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# Drop encapsulation and bubble bursting in surfactant-laden flows in capillary channels

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We present a parametric study of the unsteady phenomena associated with the flow of elongated gas bubbles travelling through liquid-filled square capillaries under high Weber number conditions. These conditions induce the formation of an indentation at the back of the bubble that commonly gives way to a deep re-entrant liquid jet penetrating the bubble. Subsequent steps include pinch-off events in the penetrating liquid to generate one or multiple encapsulated drops which may coalesce, in conjunction with the bursting of the bubble-liquid interface by either the liquid jet or the drops. Some of these interfacial instabilities have previously been reported experimentally [65] and numerically [43] for liquid-liquid flow in microchannels. We carry out three-dimensional direct numerical simulations based on a hybrid interface-tracking/level-set method capable of accounting for the presence and dynamic exchange of surfactants between the liquid bulk phase and the liquid-gas interface. Our results indicate that the delicate interplay amongst inertia, capillarity, viscosity, surfactant adsorption/desorption kinetics, and Marangoni stresses has a dramatic influence over the non-axisymmetric morphological structures of the encapsulated drops-elongated bubble. This strong coupling also influences the pinch-off time, penetration depth of the liquid, and number, size, and velocity of the encapsulated drops across the bubble. The observed phenomena are summarised in three main morphological regimes based on surfactant-related parameters and dimensionless groups. A discussion of the flow regime maps is also provided.

PACS numbers:

## I. INTRODUCTION

The problem of gas bubbles propagating in confined capillary channels has been the subject of extensive theoretical, experimental, and numerical investigation (see Etminan et al. [31] and references therein). In addition to being a classic problem in fluid mechanics, as seen in the now-seminal works of Bretherton [13], Taylor [82], and Schwartz et al. [73], these flows are central to multiple applications in technology development and are found in several real-life systems. To name a few examples, gas-liquid interfaces in the micro- and capillary scale are involved in two-phase coolers [48], CO<sub>2</sub> sequestration and storage microdevices [74], obstructed pulmonary airways [41], and volcanic conduits [14]. Since the early works mentioned above, extensive efforts have been undertaken to understand the phenomena dictating the bubble's behaviour along with the surrounding thin liquid film, largely through an idealisation of the operating conditions and the morphology of the bubble itself as a flat cylinder bounded by two semi-spherical menisci. It is well-known by experiments [82] and theory [13] that the film thickness of the flat region scales as  $h \sim Ca_b^{2/3}$  within the realms of vanishing capillary,  $Ca_b$ , and Reynolds numbers,  $Re_b$ , and as  $h \sim Ca_b^{2/3}/(1 + Ca_b^{2/3})$  for  $Ca_b < 2$  [9, 59]. As inertial effects become non-negligible, Han and Shikazono [39] proposed a correction to the scaling exponents of  $Ca_b$  and the introduction of the Weber number,  $We_b$ , in the scaling law expression. Particular attention has also been placed on exploring the influence of  $Ca_b$  on the pressure drop ahead of the bubble [88], mapping the full bubble topological profiles [25], analysing the undulatory structures that emerge in the vicinity of the bubble rear [49, 56] as inertial effects begin to dominate, and predicting the final shape of the front meniscus [34].

Other researchers have delved into the problem by deviating from the traditional idealised assumptions of circular and straight channels, absence of surface-active materials, fully dominating capillary forces, and rigid spherical interfaces. These studies have elucidated critical differences between the steady-state and axisymmetrical features of idealised bubbles and those that adhere to more realistic scenarios. For square or rectangular channel cross sections, the experiments of Chen et al. [17, 18] and numerical simulations of Magnini and Matar [55] and Magnini et al. [58] have uncovered significant angular and axial non-uniformities in the liquid film thickness, whereby the effects of the rigid walls tend to flatten the interface at the channel's centre-line ( $h_{min} \sim Ca_b$ ), depleting it of the liquid phase, and inducing the formation of large lobes protruding towards the channel corners in the cross-section ( $h \sim Ca_b^{0.445}$ ). Furthermore, non-circular channels promote an exponential thinning of the liquid film along the bubble length from the nose to the rear section, as reported in Lister et al. [53], Magnini and Matar [55]. The effects of other non-ideal geometries have been examined by Sauzade and Cubaud [72], who carried out an experimental campaign to study the distortion of a train of bubbles in a constricting-expanding microchannel under varying  $Ca_b$  and bubble packing conditions. This study provided evidence for the occurrence of bubble width-to-length ratio hysteresis across the sinusoidal

structures of the channel, and the attenuation of these effects with decreasing  $Ca_b$ .

A wide array of investigations have focused on illustrating the effects of (in)soluble surfactants on liquid-gas confined flows, placing particular emphasis on their influence on liquid film thickness, bubble deformation, and pressure drop (see Olgac and Muradoglu [67] and references). The literature largely agrees that, under typical surfactant characteristics and negligible inertia, the liquid film displays a thickening response to surfactants of the order of 1 to  $4^{2/3}$  times that predicted by Bretherton [13]’s expression for uncontaminated interfaces. This thickening has been reported to vary with  $Ca_b$ , surface elasticity, and across the bubble length due to Marangoni-related mechanisms, which act in opposition to liquid drag forces as they push the surfactants towards the bubble back. Nonetheless, Ghadiali and Gaver [33]’s numerical results in the context of pulmonary airways suggest the possibility of liquid film thinning surfactant actions under conditions of large bulk Péclet number,  $Pe_c \gg 1$ , and  $Ca_b > 10^{-1}$  due to a bifurcation and enhancement of Marangoni stresses, which drive fluid away from the thin film region and towards the bubble nose. In terms of pressure drop and bubble velocity, it has been shown that surfactants tend to increase the pressure drop across the dispersed phase [35, 54] and significantly reduce the bubble’s mobility along its length [12]. Batchvarov et al. [10] carried out a comprehensive computational exploration of multiple surfactant characteristics and dimensionless groups with the novelty of introducing inertial effects ( $Re = 443 - 728$ ) to the analysis. A key finding of this work is the surfactant-induced dampening of the bubble’s rear interfacial oscillatory structures, typical of systems with non-negligible inertia, together with the evidence provided to affirm that this process is entirely Marangoni-driven rather than a consequence of localised lowered surface tension. Moreover, this investigation addressed the effect of low solubility surfactants, detecting a clear arrangement of the bubble’s morphology into two separate regions; one fully covered in surfactants and of reduced mobility, extending from the bubble rear to a portion in the stream-wise direction that depends on  $Pe_c$ , and one of diminished surfactant concentration that stretches to the bubble nose, behaving similarly to clean interfaces.

A noteworthy sequence of events transpires in the bubble as  $Ca_b$  is increased to a critical value that strongly depends on its length and  $Re$ . As reported in a number of investigations [1, 34, 71, 81], [the combination of reduced dominance of capillary forces \(given by larger  \$Ca\$ \) and heightened inertia \(given by larger  \$Re\$ , leading to larger  \$We = CaRe\$ \)](#) has a distorting impact on the curvature of the front and back menisci, dragging both interfaces towards the main flow direction and causing the bubble to adopt an elongated, ‘bullet-like’ shape. [With the continued increase in inertial and viscous forces, the bubble’s rear undergoes a curvature inversion, enabling the liquid phase to infiltrate the bubble’s domain and form a small liquid jet.](#) The threshold conditions that induce this curvature inversion phenomenon have been reported to be  $We \approx 20$  for  $Re \sim O(10^1)$  in circular channels by Giavedoni and Saita [34] and  $Ca \gtrsim 1$  for  $Re \rightarrow 0$  in circular/square channels by Refs. [59, 71, 81]. As briefly shown in §III A, our surfactant-free simulations allow us to place the threshold within the range  $17 < We < 20$  for  $Re \sim O(10^2)$  in square capillaries.

The fate of the [liquid infiltrating the bubble](#) is highly influenced by a complex interplay of phenomena that might culminate in a continuous growth in the main axial or radial direction, or in its capillary breakup to encapsulate and entrap liquid drops within the bubble (see figure 1). This liquid infiltration process shares qualitative similarities with other systems relevant to engineering applications, such as sheet cavitation. In this type of cavitation, the adverse pressure gradient induces the development and spreading of a re-entrant liquid jet in the upstream direction underneath the cavity sheet, separating it from the solid surface [68].

The type of encapsulation described above has been observed experimentally in the works of Goldsmith and Mason [36] and Olbricht and Kung [66] in liquid-liquid flows. Similar findings are reported in a limited number of numerical pursuits on multiphase confined flows, including the investigations of Tsai and Miksis [83] and Izbassarov and Muradoglu [43] for contracting/expanding capillaries in Newtonian and viscoelastic fluids, respectively, Pozrikidis [69] for buoyancy-driven flows of viscous drops, Nath et al. [63] for liquid drops in creeping flow, and Andredaki et al. [5] and Atasi et al. [8] for gas bubbles in the absence/presence of surfactants, respectively. The latter three studies have provided an examination of the specific conditions that induce the encapsulation phenomenon, a general elucidation of the mechanisms at play, and an overview of the encapsulation aftermath. Nath et al. [63] have remarked that, in the inertialess limit and considering fully circular channels, there exists a critical  $Ca_b$  beyond which drop breakup and entrapment events will occur. The authors found this critical value to decrease (increase) with the initial drop-to-channel radius (dispersed-to-continuous phase viscosity). Andredaki et al. [5] and Atasi et al. [8] have recently laid the groundwork for the formulation of general encapsulation and breakup regime maps for  $Re \ll 1$ , bounded by  $We_B$  and the relationship between inertia and pressure drop across the channel in the former, and  $Ca_b$  and the relative importance of interface surfactant adsorption/desorption in the latter case.

Despite these initial efforts, much remains unknown and unexplored about these encapsulation events. Notably, a quantitative and phenomenological description of the underlying dynamics that govern each step of the process is needed, in combination with a thorough inspection of the re-entrant liquid jet characteristics. It is also crucial to address the multiple examples of interfacial singularities inherent to the system at hand, which have yet to be analysed from the perspective of the well-established problem of capillary breakup, from which multiple parallels naturally emerge between our system and others (e.g., contracting liquid ligaments and inkjet printing). Likewise, an analysis of the evolution of the drops beyond pinch-off is required as it is a topic much less understood than the steps that precede it [60] and could be crucial to potential applications [43]. Finally, and as will be seen in this paper, extending the set of surfactant parameters explored from those of Atasi et al. [8] uncovers new encapsulation and breakup outcomes, which allows us to expand and enhance the original regime maps. The main objective of this paper is therefore to carry out an extensive characterisation of the re-entrant liquid jet formation and (post) pinch-off dynamics, taking

into account the interaction between surfactants and inertia, capillarity and viscosity in a non-circular channel geometry, which correspond to non-idealities of interest, as seen in the foregoing literature review.

The rest of the paper is organised as follows: §II introduces our problem formulation, computational setup, and numerical approach. A discussion of our results is presented in §III, where we focus first on encapsulation in clean interfaces followed by an examination of surfactant-laden systems. Finally, concluding remarks are provided in §IV.

## II. NUMERICAL METHODS AND PROBLEM FORMULATION

### A. Numerical modelling and non-dimensionalisation

We consider a horizontal liquid-filled square capillary of width and height  $D$  in a Cartesian three-dimensional domain,  $\mathbf{x} = (x, y, z)$ , with an elongated gas bubble propagating through its interior (see figure 1). Assuming incompressible flow, Newtonian fluids, and negligible gravitational effects, we perform direct numerical simulations (DNS) based on the two-phase Navier-Stokes equations and a hybrid front-tracking/level-set method to handle the interface and surface tension forces. This method, as formulated and implemented in Shin and Juric [75], is coupled with the resolution of surfactant transport and exchange between the interface and the liquid phase bulk, as described in Shin et al. [78]. The relevant variables involved in the system have been rendered dimensionless (denoted by tildes) by using the scalings depicted in Eq. (1),

$$\tilde{\mathbf{x}} = \frac{\mathbf{x}}{D}, \quad \tilde{\mathbf{u}} = \frac{\mathbf{u}}{U_a}, \quad \tilde{t} = \frac{t}{D/U_a}, \quad \tilde{p} = \frac{p}{\rho_l U_a^2}, \quad \tilde{\sigma} = \frac{\sigma}{\sigma_s}, \quad \tilde{\Gamma} = \frac{\Gamma}{\Gamma_\infty}, \quad \tilde{C} = \frac{C}{C_\infty}, \quad \tilde{C}_s = \frac{C_s}{C_\infty}, \quad (1)$$

where  $\mathbf{u}$ ,  $t$ ,  $p$ ,  $\sigma$ ,  $\Gamma$ ,  $C$ , and  $C_s$  represent velocity, time, pressure, surface tension, interfacial surfactant concentration, bulk surfactant concentration, and bulk surfactant concentration in the vicinity of the interface, respectively. The physical parameters included in the scaling correspond to the width of the channel,  $D$ , the average inlet velocity of the liquid,  $U_a$ , the density of the liquid phase,  $\rho_l$ , the surface tension in a surfactant-free interface,  $\sigma_s$ , the saturation interfacial concentration,  $\Gamma_\infty$ , and the initial bulk surfactant concentration,  $C_\infty$ , in line with the scaling proposed by Batchvarov et al. [10] for a similar system. In what follows, we refer to each variable by its name to refer to its dimensionless version, unless stated otherwise and add the subscript  $b$  to signify that the variable is reported at the bubble nose.

