cac$ up to $\beta_m=6.0$ (i.e. $X_m=\beta_m/\beta_m^\infty=6.0/5.0=1.20$) corresponds to a value of $F=0.25$ for which the theory predicts a lamella size $X_m=1.213$. Note that, as figure 9 shows, the increase of the lamella size up to $X_m=1.213$ is the maximum possible for steady uniform lamella flow.

The theory predicts no surfactant effect on the lamella size if $F<0.25$ (figure 9). Thus a low value of parameter $F$ ($F<0.25$) can be attributed for Silwett L77 solutions which form lamellas of same size as the water ones.

4.5. Quantification of the effects of the dynamic surface tension

The constitutive parameter $F=RT(D\times cac/\delta)\ r_m^\infty/(2\gamma_s v_s)$ can be independently estimated analysing data in figure 5 on the dynamic surface tensions measured with a Maximum Bubble Pressure method apparatus. With this method the mechanism of formation of the ‘dynamic surface tension’ is the same as in our experiment and results from the Bubble Pressure method apparatus. With this method the mechanism of formation of the ‘dynamic surface tension’ is the same as in our experiment and results from the Bubble Pressure method apparatus.

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4.6. Hole nucleation

Let us consider now the spontaneous hole nucleation in the light of our theoretical results. As a first approximation, we can assume the flow in the lamella as sequences of a number of steady flows. The first flow regime corresponds to the growth of the lamella up to its maximum size. The latest flow regime is characterised by a very weak liquid ejection, the existence of the lamella being made possible by the slowing down of the liquid rim retraction due to its high inertia. Massive rim plays the role of ‘lamella holder’. Correspondingly, the first and the latest flow regimes are characterised by different values of $F$ and as a consequence by different coordinates of the steady uniform flow, $r_*$.

In dimensional form, the critical coordinate $X_*=0.60653/F^{1/2}$ is written as

$$r_*=0.606\left(\frac{2\gamma_s v_s w}{\left(\left(\gamma_w-\gamma_s\right)/\alpha\right)_w}\right)^{1/2}.$$ (19)
At the initial stage of the lamella existence $\tau \in [0, 3]$ the flow velocity is estimated as $v \sim v_0 = 3.4$ m/s, and the equivalent water lamella radius $r_m^w = 6.75$ mm. In this case according to (19) the critical radius $r_*$ has the same order of magnitude or exceeds significantly the current maximum radius of the lamella $r_- r_m^w = 6.75$ mm – see table 1. It means that the flow in the lamella occurs in a domain where a steady uniform flow is possible, $We > 1$ and it continues up to the rim, where $We = 1$.

At the latest stage of the lamella existence ($\tau \in 5$), another hydrodynamic situation exists. Using our experimental estimations of the kinematic data for the flow at $\tau \in 5$ in [6] ($h \approx 25$ $\mu$m, $v \approx 1$ m/s), the continuity equation (3), and the Taylor formula for the lamella maximum size [8] $r_m^w = \rho q_\nu / 4 \pi \gamma_w$, we obtain $r_m^w \approx 0.5$ mm. The corresponding values of $r_m$ calculated with formula (19) are presented in table 1. Relatively small values of $r_m$ are obtained whereas the lamella size $r_l$ at $\tau \in 5$ remains large $r_l \approx r_m^w = 6.75$ mm (figure 5), which illustrates well the holder role of the rim. Therefore, we observe a large decrease of $r_m$ from the level $r_m > r_l$ at the early stage of the impact, down to the level $r_m < r_l$ at the latest one (figure 11). It means that a steady uniform flow cannot occur in the part of the lamella at $\tau \in 5$. We suppose that the transition of the flow to the unsteady or/and spatially non-uniform regime is the main reason of the hole formation.

![Fig. 11. Image of a lamella displaying the position $r_*$ of the limit between stable ($r_\ast < r_\ast$) and unstable ($r_\ast < r_\ast < r_l$) domains.](image)

However the precise mechanism is still unclear. The unsteady or/and non-uniform character of the flow is not a sufficient condition for hole formation. Experiments with different liquids or even with different drops of a same liquid show that the lamella rupture has a random character. In reality, hole nucleations in the liquid film result from a thickness instability in a region beyond $r_\ast$ where $We < 1$ and where Marangoni effects cannot restore the uniformity of the thickness.

5. CONCLUSION

Surfactant additives increase the size and the lifetime of the liquid lamella resulting from the impact of drop on a small target because of the fast decrease of the surface tension forces in the case of fast surfactants. They could hinder splashes that generate spider-like liquid structure at the end of the impact. Paradoxically, we could also observe spontaneous nucleation of expanding holes giving to the lamella a web-like structure. The observed holes formation is likely connected with the coexistence of stable and unstable domains in the lamella generated by surface tension gradients in the latest stage of the experiment. These results may be of great assistance for the choice of additives for the formulation of sprays.

6. REFERENCES