The mass and momentum equations are written in dimensionless form and according to a ‘one-fluid’ formulation:

$$\nabla \cdot \tilde{\mathbf{u}} = 0, \quad \tilde{\rho} \left( \frac{\partial \tilde{\mathbf{u}}}{\partial \tilde{t}} + \tilde{\mathbf{u}} \cdot \nabla \tilde{\mathbf{u}} \right) = -\nabla \tilde{p} + \frac{1}{Re} \nabla \cdot [\tilde{\mu} (\nabla \tilde{\mathbf{u}} + \nabla \tilde{\mathbf{u}}^T)] + \frac{1}{ReCa} \int_{\tilde{A}(\tilde{t})} (\tilde{\sigma} \tilde{\mathbf{k}}\mathbf{n} + \nabla_s \tilde{\sigma}) \delta(\tilde{\mathbf{x}} - \tilde{\mathbf{x}}_f) d\tilde{A}, \quad (2)$$

$$\begin{aligned} \tilde{\rho}(\mathbf{x}, t) &= \rho_g + (\rho_l - \rho_g) \mathcal{S}(\mathbf{x}, t), \\ \tilde{\mu}(\mathbf{x}, t) &= \mu_g + (\mu_l - \mu_g) \mathcal{S}(\mathbf{x}, t), \end{aligned} \quad (3)$$

where  $\mathcal{S}(\mathbf{x}, t)$  is a smoothed Heaviside function that adopts the value of zero in the gas phase and unity in the liquid phase,  $\rho$  is the density and  $\mu$  the viscosity of the fluids. Here, we denote the liquid and gas phases with the subscripts  $l$  and  $g$ , respectively. The normal and tangential components of the surface tension forces are represented by the last two terms on the right-hand-side (RHS) of Eq. (2), wherein  $\tilde{\mathbf{k}}$  corresponds to twice the mean interface curvature,  $\mathbf{n}$  to a unit normal to the interface,  $\delta(\tilde{\mathbf{x}} - \tilde{\mathbf{x}}_f)$  to a Dirac delta function that is zero everywhere except for the interface (located at  $\tilde{\mathbf{x}} = \tilde{\mathbf{x}}_f$ ),  $\tilde{A}(\tilde{t})$  to the time-dependent interface area, and  $\nabla_s$  to the surface gradient operator [76, 77]. We adopt the convention that positive  $\mathbf{n}$  vectors point outwards from the interface towards the liquid phase. Accordingly, positive values of  $\tilde{\mathbf{k}}$  describe a convex interface, as exemplified in figure 1. **As is typical in front-tracking, the Lagrangian interface is advected by integrating  $d\mathbf{x}_f/dt = \mathbf{V}_f$ , where the vertices of the Lagrangian triangular mesh discretising the interface,  $\mathbf{x}_f$ , are displaced in time by the fluid velocity,  $\mathbf{V}_f$ , at those vertices.**

Note that the gas phase dynamics are not neglected within this one-fluid formulation and are resolved along with that of the liquid via solution of Eqs. (2)-(3) with the density and viscosity defined by Eqs.(3). Consequently, the tangential component of the interfacial stress is finite with further contributions to it arising from the surface tension gradients; the latter owe their existence to the interfacial surfactant concentration gradients, as will be discussed below.

The mass conservation equations of surfactant species at the interface and bulk are given by Eq. (4)-(5) and the source term representing surfactant exchange between the interface and the bulk region immediately adjacent is given by Eq. (6):

$$\frac{\partial \tilde{\Gamma}}{\partial \tilde{t}} + \nabla_s \cdot (\tilde{\Gamma} \tilde{\mathbf{u}}_t) = \frac{1}{Pe_s} \nabla_s^2 \tilde{\Gamma} + Bi (k \tilde{C}_s (1 - \tilde{\Gamma}) - \tilde{\Gamma}), \quad (4)$$

$$\frac{\partial \tilde{C}}{\partial \tilde{t}} + \tilde{\mathbf{u}} \cdot \nabla \tilde{C} = \frac{1}{Pe_c} \nabla \cdot (\nabla \tilde{C}), \quad (5)$$

$$\mathbf{n} \cdot \nabla \tilde{C}|_{interface} = -Pe_c Da Bi (k \tilde{C}_s (1 - \tilde{\Gamma}) - \tilde{\Gamma}), \quad (6)$$

where  $\tilde{\mathbf{u}}_r = (\tilde{\mathbf{u}}_s \cdot \mathbf{t})\mathbf{t}$  is the projection of the interface velocity vector,  $\tilde{\mathbf{u}}_s$ , on the interface unit tangent,  $\mathbf{t}$ . The dependence of surface tension on local interface surfactant concentration is represented by a non-linear equation of state derived from Langmuir adsorption isotherm, as expressed in Eq. (7)

$$\tilde{\sigma} = \max(\varepsilon_\sigma, 1 + \beta_s \ln(1 - \tilde{\Gamma})), \quad (7)$$

where  $\beta_s = \mathcal{R}T\Gamma_\infty/\sigma_s$  is the surfactant elasticity number;  $\mathcal{R}$  and  $T$  are the thermodynamic ideal gas constant and temperature, respectively. The non-linear equation of state described above is valid for very dilute systems in which  $\Gamma \ll \Gamma_\infty$ . As  $\Gamma$  increases, the equation of state yields unphysical negative values of surface tension. To circumvent this, a limiting value for  $\tilde{\sigma}$ ,  $\varepsilon_\sigma = 0.05$ , has been introduced, in accordance with Muradoglu and Tryggvason [62] and Shin et al. [78]. **The tangential surface stresses or Marangoni stresses, heavily discussed throughout the manuscript, are calculated as  $\tilde{\tau}_m \equiv \nabla_s \tilde{\sigma} \cdot \mathbf{t} = -\beta_s \frac{1}{1-\tilde{\Gamma}} \nabla_s \tilde{\Gamma} \cdot \mathbf{t}$  (see last term of Eq. (2)).** The dimensionless groups that appear in the above equations and that characterise the system are defined as follows:

$$Re = \frac{\rho_l U_a D}{\mu_l}; Ca = \frac{\mu_l U_a}{\sigma_s}; Pe_c = \frac{U_a D}{D_c}; Pe_s = \frac{U_a D}{D_s}; Bi = \frac{k_d D}{U_a}; Da = \frac{\Gamma_\infty}{DC_\infty}; k = \frac{k_a C_\infty}{k_d}, \quad (8)$$

where  $Re$  and  $Ca$  are the Reynolds and Capillary numbers, respectively ( $We = CaRe$ , and  $Oh = \sqrt{Ca/Re}$ ).  $Pe_c$  and  $Pe_s$  are the bulk and interfacial Péclet numbers, which provide a measure of the importance of inertia relative to surfactant mass diffusion across the interface (modulated by a diffusivity  $D_c$ ) or the bulk (modulated by a diffusivity  $D_s$ ), respectively;  $Bi$  represents the Biot number, describing the competition between convective,  $t_{conv} = D/U_a$ , and interface surfactant desorption,  $t_{des} = k_d^{-1}$ , time scales. The Damköhler number,  $Da$ , provides a dimensionless measure of the initial interface saturation, and the number  $k$  reflects the interplay between the characteristic time scales of interface surfactant adsorption,  $t_{ad} = (k_a C_\infty)^{-1}$ , and desorption. **We note that gravitational effects have not been taken into account in this study. Hence, we run our simulations with vanishing Bond number,  $Bo \equiv (\rho_l - \rho_g)gD^2/\sigma_s = 0$ .**

For an exhaustive description of the discretisation schemes and numerical framework employed for solving the above mentioned governing equations, we refer the reader to Shin et al. [77, 78], and to Shin and Juric [76] for a comprehensive account of the hybrid front-tracking/level-set method, also called Level Contour Reconstruction Method (LCRM). Here, we present a short summary of the most relevant aspects. The LCRM considers the combination of a fixed structured Eulerian grid for the resolution of field equations and a moving and deforming Lagrangian grid for the interface, discretised via an unstructured triangular mesh. These interface elements are consequently advected through the integration of the Lagrangian equation  $d\mathbf{x}_f/dt = \mathbf{v}$ , where  $\mathbf{v}$  corresponds to the interface velocity, which is interpolated from the Eulerian grid. This integration is carried out with a second-order Runge-Kutta method. The spatial derivatives of the fields in the Eulerian grid are discretised through a standard cell-centred scheme for all terms, with the exception of the non-linear convective terms, for which a second-order essentially non-oscillatory (ENO) procedure is used [79, 80]. For the viscous term in the momentum equation, a second-order centred difference scheme is employed. A second-order Gear method [86] is adopted for the temporal terms, with an implicit time integration for the viscous terms. The time step is set to be adaptive, according to the following criterion:

$\Delta t = \min\{\Delta t_{cap} = \frac{1}{2} \sqrt{\frac{(\rho_l + \rho_g)\Delta x^3}{\pi\sigma_s}}, \Delta t_{vis} = \frac{\rho_g \Delta x^2}{6\mu_l}, \Delta t_{CFL} = \frac{\Delta x}{|u_{max}|}, \Delta t_{int} = \frac{\Delta x}{|V_{int}|}\}$ , where the time steps relate to viscosity, capillarity, Courant-Friedrichs-Lewy condition, and interface, respectively;  $\Delta x$  is the minimum cell size,  $u_{max}$  is the maximum fluid velocity, and  $V_{int}$  is the maximum interface velocity. The time-step related to the interface was found to be the limiting time-step in all of our simulations ( $\Delta t_{int} \sim O(10^{-7})$ s).

## B. Simulation setup

As depicted in figure 1, our simulation domain consists of a three-dimensional rectangular box of dimensions  $27D \times D \times D$ , corresponding to the  $x$  (channel length),  $y$  (channel height), and  $z$  (channel width)-directions, respectively. The value of the channel length was selected to ensure the attainment of a steady-state bubble propagation for  $We < 10$ , following the results of Batchvarov et al. [10] and Magnini and Matar [55]. Under conditions of  $We > 10$ , highly unsteady encapsulation/bursting phenomena are detected in the system. The channel length chosen allowed for the observation of all relevant phenomena, including the formation of the re-entrant jet, drop encapsulations and coalescence events, and bubble bursting, for all conditions

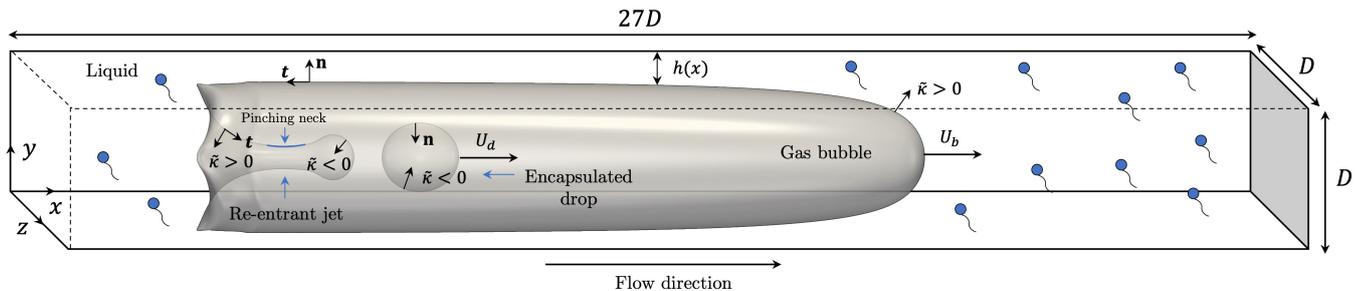


FIG. 1: (not to scale) Schematic of simulation setup, re-entrant liquid jet penetrating the bubble, and encapsulated drop. The direction of outwards pointing normal vectors, liquid film thickness,  $h(x)$ , and curvature sign according to the adopted convention, are also highlighted. The location of the Cartesian axis represents the outset of the geometrical domain ( $x = 0, y = 0, z = 0$ )

tested. The elongated bubble was initialised in quiescent conditions close to the channel inlet and at the centre-line of the  $y$  and  $z$ -directions as a horizontal cylinder of length  $3.12D$  with two hemispherical caps of diameter  $0.94D$  on each end. A fully-developed liquid velocity profile was imposed at the channel inlet with a Neumann condition for pressure ( $\partial p / \partial \mathbf{n} = 0$ ). All walls were treated as no-slip boundaries, and Neumann conditions were imposed for all variables at the outlet. The bubble-liquid interface was initially covered with surfactant of uniform concentration  $\Gamma_0$ , determined from an initial equilibrium between surfactant adsorption and desorption (last term on the RHS of Eq. (4) equal to zero), which, assuming  $C_s = C_\infty$  initially, reads:

$$\Gamma_0 = \frac{k}{k+1} \Gamma_\infty. \quad (9)$$

This set-up closely resembles the approaches followed in previous numerical investigations for similar systems [8, 54, 63].

We explore the effect of the dimensionless numbers and flow parameters that characterise the system (see Eq. (8)) considering the following ranges:  $Ca = 0.0089 - 0.0693$  ( $We = 3.94 - 30.70$ ,  $Oh = 4.48 \times 10^{-3} - 1.25 \times 10^{-2}$ ),  $Pe_{c,s} = 100$ ,  $Bi = 0.10 - 1$ ,  $Da = 0.01 - 1$ ,  $k = 0.10 - 10.00$ , and  $\beta_s = 0.25 - 1$ . For all cases simulated, the viscosity and density ratio between the phases is kept constant and in line with representative values for water and air:  $\rho_l / \rho_g = 1000$  and  $\mu_l / \mu_g = 100$ . We define a ‘base’ case from which our parametric sweep was carried out with the following conditions:  $Re = 443$ ,  $Ca = 0.0693$ ,  $Pe_{c,s} = 100$ ,  $Bi = 0.10$ ,  $Da = 1$ ,  $k = 0.10$ , and  $\beta_s = 0.50$ . Unless stated otherwise, all results correspond to simulations under these conditions. The selection of the testing ranges was based on common values encountered in bubbly flow systems in the presence of surfactants, as reported in Atasi et al. [8], as well as values of  $We$  that would allow a significant disruption of the bubble back to generate the aforementioned penetrating liquid and consequent drop encapsulation [5, 34]. We refer the reader to Appendix A for typical values of surfactant properties, which support our choice of dimensionless ranges, and for a description of the competing timescales in our system.

Following numerous studies making use of the numerical methods described above for inertial and capillary phenomena (see Refs. [10, 19, 21]), we employ a fully-structured and uniform Cartesian grid divided into  $54 \times 4 \times 4$  sub-domains, further divided into  $64 \times 32 \times 32$  cells per sub-domain, rendering a global resolution equal to  $3456 \times 128 \times 128$  cells. On account of ensuring this resolution produces mesh-independent results, we have performed a mesh analysis in surfactant-free and surfactant-laden conditions, detailed in Appendix B along with other resolution considerations. **In this analysis, we showcase how the chosen grid resolution guarantees mesh-independent outcomes for both global variables like kinetic energy, interfacial area, and bubble velocity, and local variables associated with interfacial singularities. These encompass the curvature inversion time, the time until the first pinch-off event of the re-entrant jet, and the size of the encapsulated drops, all of which show less than 3.5% difference between mesh resolutions. Furthermore, we illustrate that our resolution is adept at accurately representing the thin liquid film encasing the bubble, as outlined in Ref. [55].**

The numerical framework and simulation setup presented herein have previously been carefully validated in the context of elongated bubbles in circular capillary channels. For this validation, we refer to the work of Batchvarov et al. [10], in which the code was benchmarked against the well-known correlation of Han and Shikazono [39] for steady-state liquid film thickness in conditions of non-negligible inertia. For  $Ca \sim O(10^{-2})$  and  $Re \sim O(10^2)$ , deviations of up to 10% were reported, which are within the uncertainty of the correlation. A complementary validation is reported in Appendix C to demonstrate our faithful capturing of the bubble’s main morphological regimes in square capillary channels, as proposed by Magnini and Matar [55].

To attach a physically-relevant meaning to the dimensionless times quoted throughout the manuscript, we henceforth report all times starting from  $\tilde{t} = 0.0$  which corresponds to the instance of the bubble rear’s curvature inversion (mentioned in §I and detailed in figure 3) for each set of conditions. For the surfactant-free and low  $Ca$  cases that do not exhibit a curvature inversion (see figure 2),  $\tilde{t} = 0.0$  was taken as the inversion time of the surfactant-free case that did undergo such process (see §III A).

### III. RESULTS AND DISCUSSION

The results of our investigation are presented in three parts. The first, §III A, introduces the phenomenon of drop encapsulation by contrasting the behaviour of three surfactant-free cases of varying  $Ca$  and exposed to non-negligible inertial effects ( $Re = 443$ , figure 2). The second part of the results, §III B, analyses selected cases from our surfactant-related parameter sweep ( $\beta_s$ ,  $Bi$ ,  $k$ , and  $Da$ ) and quantifies their influence on drop encapsulation. The final set of results, §III C, summarises all encapsulation behaviours observed in two regime maps in the  $\beta_s - k$  and  $Bi - k$  spaces (figure 11).

#### A. Bubble dynamics in surfactant-free cases: tail undulations, curvature disruptions, and encapsulations

We start our discussion by showcasing the bubble topology at a two-dimensional projection in the  $(x,y)$  plane at the channel centre-line ( $\tilde{z} = 0.5$ ), as well as the normalised mean curvature,  $\tilde{\kappa}$ , and the normal,  $\tilde{\tau}_n$ , and tangential,  $\tilde{\tau}_t$ , components of the viscous stress along the interface (see figures 2(a)-(b)). Here, these quantities are defined as  $\tilde{\kappa} = \kappa/\kappa_c$ ,  $\tilde{\tau}_n = \tau_n/(D/U_a)$ , and  $\tilde{\tau}_t = \tau_t/(D/U_a)$ , respectively, where  $\tau_n = (\mathbf{D} \cdot \mathbf{n}) \cdot \mathbf{n}$ ,  $\tau_t = (\mathbf{D} \cdot \mathbf{n}) \cdot \mathbf{t}$ ,  $\mathbf{D} = (\nabla \mathbf{u} + \nabla \mathbf{u}^T)/2$  (rate of deformation tensor), and  $\kappa_c$  corresponds to the curvature of a sphere with a volume equal to the initial bubble volume. Under the sign conventions adopted,  $\tilde{\tau}_n > 0$  denotes stresses exerted on the interface towards the liquid phase. Conforming to previous observations [1, 34, 71, 81], our plots reveal a loss of sphericity (typical of fully dominant capillary forces) at the bubble front and back with increasing  $Ca$ , in conjunction with an overall axial bubble elongation. The interface at the bubble front sharpens and expands, whilst the bubble back becomes progressively flattened with  $Ca$ , adopting a ‘bullet-like’ shape ( $\tilde{\kappa}_{front} > \tilde{\kappa}_{back}$ ). This process continues until a curvature inversion emerging from  $\tilde{y} = 0.5$  is detected for  $Ca = 0.0693$ , which eventually leads to a drop encapsulation. As mentioned in §I, this type of curvature sign inversion is triggered by a combination of large  $Ca$  and  $Re$ . From the surfactant-free results shown in figure 2(a) at  $Re = 443$ , as well as those reported in Appendix C, which encompass various  $Ca$  and  $Re$  values ( $We = 2.5 - 50$ ), we place the threshold value for curvature inversion in the range  $17 < We < 20$ . This range is fully consistent with that previously found in Giavedoni and Saita [34] ( $We \approx 20$  for  $Re \sim O(10^1)$ ) in circular channels.

We note that the bubble shapes presented in figure 2(a) correspond to the systems’ initial stages of development in terms of the re-entrant jet and the front/rear menisci ( $\tilde{r} = 0.76$ ). During these early stages, the systems are still distant from equilibrium, which is typically achieved at  $\tilde{r} \approx 6$  for the non-encapsulated cases  $Ca = 0.0089, 0.0377$  (and notably not attained for the encapsulated case, as demonstrated in both our present study and prior research, see §I). Consequently, the well-established direct relationship between liquid film thickness,  $h(x)$ , and  $Ca$  [13, 39, 55] has not materialised fully in the systems at this early stage, explaining the apparent absence of differences in  $h(x)$  for varying  $Ca$ . A further discussion of the effects of  $Ca$  on  $h(x)$  is provided in Appendix C, where the cross-section bubble shapes of the systems depicted in figure 2(a) are displayed and analysed at a later time (see figure 13).

Detailing the behaviour of  $\tilde{\kappa}$  for  $Ca = 0.0693$  along the interface path going from the re-entrant liquid jet vertex to the bubble nose, a zone of negative curvature is first observed at  $3.8 \lesssim \tilde{x} \lesssim 4.1$ , followed by a positive curvature meniscus at  $3.3 \lesssim \tilde{x} \lesssim 3.8$ . This particular curvature sign inversion is a manifestation of the early stages that precede pinch-off and drop encapsulation. This process, and its connection to other well-known systems that exhibit capillary instabilities, are examined further along in this paper. Exiting the interior of the liquid jet towards the region adjacent to the liquid film ( $3.3 \lesssim \tilde{x} \lesssim 7.0$ ), a non-uniform low curvature zone that encompasses the majority of the bubble is identified, later culminating in the high-curvature bubble nose seen for  $\tilde{x} > 7.0$ . Contrasting this evolution to that of the two lowest  $Ca$ , it can be noticed that the spatial location that marks the beginning of the bubble nose (sudden curvature rise from the thin-liquid film region, highlighted in gray in figure 2(a)) shifts to lower  $\tilde{x}$  for decreasing  $Ca$ . Notable curvature inversions at the bubble tail are also identified for the low  $Ca$  cases, expressed in the undulatory structures emphasized in figure 2(b). These interfacial waves, previously reported in Ref. [27, 34, 56], moderately diminish in frequency and amplitude with  $Ca$  at the same  $Re$ , as illustrated in the figure.

The dynamics of the bubble-liquid interface and its curvature gradients are readily mapped onto the  $\tilde{\tau}_{n,t}$  spatial profiles of figure 2. Pertaining to the trailing undulatory structures (highlighted in yellow and green for  $Ca = 0.0089$  and  $Ca = 0.0377$ , respectively), it is found that these are first characterised by a sudden surge in  $\tilde{\tau}_n$  that terminates in a local maximum, which pushes the interface towards the channel walls and thus creates the crests of the interfacial waves. These peaks are promptly followed by a rapid  $\tilde{\tau}_n$  decrease approaching a local minimum that contributes to the formation of the wave troughs by pulling the interface back towards the bubble. Concurrently, the tangential stresses feature similar inverse patterns of local maxima/minima that promote the sharpening of the crests/troughs. A substantial drop in  $\tilde{\tau}_{n,t}$ , mirrored by  $\tilde{\kappa}$ , is observed by departing from the rear undulatory region to the high-pressure central section of the bubble.  $\tilde{\tau}_{n,t}$  remain quasi-constant and close to zero throughout this section until the reappearance of local maxima/minima marks the onset of the bubble nose. Note the generally higher  $|\tilde{\tau}_t|$  exerted on the bubble nose at  $\tilde{y} = 0.5$  for decreasing  $Ca$ , partly contributing to a lower curvature (more spherical) bubble front. Similar remarks about the close relationship between curvature, capillarity, and interfacial stresses have been made by Atasi et al. [8].

We now proceed to detail the encapsulation process by first illustrating the numerous flow singularities produced by high  $Ca$  conditions in uncontaminated interfaces. In figure 3(a) we plot the temporal evolution of the maximum normalised velocity in

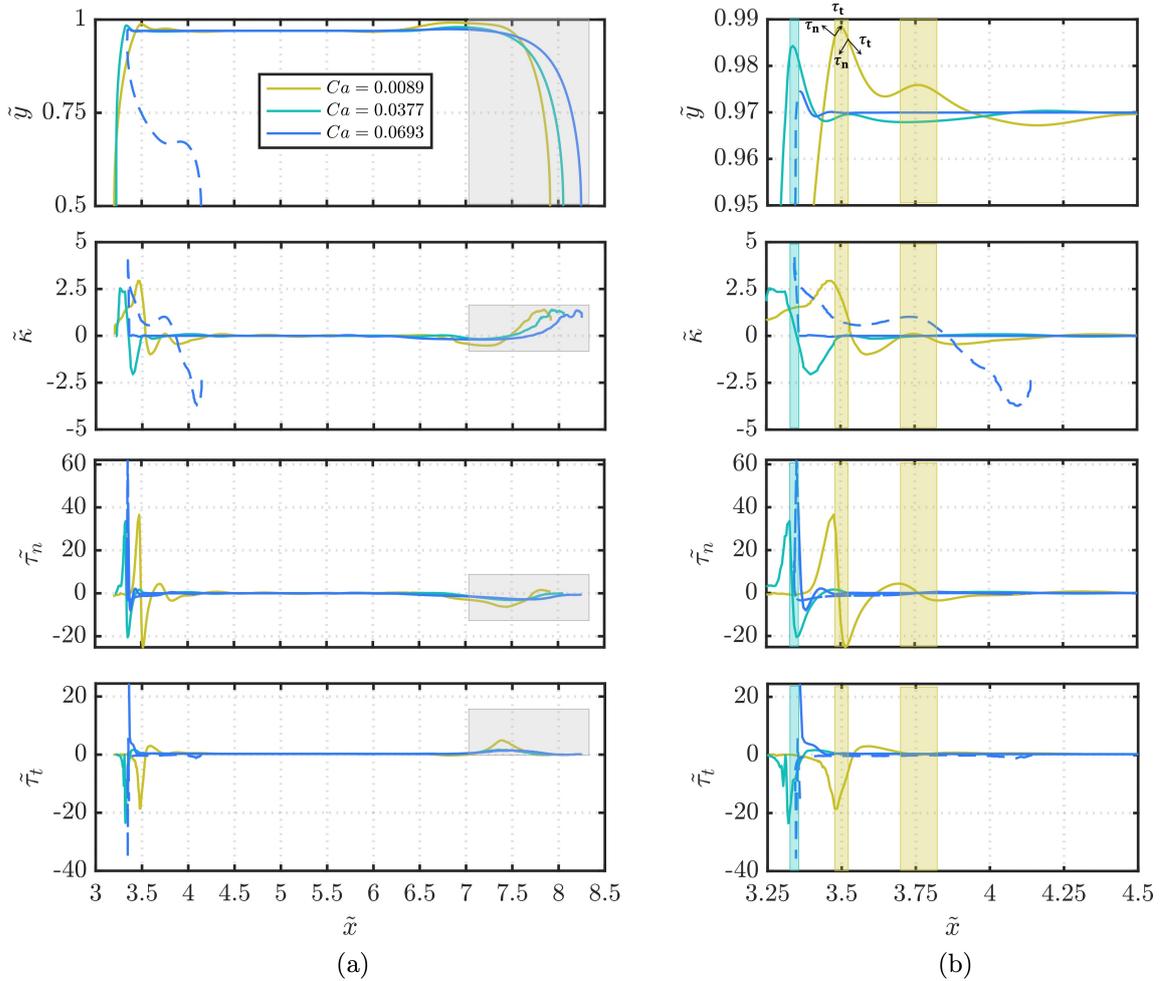


FIG. 2: Effect of  $Ca$  on bubble characteristics for surfactant-free cases at  $\tilde{t} = 0.76$ . (a) Full bubble length in the axial direction. (b) Zoom over the trailing undulatory structures at low  $Ca$  ( $3.25 < \tilde{x} < 4.5$ ). From top to bottom the plots correspond to bubble-liquid interface shape, normalised mean curvature,  $\tilde{\kappa}$ , normalised normal component of viscous stress on the interface,  $\tilde{\tau}_n$ , and normalised tangential component of viscous stress on the interface,  $\tilde{\tau}_t$ , respectively. Dotted line for  $Ca = 0.0693$  highlights the re-entrant liquid jet. All other parameters remain unchanged from those specified in §II B for the base case. The highlighted zones in (b) represent the areas bounded by  $\tilde{\tau}_n$  local maxima and minima their projections in the  $\tilde{y}$ ,  $\tilde{\kappa}$ , and  $\tilde{\tau}_t$  profiles

$x$ -direction,  $Max \tilde{u}_x$ . This evolution reveals five major local peaks in  $Ca = 0.0693$  in the interval  $0 < \tilde{t} < 3.5$ , which materialise in (i) the previously analysed curvature inversion at the bubble back, (ii)-(iii) two pinch-offs that give rise to a ‘satellite’ and a primary drop, (iv) an abrupt retraction and subsequent expansion of the former, and (v) a coalescence event between the two drops (see figure 3(b)). A first description of the mechanisms by which these interfacial singularities come about is shown in figure 3(c), where a liquid infiltration deepening in the positive  $x$ -direction takes place at  $0.32 < \tilde{t} < 0.99$ . The fate of this liquid structure can be understood through the hybrid lens of the well-known problems of liquid filament breakup surrounded by a gas phase [15, 29] and liquid dripping faucets [4]. Our liquid jet at  $\tilde{t} < 0.99$  can be thought of as a quasi-cylindrical liquid filament ( $Oh = 0.0125$ ) with a low ratio between its length,  $2L_{jet}$ , and radius,  $R_{jet}$  undergoing end-pinching.

At around  $\tilde{t} = 0.99$  in figure 3(c), we observe a halt in jet axial growth as capillary pressure builds at the bubble rear, inducing a vertical expansion towards the channel centre-line ( $\tilde{y} = 0.5$ ), creating an essentially flat bubble back at  $\tilde{t} = 1.87$  and a nascent pinching neck characterised by a high  $|\tilde{\omega}_z|$  ring. The interface areas covered by the ring are further pulled in the counter-flow direction, prompting an additional curvature inversion at the bubble rear and the first pinch-off thereupon (see  $\tilde{t} = 2.09$ ). This capillary singularity promotes a swift and opposite-direction recoil of the bubble back and the newly-entrapped liquid structure, thus forming the globoid-shaped bubble rear observed in  $\tilde{t} = 2.27 - 2.51$ . The rapidly forming rear pinching neck witnesses the incipience of an additional capillary neck at the interior of the bubble (see highlight and high  $\tilde{\omega}_z$  ring in  $\tilde{t} > 2.09$ ). This concludes in a second pinch-off and a precipitous recoil into a satellite drop (see  $\tilde{t} = 2.37$ ). Henceforth, we will refer to the first of these

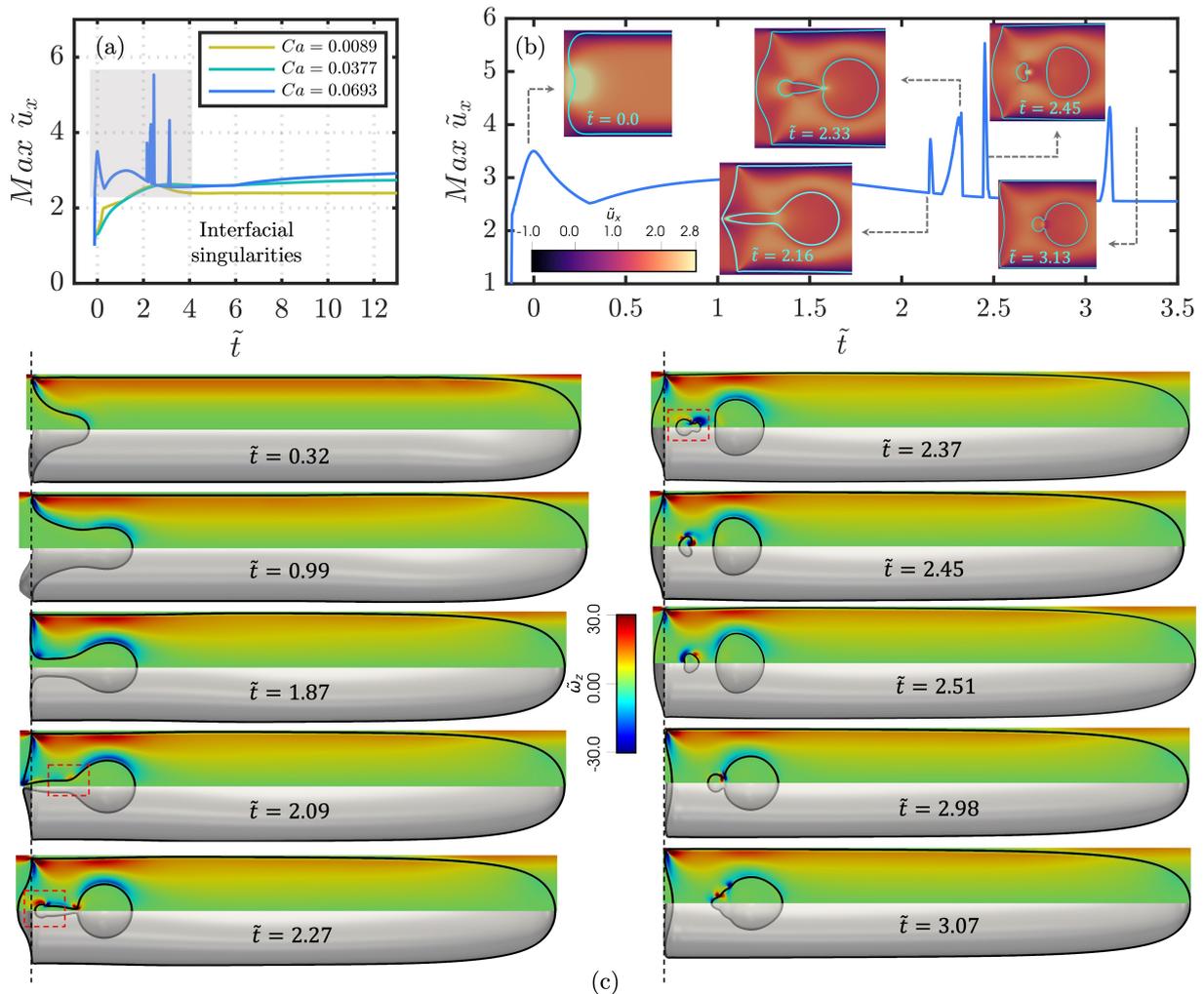


FIG. 3: Temporal evolution of interface characteristics in surfactant-free cases. (a) Maximum dimensionless velocity in  $x$ -direction. (b) Zoom on interfacial singularities found for  $Ca = 0.0693$ . (c) Detailed evolution of encapsulation process for  $Ca = 0.0693$ . Contour plots of normalised azimuthal vorticity,  $\tilde{\omega}_z = \omega_z/(D/U_a)$  (top) and three-dimensional bubble shape (bottom). The interfaces of these contour plots have been translated in the  $x$ -direction to have a common point of reference (marked by the black dotted line)

pinch-off events as ‘back pinch-off’, and to the second as ‘interior pinch-off’; the times for these events are denoted as  $\tilde{t}_{p-o,bk}$  and  $\tilde{t}_{p-o,int}$ , respectively. These phenomena of successive back and interior pinch-offs in surfactant-free cases are in agreement with previous numerical observations [5].

The post second pinch-off dynamics are driven by rich rotation mechanisms around both the satellite and primary drops. The two pinch-off events create a small contracting liquid filament, characterised by local  $Oh \sim 0.05$  and  $L_{ligament}/R_{ligament} \sim 2.62$  and similar to those studied previously [26, 64]. This small aspect ratio, in conjunction with the strong contracting kinetic energy derived from the second pinch-off, overcome capillary pressure forces to avoid an additional pinch-off instance in the small ligament. Observing the vorticity contours and the highlights at  $\tilde{t} = 2.27 - 2.37$ , an example of a vortex ring detachment can be noticed at the forming neck of the filament, where the vorticity boundary layer detaches and advects towards the centre of the bulbous region, pushing flow away from the neck and promoting the escape from pinch-off [19, 42, 84]. As seen in  $\tilde{t} = 2.45$ , succeeding the small liquid filament retraction in the counter-flow direction ( $\tilde{t} = 2.37$ ) we spot a counter expansion (see positive  $\tilde{\omega}_z$  at  $\tilde{t} = 2.45$ ) that generates the bullet-like satellite drop depicted at  $\tilde{t} = 2.51$  and that eventually coalesces with the primary drop. The process of incorporation of the satellite into the primary drop exhibits the progressive growth of the finite liquid bridge [30], driven by the two opposite-signed high  $|\tilde{\omega}_x|$  rings in its vicinity (see  $\tilde{t} = 2.98$ ) as well as complex dynamics of interfacial waves across the newly-formed drop (see  $\tilde{t} = 3.07$ ).

## B. Influence of surfactant parameters on re-entrant liquid jet formation and encapsulation dynamics

Having examined in detail the different encapsulation phenomena observed for high  $Ca$  in surfactant-free interfaces, we continue our analysis to explore the influence of key surfactant dimensionless groups. We begin by exploring the locality of the pinch-off in surfactant-free and surfactant-laden cases. We follow this by highlighting the influence of Marangoni stresses on the infiltration liquid shape, followed by describing the relative effect of  $\beta_s$  and  $Bi$ . Thereafter, we analyse the competition between the rates of interface surfactant adsorption and desorption, represented by  $k$  and  $Da$  simultaneously.

### 1. Focus on the pinch-off dynamics for surfactant-free and surfactant-laden cases

We examine in this subsection the locality of the re-entrant jet breakup in our simulations and contrast the neck thinning behaviour to that of the well-established theoretical scalings of Eggers [28] and Day et al. [24] for the minimum neck radius,  $r_{min}$ , and its evolution over time,  $t_p - t$ , where  $t_p$  is the breakup time. We focus on the first observed breakup singularity for the surfactant-free case, which corresponds to a ‘back pinch-off’ event, as defined in §III A and figure 3(a,c) at  $\tilde{t} = 0.99 - 2.16$ . Figure 4(a) contrasts the thinning progression of the pinching necks using the two mesh resolutions extensively tested in Appendix B (M1 and M2), along with an additional ‘medium’ mesh with global resolution  $M1.5 = 5376 \times 192 \times 192$ . Figure 4(b) depicts zoomed-in snapshots, colour-coded by  $\tilde{\omega}_z$ , of the liquid jet as it approaches the singularity.

An inspection of this figure provides further evidence of the nearly complete independence of our results on the mesh resolution from M1 and beyond, as well as the robustness of our numerical approach, which is able to correctly capture the system’s universal dynamics at the scale of the pinch-off in terms of both thinning rate and scaling exponents. Reassuringly, we observe an almost complete overlap of the curves for the three resolutions as  $r_{min}(t) \rightarrow 0$  ( $\tilde{t}_b - \tilde{t} \lesssim 4 \times 10^{-3}$ ), where the universal scaling for the inertial-viscous (IV) regime of Eggers [28] ( $r_{min} \sim (t_p - t)$ ) is correctly recovered. At early thinning stages ( $\tilde{t}_b - \tilde{t} \approx 2 \times 10^{-1} - 6 \times 10^{-2}$  in our systems), the competition between fluid inertia the opposing capillary forces yields the inertial-capillary regime (IC [24], dictated by  $r_{min} \sim (t_p - t)^{2/3}$  and highlighted in red) for the liquid thread, which later transits into intermediate regimes, as predicted in Castrejón-Pita et al. [16]. It is important to note that although our obtained solution approximates the IC regime, it does not precisely match the theoretical pre-exponent of  $\approx 0.7$  [29] or the  $2/3$  exponent proposed by Ref. [24]. For reference, the closest matches to the scaling were found to be  $\tilde{r}_{min} \approx 0.67(\tilde{t}_b - \tilde{t})^{2/3}$ ,  $\tilde{r}_{min} \approx 0.6(\tilde{t}_b - \tilde{t})^{0.62}$ , and  $\tilde{r}_{min} \approx 0.55(\tilde{t}_b - \tilde{t})^{0.56}$  for M1, M1.5, and M2, respectively. Moreover, it can be noticed that the intermediate stages in our plot appear to be moderately sensitive to the resolution.

The above mentioned deviations from the theoretical scalings can be attributed to the dissimilarities between our system and those those studied in Refs. [24, 28]. Our system consists of a three-dimensional confined capillary channel wherein inertial effects are non-negligible, the pinching thread grows axially due to the action of the main flow, and the pinching domain remains in motion across the channel. These intricate dynamics are not accounted for in the theoretical scalings, which were initially proposed in an axisymmetric context under free-flow conditions. It is these dissimilarities between our system and the theoretical framework that likely contribute to the observed discrepancy between our solution and the IC regime. As mentioned in our previous work [19, 21], the nature of our 3-D simulations makes it prohibitively difficult to capture the dynamic intermediate regimes reported in Castrejón-Pita et al. [16], a fact that likely explains the deviations found between mesh resolutions at intermediate times.

We further investigate the pinching phenomenon of our system by tracking the re-entrant jet breakup process in the presence of surfactants. To do so, we take as a case study the first ‘interior pinch-off’ event of a simulation run characterised by  $k = 0.25$  ( $Da = 0.4$ ), with the remaining parameters kept constant from those ascribed to the base case in §II B. The results from this analysis are summarised in figure 5. Remarkably, these results are in agreement with the existing literature on surfactant-covered liquid thread breakup surrounded by a static gas phase (see for example Craster et al. [23], Kamat et al. [46], Liao et al. [52], Wee et al. [87], Xu et al. [89]); our surfactant-laden system, which is characterised by a moving liquid jet in confined flow with non-negligible inertia, adheres to a breakup mode in the inertial-viscous regime of Eggers [28] as  $r_{min}(t) \rightarrow 0$  and to the inertial-capillary regime in the early thinning stages, akin to our observations for the surfactant-free case. As well reported in the references provided above, at the locality of the pinch-off, the competing inertial, viscous, capillary, and Marangoni-related forces balance in a way that makes the thinning neck evolve as if there were no surfactants present, even if large Marangoni stresses arise near the pinching point to re-open the neck [46]. The intermediary dynamics between that characterised by the  $(\tilde{t}_b - \tilde{t})^{2/3}$  and  $(\tilde{t}_b - \tilde{t})$  scalings, however, is influenced by the presence of surfactant. The existence of intermediary scalings that eventually lead to the inertial-viscous regime has been reported previously in Xu et al. [89] for small  $Pe$  and seen in Kamat et al. [46], Wee et al. [87] for  $Pe \rightarrow \infty$  (see figure 5 of the first reference). This is illustrated in figure 5(b) where the lower half of the jet is coloured by interfacial surfactant concentration,  $\tilde{\Gamma}$ . Due to the confinement effects present in our system, the bulk flow in the axial direction drags surfactants from their accumulation zones at the bubble’s rear and distributes them along the liquid jet. In our system, this action appears to counteract the surfactant depletion process at the pinching point typically reported in surfactant-covered capillary breakup [23, 87].

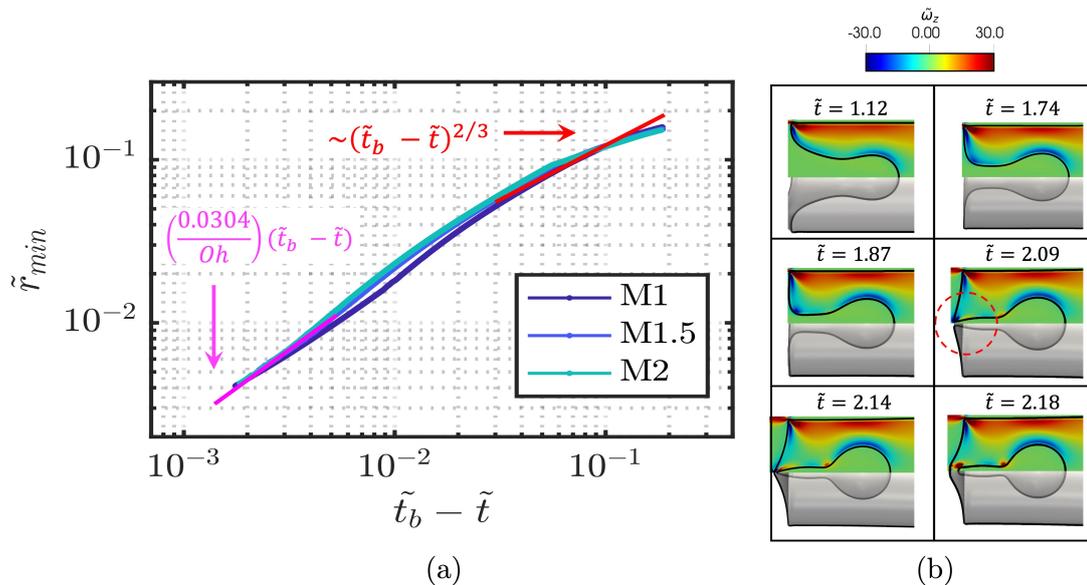


FIG. 4: Neck thinning analysis for the ‘back pinch-off’ event of a surfactant-free interface. (a) Plot of the minimum neck radius,  $r_{min}$ , vs. the time before pinch-off,  $t_p - t$ , depicting how our simulations correctly retrieve the well-known inertial-viscous (pink line) pinching regime of Eggers [28] and approximate the inertial-capillary regime predicted by Day et al. [24]. Here, the time has been scaled by the inertial-capillary time scale,  $t_{in-cap} = \sqrt{\rho_l D^3 / \sigma_s}$ . (b) Zoomed-in  $\tilde{\omega}_z$  contour plots of the pinch-off. All other parameters remain unchanged from those specified in §II B for the base case.

## 2. Marangoni stresses, surface elasticity, and Biot number

Herein we elucidate the behaviour of a surfactant-laden system under the conditions ascribed to the ‘base’ case (refer to §II B) and compare it to that of clean interfaces. To isolate the effects of lower surface tension from those arising from Marangoni stresses on the encapsulation dynamics, we have set up an additional case, denoted by  $|\tilde{\tau}_m| = 0$ , in which we have suppressed the last term on the RHS of Eq. (2) to inhibit Marangoni stresses while simultaneously allowing the reduction of surface tension. Figures 6(a)-(b) show the shape of the liquid jet and a few other characteristics along its interface at  $\tilde{t} = 2.09 - 2.11$ . We have duplicated the plots of interfacial shape on both column panels for clarity. Examination of the plots uncovers an almost surfactant-free interface at the liquid jet for  $|\tilde{\tau}_m| = 0$ , which exhibits a significant drop in  $\tilde{\Gamma}$  going from the edge to the interior of the re-entrant jet in the stream-wise direction and a slight surge after surpassing the incipient pinching neck. This rapid loss of surfactant for  $|\tilde{\tau}_m| = 0$ , as opposed to the Marangoni-supported case,  $|\tilde{\tau}_m| > 0$ , is explained by noticing that the base case’s operating conditions promote surfactant desorption (large  $Da$  and small  $k$ ) as  $\tilde{\Gamma} \rightarrow 1$  (see Eq. (6)) as well as slow interface surfactant diffusion (large  $Pe_s$ ). Reverting back to the remarks by Batchvarov et al. [10] and Atasi et al. [8], the motion of the bubble prompts surfactant accumulation at the back due to drag forces from the liquid phase, partially counteracted by Marangoni stresses in the opposite direction for  $|\tilde{\tau}_m| > 0$ . The absence of Marangoni stresses in the  $|\tilde{\tau}_m| = 0$  case leads to a continuous process of local surfactant accumulation at the back and desorption into the bulk, depleting the interface of surfactants.

Figure 6(c) portrays a key difference in the encapsulation behaviour between the three cases, where the succession of back pinch-off first and interior pinch-off second observed for clean interfaces is reversed in the presence of surfactants and the overall pinch-off dynamics are delayed (see the figure caption for the pinch-off times). This delay is accentuated for  $|\tilde{\tau}_m| > 0$ , as can be observed from the local peaks in  $\tilde{\kappa}$  and  $\tilde{\tau}_n$  of figure 6(b), which, in the case of  $|\tilde{\tau}_m| = 0$ , are already pulling the interface towards the channel centre, signalling the onset of capillary neck closure. Part of the role of Marangoni stresses can be seen in the green-shaded region of figure 6(a), where these tangential stresses counteract capillary forces at both the back and interior pinch-off regions. These observations are in complete agreement with Refs. [19, 46, 47] for liquid threads, although we note that in our system the Marangoni forces exerted on the interface are not sufficient to escape pinch-off completely. The inversion of back and interior pinch-off times in contaminated interfaces can be attributed to the noticeably non-uniform surfactant distribution along the liquid jet, which displays its highest values in the regions neighbouring the capillary neck at the back of the jet, lowering surface tension and providing additional disruption to neck closure in comparison to the interior neck.

The effect of the surfactant-induced interior and back pinch-off delays can be further described through the evolution of the satellite drop, illustrated in figure 6(c) for  $|\tilde{\tau}_m| > 0$ . Unlike its surfactant-free counterpart (see figure 3(c),  $\tilde{t} = 2.37 - 2.98$ ), this satellite drop does not re-coalesce with the main drop before the bubble reaches the end of the channel domain in our

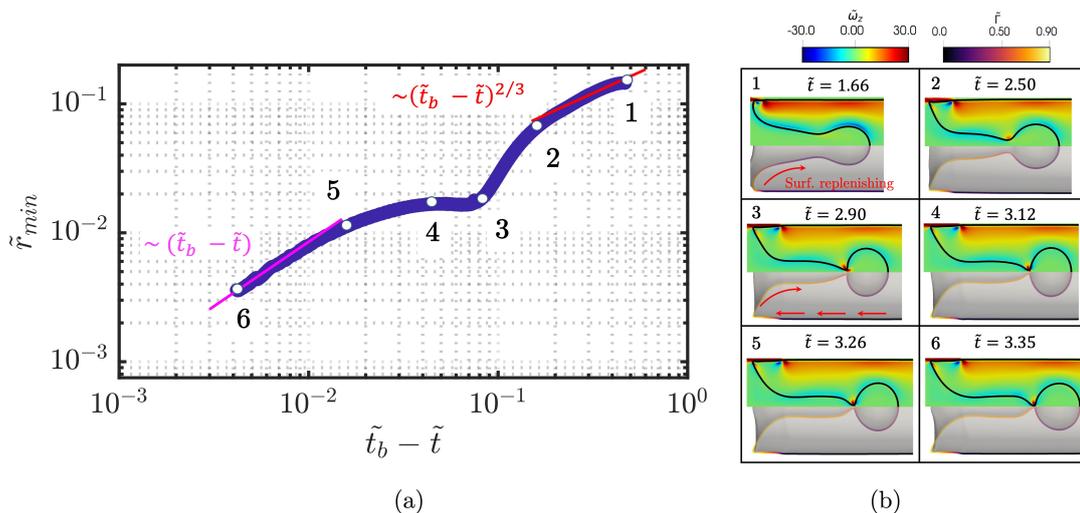


FIG. 5: Neck thinning analysis for the ‘interior pinch-off’ event of a surfactant-laden case characterised by  $k = 0.25$  ( $Da = 0.4$ ). (a) Plot of the minimum neck radius,  $r_{min}$ , vs. the time before pinch-off,  $t_p - t$ , depicting how our simulations correctly retrieve the well-known inertial-capillary (red line) and inertial-viscous (pink line) pinching regimes predicted by Eggers [28] and Day et al. [24], and the occurrence of three intermediary regimes. Here, the time has been scaled by the inertial-capillary time scale,  $t_{in-cap} = \sqrt{\rho_l D^3 / \sigma_s}$ . (b) Zoomed-in  $\tilde{\omega}_z$  (top) and  $\tilde{\Gamma}$  (bottom) contour plots of the pinch-off. All other parameters remain unchanged from those specified in §II B for the base case.

simulations. Nevertheless, we note that the satellite drop’s average velocity exceeds that of the main drop by about 30% under the conditions displayed in figure 6(c), suggesting an eventual drop coalescence in an extended channel. In surfactant-laden scenarios, the delayed coalescence of drops is attributed to the re-entrant jet’s lower surface tension, allowing faster infiltration into the bubble due to reduced resistance to liquid flow. This, in combination with Marangoni stresses opposing the jet’s breakup, causes a delay in the back and interior pinch-off events and increases the spatial separation between the main and satellite drop. Measurements at  $\tilde{t} = 2.66$  reveal a dimensionless distance ( $\tilde{d}_{main,sat}$ ) of 0.13 for surfactant-free cases and 0.31 for the surfactant-laden case depicted in figure 6(c). We note that the formation of the satellite drop is confined to surfactant-laden cases with highly desorptive characteristics, as explained in §III C.

For the first part of our parametric investigation, we study the effect of  $\beta_s$  on encapsulation and the post pinch-off dynamics. As figure 7(a) depicts, and in line with previous studies in the context of ink-jet printing [7, 90], increasing surfactant strength gradually increases the speed of liquid infiltration and decreases the rate of neck thinning at both the back and interior of the re-entrant jet (see locations of jet nose in figure 7(a) and pinch-off times in the figure caption). This delay is materialised in the larger  $|\tilde{\kappa}|$  peaks seen for  $\beta_s = 0.25$  at the incipient pinching necks, as well as the higher  $|\tilde{\tau}_m|$  that arise at their vicinity, seeking to counteract capillary draining [3]. Figure 7(b) illustrates the progression of the primary encapsulated drop as it travels across the bubble domain in the form of its volume-averaged  $x$ -velocity,  $u_{x,d}$ , normalised by  $U_b$  (top), interfacial area,  $A_d$ , normalised by its area after pinch-off,  $A_{d,0}$  (middle), and ratio between drop length in the  $x$  and  $y$ -directions,  $a/b$  (bottom). The encapsulated drops exhibit three common characteristics irrespective of the presence of surfactants; namely, a higher stream-wise velocity than the bubble nose, highlighting the potential for a scenario in which the bubble nose interface is ruptured by the drop, an overall decrease in interfacial area, and dampening  $a/b$  oscillations in time, which culminate in virtually spherical drops. This bubble nose rupture is further analysed in §III C. Under the conditions considered, the addition of a strong surfactant ( $\beta_s = 1$ ) decreases the rate of surface area reduction and significantly increases drop velocity relative to the bubble nose. The non-monotonic behaviour of  $u_{x,d}/U_b$  with regard to a clean interface and weak surfactants ( $\beta_s = 0.25 - 0.50$ ) can be explained by observing in figure 7(c) the spatial distribution of  $\tilde{\Gamma}$ ,  $\tilde{\sigma}$  and  $\tilde{\tau}_m$  in the encapsulated drop at  $\tilde{t} = 7.70$ . Despite the largest  $|\tilde{\tau}_m|$  opposing drop propagation in the stream-wise direction for the strongest surfactant, it is seen that these effects are fully countered by the lower surface tension across the drop’s domain, yielding a higher drop velocity than the surfactant-free case. In contrast, the weak surfactant cases feature an almost clean interface with surface tension values very close to the clean case, but with non-zero Marangoni effects that induce a delay in drop propagation.

The morphological deformations that characterise drop behaviour after pinch-off, as depicted in figure 7(b)(bottom), demonstrate the presence of a semi-periodic oscillating pattern monotonically influenced by surface elasticity. In the inset of this figure, a higher deviation from a fully spherical drop can be appreciated for the surfactant-free case in both the major crests and troughs of the oscillatory structures. Considering these results, we draw parallels with the study of Wang et al. [85] in an ink-jet system, whose main results suggest that the extent of deformation decreases with  $Ca$  (after a critical  $Ca$  has been reached), in line with

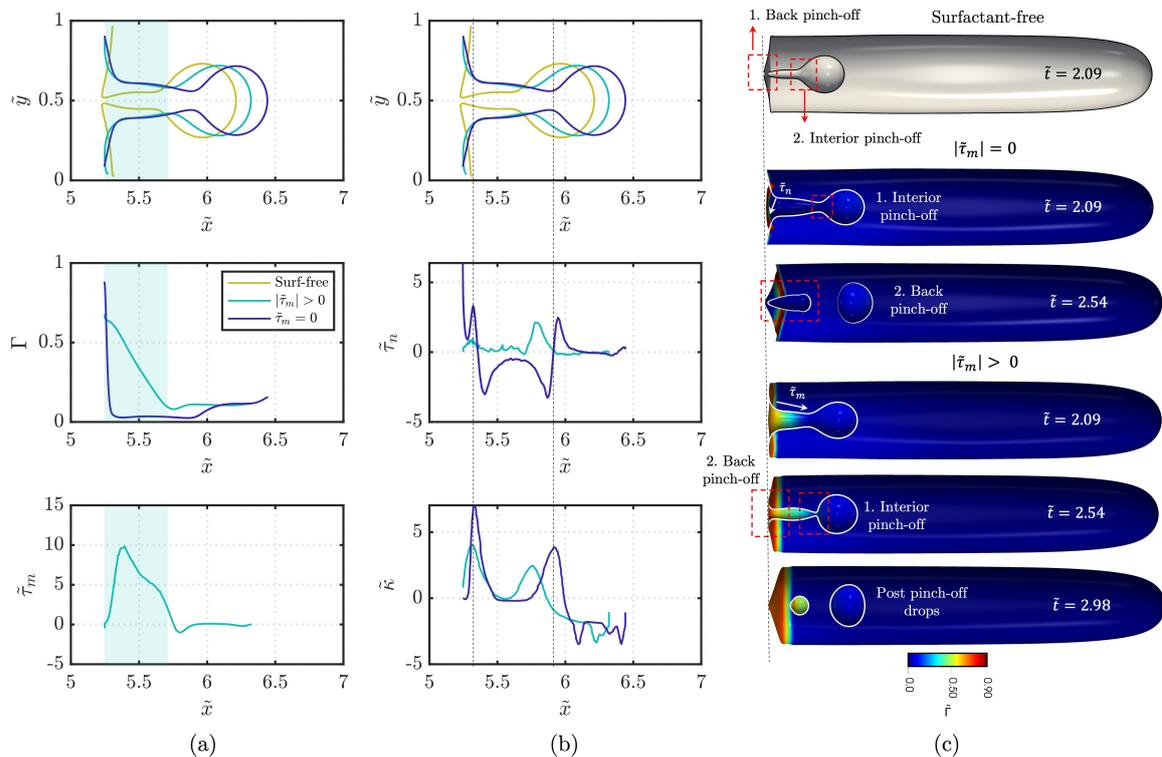


FIG. 6: Effect of Marangoni stresses on re-entrant liquid jet formation and encapsulation. (a) Liquid-filled jet shape (top), surfactant interfacial concentration (middle), and Marangoni stresses (bottom). (b) Liquid-filled jet shape (top), normal stresses (middle), and interface curvature (bottom).  $|\tilde{\tau}_m| > 0$  corresponds to Marangoni-supported, while  $|\tilde{\tau}_m| = 0$  to Marangoni-free. Plots on two-dimensional projection in  $x$ - $y$  plane ( $\tilde{z} = 0.5$ ) and at  $\tilde{t} = 2.09, 2.10, 2.11$  for surfactant-free,  $|\tilde{\tau}_m| = 0$ , and  $|\tilde{\tau}_m| > 0$ , respectively. (c) 3-D schematic of pinch-off mechanisms. The interfaces of these images have been slightly translated in the  $x$ -direction to have a common point of reference (marked by the black dotted line); for their real location in the channel, refer to (a, top).  $\tilde{t}_{p-o,bk} = 2.16, 2.55, 2.90$  and  $\tilde{t}_{p-o,int} = 2.33, 2.37, 2.62$  for surfactant-free,  $|\tilde{\tau}_m| = 0$ , and  $|\tilde{\tau}_m| > 0$ , respectively. All other parameters remain unchanged from those specified in §II B for the base case.

our numerical observations for increasing  $\beta_s$ . Estimating the frequency of  $a/b$  oscillations through the classic expression of Rayleigh [70] ( $f \sim \sqrt{\sigma/\rho_c r_d^3}$ , where  $r_d$  is the drop's spherical equivalent radius) at the latest time recorded ( $\tilde{t} \approx 13.17$ ), we obtain  $\tilde{f} \sim 1.49, 1.55, 1.57$  for surfactant-free and  $\beta_s = 0.25, 0.50, 1$ , respectively, implying a dominating effect of lower drop radius over lower surface tension on the frequency of deformation cycles for increasing  $\beta_s$ .

The effects arising from altering  $Bi$  in the range  $0.01 - 1$  are depicted in the plots of figure 8. An interface covered by a highly soluble surfactant (i.e., high  $Bi$ ) undergoes rapid rates of mass exchange between the interface and bulk as a result of its comparatively low (high) characteristic desorptive (inertial) time. This is demonstrated by noticing the significantly lower  $\tilde{\Gamma}$  distributed along the liquid jet for the highest  $Bi$  considered, which, barring the surfactant-free case ( $Bi \rightarrow \infty$ ), exhibits the slowest rate of jet formation and the fastest pinch-off events (see location of jet nose in figure 8(a)(top) and figure caption for the pinch-off times). Similar to our previous remarks, the signature  $|\tilde{\tau}_m|$ ,  $|\tilde{\kappa}|$ , and capillary pressure peaks (not shown) that signal the onset of capillary neck closure and the advent of pinch-off are already developing in a noticeable manner for  $Bi = 1$ , in contrast to  $Bi = 0.01 - 0.10$ . The post pinch-off temporal evolution of the primary drop, as depicted in figure 8(b), reveals similar trends to those referenced in the above analysis of  $\beta_s$  in terms of drop shrinkage, its attenuation in surfactant-laden cases, and semi-periodic drop size oscillations.

Figure 9 elucidates the functional relationships that interlink  $\beta_s$  and  $Bi$  to key outcomes characterising the encapsulating system. These outcomes encompass the first pinch-off event ( $\tilde{t}_{p-o,1}$ ), the post-detachment volume-averaged drop velocity ( $\tilde{u}_{x,d}$ ), the first encapsulated drop's volume (normalised by the bubble's initial volume,  $V_d/V_0$ ), and the drop's interface surfactant concentration (normalised by the initial concentration,  $\tilde{\Gamma}_d/\tilde{\Gamma}_0$ ). The figure incorporates best-fit curves for each variable for reference. An examination of the relationships pertaining to  $\beta_s$  in figure 9(a) unveils a distinct divergence between behavioural modes for  $\beta_s < 0.4$  (highlighted in blue) and  $\beta_s \geq 0.4$  (highlighted in green). This divergence is particularly evident for  $\tilde{t}_{p-o,1}$  and  $\tilde{u}_{x,d}$ . In the latter  $\beta_s$  range, well-established surfactant-induced effects, discussed in this paper and widely reported in the

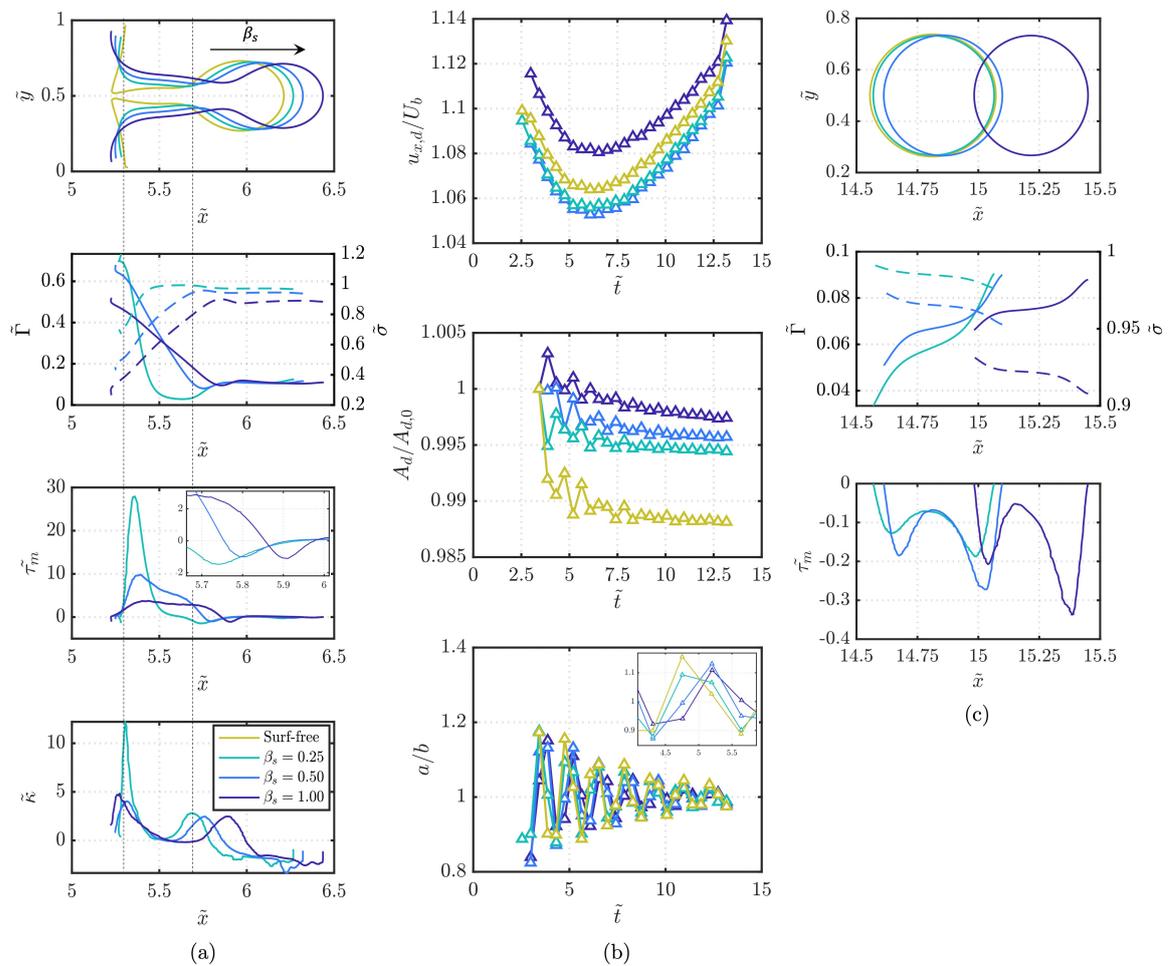


FIG. 7: Effect of  $\beta_s$  on re-entrant liquid jet formation and encapsulation. (a) Liquid-filled jet shape, surfactant interfacial concentration (left axis, continuous line) and surface tension (right axis, dotted line), Marangoni stresses, and interface curvature at  $\tilde{t} = 2.09, 2.10, 2.10, 2.09$  for surfactant-free, and  $\beta_s = 0.25, 0.50, 1$ , respectively. (b) Temporal evolution of primary encapsulated drop velocity normalised by bubble velocity, primary drop interfacial area normalised by area after pinch-off, and ratio between drop length in the  $x$  and  $y$  directions. Data points computed for all cases every  $10^{-3}$  s. (c) Primary encapsulated drop shape, surfactant interfacial concentration (left axis, continuous line), and surface tension (right axis, dotted line) and Marangoni stresses at  $\tilde{t} = 7.70$ . Plots on two-dimensional projection in  $x$ - $y$  plane ( $\tilde{z} = 0.5$ ) for (a,c).  $\tilde{t}_{p-o,bk} = 2.16, 2.52, 2.90, 3.83$  and  $\tilde{t}_{p-o,int} = 2.33, 2.40, 2.62, 2.77$  for surfactant-free, and  $\beta_s = 0.25, 0.50, 1$ , respectively. All other parameters remain unchanged from those specified in §II B for the base case

literature, come to the forefront. These include the concurrent impact of stabilising Marangoni stresses and reduced local surface tension (i.e., increase in local  $Oh$ ) contributing to thwart/delay capillary breakup and reduce drop size, as previously reported in Refs. [45–47].

Our simulation results reveal that a surface elasticity surge (maintaining all other dimensionless groups unchanged) brings about the generation of encapsulated drops of increasing surfactant concentration and smaller size (see plots of  $\Gamma_d/\Gamma_0$  and  $V_d/V_0$ ). A larger  $\Gamma_d/\Gamma_0$ , in tandem with an augmented sensitivity of  $\tilde{\sigma}$  to  $\tilde{\Gamma}$  for increased  $\beta_s$  (indicated by the Langmuir relation in Eq. (7)) point to lower  $\tilde{\sigma}$  during the re-entrant jet infiltration and pinch-off events. In fact, applying the Langmuir relation to the  $\Gamma_d/\Gamma_0$  data of figure 9(a), we find that  $\tilde{\sigma}$  ranges from 1 to 0.869 for  $\beta_s = 0$  and  $\beta_s = 1$ , respectively. Coupling this analysis with the understanding that  $|\tau_m| \sim \beta_s$  (see §II A), it is possible to rationalise the direct relationship between  $\tilde{t}_{p-o,1}$  and  $\beta_s$  by invoking the above mentioned compounded action of larger Marangoni stresses and lower surface tension to resist capillary breakup for larger values of  $\beta_s$ .

The direct relationship registered between  $\tilde{u}_{x,d}$  and  $\beta_s$  for  $\beta_s \geq 0.4$ , featuring a linear growth under our studied conditions, owes its behaviour to the diminished capillary resistance to liquid infiltration into the bubble's domain, as mentioned previously in §I. This directly proportional response of  $\tilde{u}_{x,d}$  to  $\beta_s$  is akin to the well-established links between velocity of drops formed

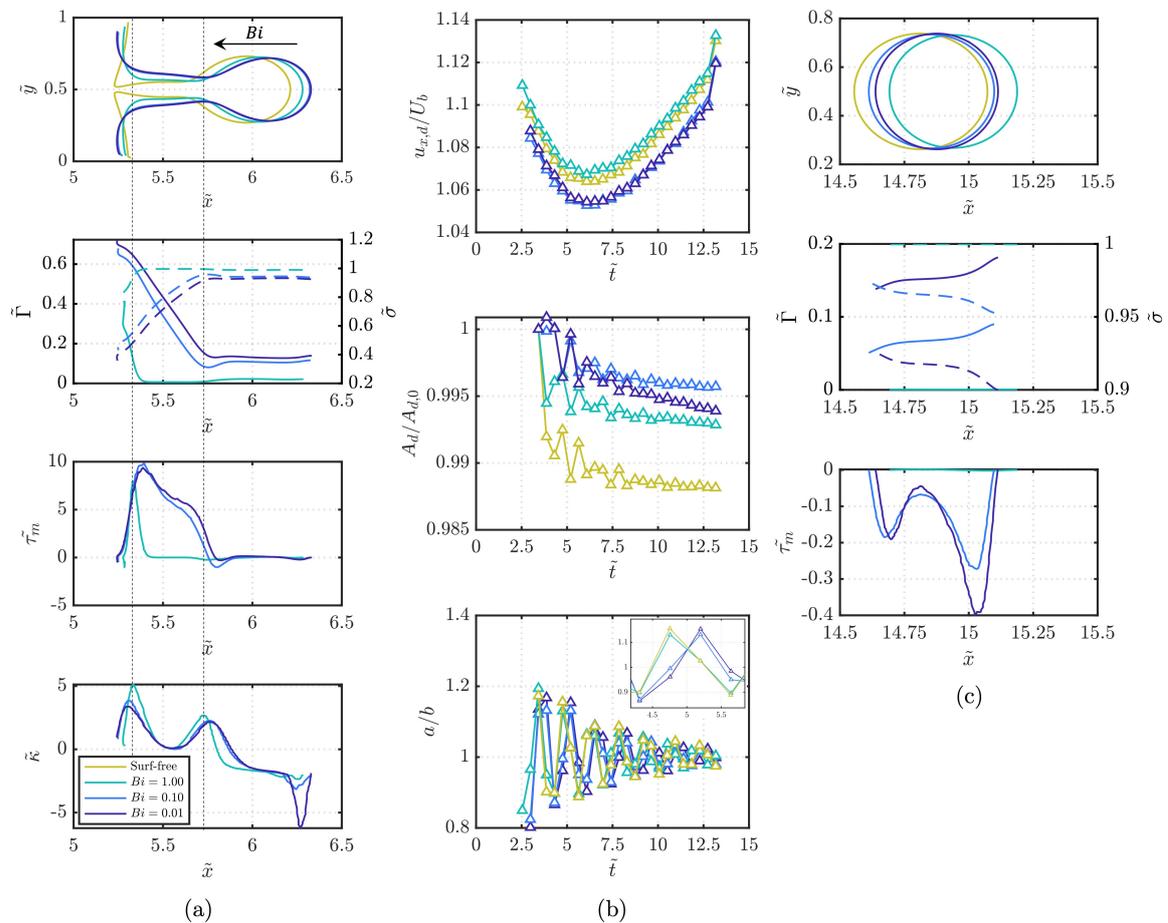


FIG. 8: Effect of  $Bi$  on re-entrant liquid jet formation and encapsulation. (a) Liquid-filled jet shape, surfactant interfacial concentration (left axis, continuous line) and surface tension (right axis, dashed line), Marangoni stresses, and interface curvature at  $\tilde{t} = 2.09, 2.10, 2.10, 2.10$  for surfactant-free and  $Bi = 1, 0.10, 0.01$ , respectively. (b) Temporal evolution of primary encapsulated drop velocity normalised by bubble velocity, primary drop interfacial area normalised by area after pinch-off, and ratio between drop length in the  $x$  and  $y$  directions. Data points computed for all cases every  $10^{-3} s$ . (c) Primary encapsulated drop shape, surfactant interfacial concentration (left axis, continuous line), surface tension (right axis, dashed line), and Marangoni stresses at  $\tilde{t} = 7.70$ . Plots on two-dimensional projection in  $x - y$  plane ( $\tilde{z} = 0.5$ ) for (a,c).  $\tilde{t}_{p-o,bk} = 2.16, 2.63, 2.90, 3.09$  and  $\tilde{t}_{p-o,int} = 2.33, 2.37, 2.62, 2.68$  for surfactant-free and  $Bi = 1, 0.10, 0.01$ , respectively. All other parameters remain unchanged from those specified in §II B for the base case

through end-pinching and  $Oh$  ( $\sim \sigma^{-1/2}$ , precipitated by larger  $\beta_s$  in our simulations, as previously explained) in the problem of jet droplets released to the atmosphere through bursting bubbles [37]. The inversely monotonic  $\tilde{u}_{x,d}$  vs.  $\beta_s$  relationship, evident for  $\beta_s < 0.4$ , embodies the nuanced effects of surface tension and Marangoni stresses, as briefly discussed in our analysis of figure 7. The uneven distribution of surfactant across encapsulated drops (larger concentration at the drop's nose, refer to figure 7(c, bottom)) instigates counter-flow Marangoni stresses ( $\tilde{\tau}_{m,x} < 0$ ), hindering the motion of the drop. These Marangoni stresses, though small, are non-negligible, constituting approximately 10% of the Marangoni forces exerted on the infiltrating jet prior to pinch-off (see figure 7). For instances where  $\beta_s < 0.4$ , the anticipated acceleration of the drop due to reduced surface tension is offset by these Marangoni stresses, thereby resulting in a slightly decreased drop velocity with increased surfactant strength.

The parameter sweep of surfactant solubility elucidated in figure 9(b) mirrors that of  $\beta_s$  in the discernible emergence of dual behavioural modes, this time noticeable for  $\tilde{u}_{x,d}$  and  $V_d/V_0$  and clearly separated by  $Bi \approx 0.30$ . Before understanding this duality (highlighted in gray and magenta), we examine the expected behaviour detected for  $\tilde{t}_{p-o,1}$  and  $\Gamma_d/\Gamma_0$ . Both variables conform to a conspicuous exponential decline with increased  $Bi$ . This signals that elevated rates of mass transfer between the interface and the bulk compared to convection—indicative of enhanced solubility or larger  $Bi$ —predominantly deplete the interface of surfactant and concurrently accelerate capillary breakup with respect to lower  $Bi$ . Notice that these cases correspond to  $k = t_{des}/t_{ad} = 0.1$ , stipulating that surfactant desorption is heavily promoted over adsorption, leading to the high-rate plummeting of  $\tilde{\Gamma}/\tilde{\Gamma}_0$  with  $Bi$  and the roughly surfactant-free drops observed for  $Bi > 1$  ( $\tilde{\sigma} \approx 1 - 0.08e^{-3.81Bi}$ , not shown in the plot). The inverse

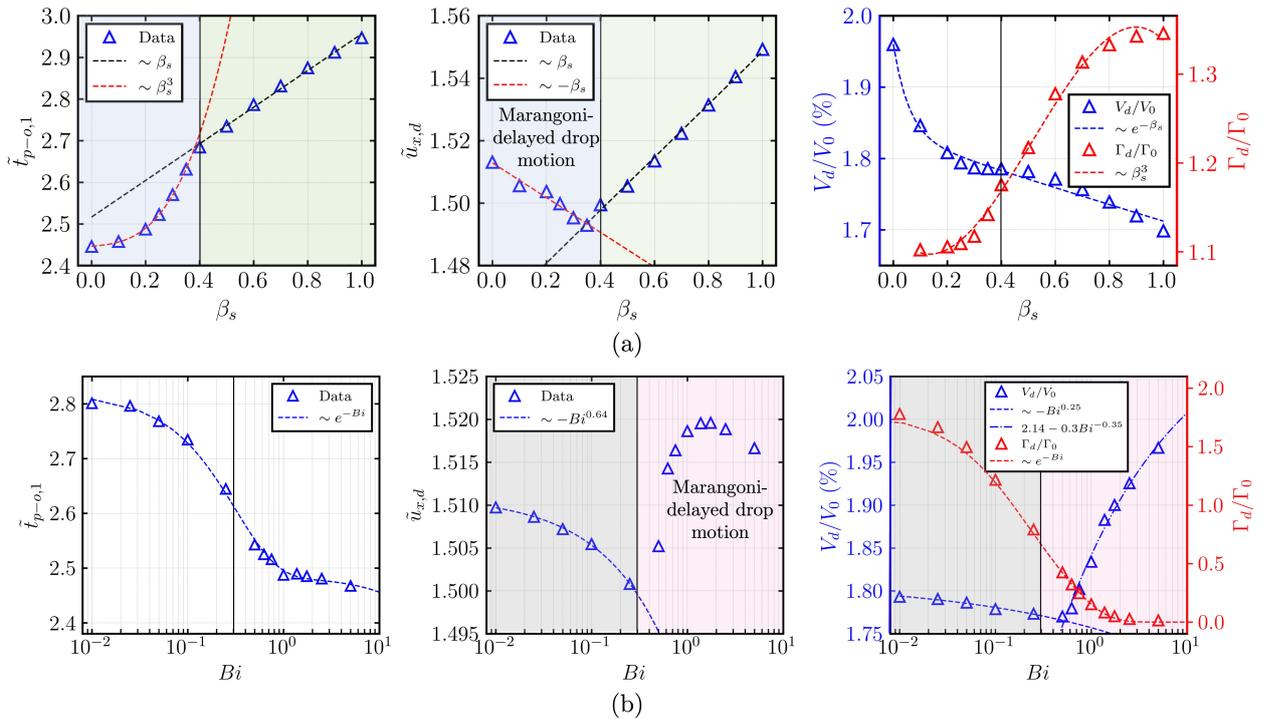


FIG. 9: Impact of surfactant parameters on pinch-off dynamics and evolution of first encapsulated drop for a highly desorptive surfactant ( $k = 0.1$ ). (a) Effect of  $\beta_s$  at  $Bi = 0.1$ . (b) Effect of  $Bi$  at  $\beta_s = 0.5$ . All other parameters remain unchanged from those specified in §II B for the base case

exponential dependence recovered for  $\tilde{t}_{p-o,1}$  and  $\Gamma_d/\Gamma_0$  imposes explicit bounds for these values between the two limiting cases: surfactant-free ( $Bi \rightarrow \infty$ ,  $\tilde{t}_{p-o,1} = 2.48$  and  $\Gamma/\Gamma_0 = 0$ ) and insoluble surfactant ( $Bi = 0$ ,  $\tilde{t}_{p-o,1} = 2.81$  and  $\Gamma/\Gamma_0 = 1.87$ ). The approximately two-fold increase in drop surfactant concentration with respect to initial surfactant coverage as the insoluble limit is approached is consistent with other applications. For instance, in the context of jet drops formed via bursting bubbles, Ref. [22] reports a 1.6 times increase in  $\tilde{\Gamma}$  of the released drops with respect to that of the equilibrated bubble interface before bursting.

Focusing on the behavioural divergence obtained for  $Bi \approx 0.3$ , an inverse relation between  $V_d/V_0$  and  $Bi$  is recovered for  $Bi > 0.3$ . In a similar fashion to the trends elucidated for surface elasticity, the diminished  $\tilde{\sigma}$  associated with augmented surfactant coverage as  $Bi$  decreases generates smaller encapsulated drops. As we venture into the  $Bi \lesssim 0.3$  territory, a pronounced plunge in the sensitivity of  $V_d/V_0$  to  $Bi$  is identified with respect to that of  $Bi > 0.3$ . The  $Bi < 0.3$  range adheres to  $|d(V_0/V_d)/dBi| = -0.0125Bi^{-3/4}$ , whereas  $Bi > 0.3$  is driven by  $|d(V_0/V_d)/dBi| = 0.105Bi^{-1.35}$  (1-2 orders of magnitude larger  $|d(V_0/V_d)/dBi|$  for  $Bi > 0.3$  within the entire  $Bi$  range studied). This analysis hints to a saturation of surfactant effects for increasingly insoluble contaminants. To finalise the present discussion of figure 9, we bring attention to the similar behaviour of  $\tilde{u}_{d,x}$  in  $Bi > 0.3$  as that in  $\beta_s < 0.4$ . That is, a neutralisation of the drop acceleration effects introduced by lower surface tension (prompted by larger  $\beta_s$  or smaller  $Bi$ ) by counter-flow Marangoni stresses ( $\tilde{\tau}_{m,x} < 0$ ).

### 3. Effect of surfactant adsorption/desorption kinetics ( $Da$ and $k$ )

We now proceed to examine the influence of directly contrasting the characteristic times of adsorption and desorption, along with the initial interface saturation through varying  $k$  and  $Da$  simultaneously. This concurrent variation allows us to maintain all other parameters constant. Figure 10(a) portrays a monotonic response of the liquid infiltration depth and  $\tilde{\Gamma}$  with  $k$  and  $Da$ , where the rapid rates of surfactant adsorption inherent to the high  $k$  (low  $Da$ ) cases are materialised in the relatively large and comparatively constant  $\tilde{\Gamma}$  distributions along the re-entrant jet. In line with our previous results, general reductions in local surface tension at the bubble rear, brought about by higher  $k$  or  $\beta_s$  and lower  $Bi$  conditions, promote a faster and deeper infiltration of liquid into the bubble domain. In figure 10(b) we record the time until the first pinch-off instance at the bubble interior as a function of  $k$  for the entire parameter space explored. A retardation of the capillary instabilities that lead to end-pinching is seen as  $k$  is increased in the range  $0$  (surfactant-free)  $< k < 1$ , consistent with our previous remarks about the combined effect

of  $\tilde{\sigma}$  and Marangoni stresses on opposing drop encapsulation. This behaviour is mirrored by the  $\tilde{\tau}_m$  and  $\tilde{\kappa}$  peaks exposed near the nascent pinching necks for the lowest  $k$  cases displayed in figure 10(a) ( $k = 0.1 - 1.0$ ), while the oscillations seen for  $k = 10$  correspond to capillary waves in the infiltrating liquid, reported by Constante-Amores et al. [19] for long liquid ligaments.

Similar to our previous analyses of  $\beta_s$ ,  $Bi$ , and Marangoni-free case for  $k = 0.10$ , it is found that increasing  $k$  in the range  $k < 0.10$  results in a sequence inversion of the two main pinch-off events ( $\tilde{t}_{p-o,bk} > \tilde{t}_{p-o,int}$ ) identified in clean interfaces ( $\tilde{t}_{p-o,bk} < \tilde{t}_{p-o,int}$ , see caption of figure 10). Interestingly, it is also found that for all cases above  $k = 0.25$ , the large accumulation of surfactants nearby the bubble rear provides a strong resistance to capillary pressure buildup in those regions, resulting in the partial elimination of the ‘back pinch-off’ event until bubble bursting events occur. This will be further discussed in the following section. Another notable feature to highlight from figure 10(b) is the sharp decrease in  $\tilde{t}_{p-o,int}$  above the critical value of  $k = 1$ . This discontinuity in the function  $\tilde{t}_{p-o,int}$  vs.  $k$ , in conjunction with our observations about the elimination of the back pinch-off suggest a partition of the encapsulation dynamics into two major regions according to the rates of adsorption/desorption: i) the above described region for  $k < 1$ , in which the presence of surfactants exerts a stabilising effect, leading to a delay in neck thinning in relation to a fully clean interface; and, ii) a region of rapid liquid-filled jet formation in the axial direction alongside a significant acceleration of the first interior pinch-off event for  $k > 1$ . The first region is further segregated by the occurrence ( $k < 0.25$ , region i.a) or elimination ( $k > 0.25$ , region i.b) of the back pinch-off. An explanation for the marked transition from the first to the second region in terms of thinning rates is as follows. A general increase of surfactant coverage due to high rates of adsorption introduces high rates of viscous and inertial deformation to the interface, as noted by Bazhlekov et al. [11], hence, increasing the axial velocity of the infiltrating liquid and its aspect ratio. Recalling the observations of Wang et al. [84] and Ambravaneswaran and Basaran [3] in contracting/expanding liquid filaments, we show in figure 10(c) how in cases above the critical  $k$ ,  $L_{jet}/R_{jet}$  surpasses a given threshold value to enter the breakup regime and accelerate end-pinching, even if the local values of surface tension are lower than those of smaller  $k$  conditions.

### C. Bubble bursting, back deformations, and regime maps

This section provides a more detailed account of the post pinch-off dynamics, focusing specifically on the process of bubble bursting via the liquid jet/entrapped drops and the temporal fate of the bubble back and accompanying trailing structures. In figure 11(a)-(b) we summarise the three main bursting behaviours observed in the  $\beta_s$ - $k$  and  $Bi$ - $k$  ( $Da$ ) spaces, leaving all other conditions specified in the baseline case unchanged. As captured in the figure, a first regime (I), distinctly bounded by a surfactant-free interface and  $k$  below the critical value defined in the previous section, is identified for all  $\beta_s$  and  $Bi$  tested. This regime adheres to a behaviour mode of slow infiltration of liquid, monotonic delay of the first interior pinch-off event with increasing (decreasing)  $\beta_s$  ( $Bi$ ) (see figure 9), and most importantly, eventual restoration of the interfacial morphology of the bubble back, depicted in figure 11(c, top). This restoration ensures a pseudo-stable entrapment of one or multiple drops of varying size and  $\tilde{\Gamma}$  within the bubble domain, only disrupted by a potential rupture of the bubble nose by the first drop. We note that this rupture was not observed for any of the cases encompassed within (I) given the channel length employed in our simulations and the significant elongation of the bubble across the channel. The stability of the encapsulated drops-bubble compound in regime I poses a potential avenue for future research.

A second regime (II) emerges as  $k$  approaches the critical value, where a much faster and deeper liquid jet develops across the bubble and, ensuing from diminished capillary forces, further delays to pinch-off are recorded. These delays lead to the entrapment of the first drop in regions approaching the bubble nose in the axial direction, promptly leading to its rupture, as illustrated at  $\tilde{t} = 5.19$  in figure 11(c, middle). Following a series of end-pinching events, the liquid jet undergoes several instances of escapes of pinch-off, in conjunction with a surge of surface capillary waves across its domain, and a radial growth that gives rise to a large bulbous end (see  $\tilde{t} = 8.73$ ). These mechanisms are closely related to the intricate phenomena stemming from retracting liquid filaments, as addressed in Refs. [6, 19]. This substantial growth causes the liquid jet to burst the interface near the channel walls, thereby fully separating the original bubble into two individual entities.

The third regime (III) is confined to regions of above critical  $k$  with moderate  $\beta_s$  and  $0.01 < Bi < 1$  and closely resembles the mechanisms of II, with the pinch-off times being the key differentiating characteristic. In III, the fast expansion of the liquid jet expedites the first pinch-off with respect to the surfactant-free case (see previous sub-section). Although the location of the first entrapment is much farther away from the bubble nose than in II, the encapsulated drops exhibit a comparatively faster velocity (e.g.,  $u_{x,d}/U_b = 1.27$  and  $u_{x,d}/U_b = 1.47$  at  $\tilde{t} = 4.75$  for the cases in II and III emphasised in figure 11(a), respectively), also leading to bubble nose ruptures by the drops and the consequent radial rupture by the bulbous end of the liquid jet mentioned above (see  $\tilde{t} = 10.50$ ).

Figure 11 also displays the numerical data derived from the regime maps proposed in Atasi et al. [8] for surfactant-laden systems. A qualitative comparison is drawn between the bubble rear bursting phenomena characteristic of regime III (see  $\tilde{t} = 10.50$  in figure 11(c)) and the observations of Nath et al. [63] for liquid-liquid flow in clean interfaces (see Figure 15 of this reference). As depicted in the figure, our proposed transition from regime I (curvature inversion without bursting) to regimes II and III (bubble bursting) at  $k \approx 0.45$  for varying  $\beta_s$  and  $Bi$  aligns remarkably well with that found in Atasi et al. [8] for  $Ca \rightarrow 1$  (see figure 16 of this reference). Notice that Ref. [8] does not dive into the pinch-off dynamics of the system, precluding us from

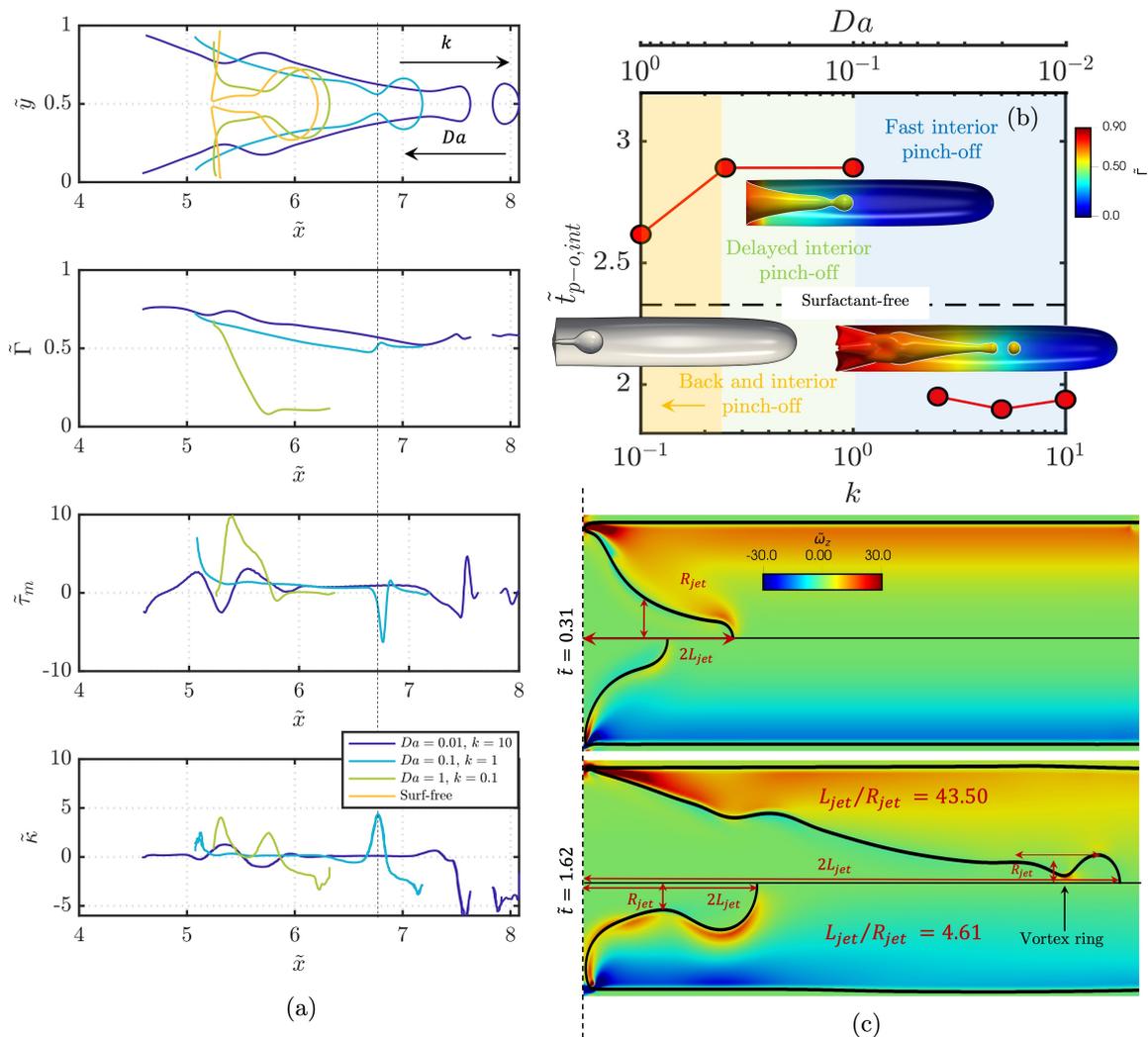


FIG. 10: Effect of  $k$  and  $Da$  on re-entrant liquid jet formation and encapsulation. (a) Liquid-filled jet shape, surfactant interfacial concentration, Marangoni stresses, and interface curvature at  $\tilde{t} = 2.09, 2.10, 2.09, 2.09$  for surfactant-free and  $k = 0.1, 1, 10$ , respectively. (b) Interior pinch-off time vs.  $k$  and  $Da$ , and schematic of the three regions that divide thinning behaviour. (c) Contour plots of normalised vorticity in  $z$ -direction and comparison of infiltrating liquid shape for  $k = 10.0, Da = 0.01$  (top) and  $k = 0.1, Da = 1.0$  (bottom). The interfaces of these contour plots have been translated in the  $x$ -direction to have a common point of reference (marked by the black dotted line). Plots on two-dimensional projection in  $x$ - $y$  plane ( $\tilde{z} = 0.5$ ). All other parameters remain unchanged from those specified in §II B for the base case

making a direct classification of their bubble bursting cases into our regimes II and III.

A few caveats are worth mentioning concerning these comparisons against previous investigations. While our regimes and observations of the bubble's behaviour are entirely in line with previous results, we must emphasise that our operating conditions vary significantly from those of Atasi et al. [8] and Nath et al. [63]. The former reference explores a system with vanishing inertia, a close-to-spherical bubble, and a circular cross-section channel. The latter reference, as previously mentioned, involves the motion of a drop in the absence of surfactant material. These characteristics explain why their encapsulation phenomena are observed for  $Ca$  much larger than ours ( $Ca \sim O(10^0)$  compared to  $Ca = 0.0693$ ) and why it is not possible to map the exact operating conditions of Nath et al. [63] into ours.

To finalise our analysis, we wish to highlight the limitations of our proposed encapsulation regime maps. As briefly mentioned in §II A, gravitational forces are neglected in our simulations ( $Bo = 0$ ) as their effects typically become significant only in channels larger than 1 mm [57], given the prevailing influence of capillary forces at the microscale. Our primary focus in this study was to characterise encapsulation mechanisms within scenarios involving inertia, non-circular geometries, and the presence of contaminants. Hence, isolating these influences from other non-idealities, including gravitational effects, was essential for

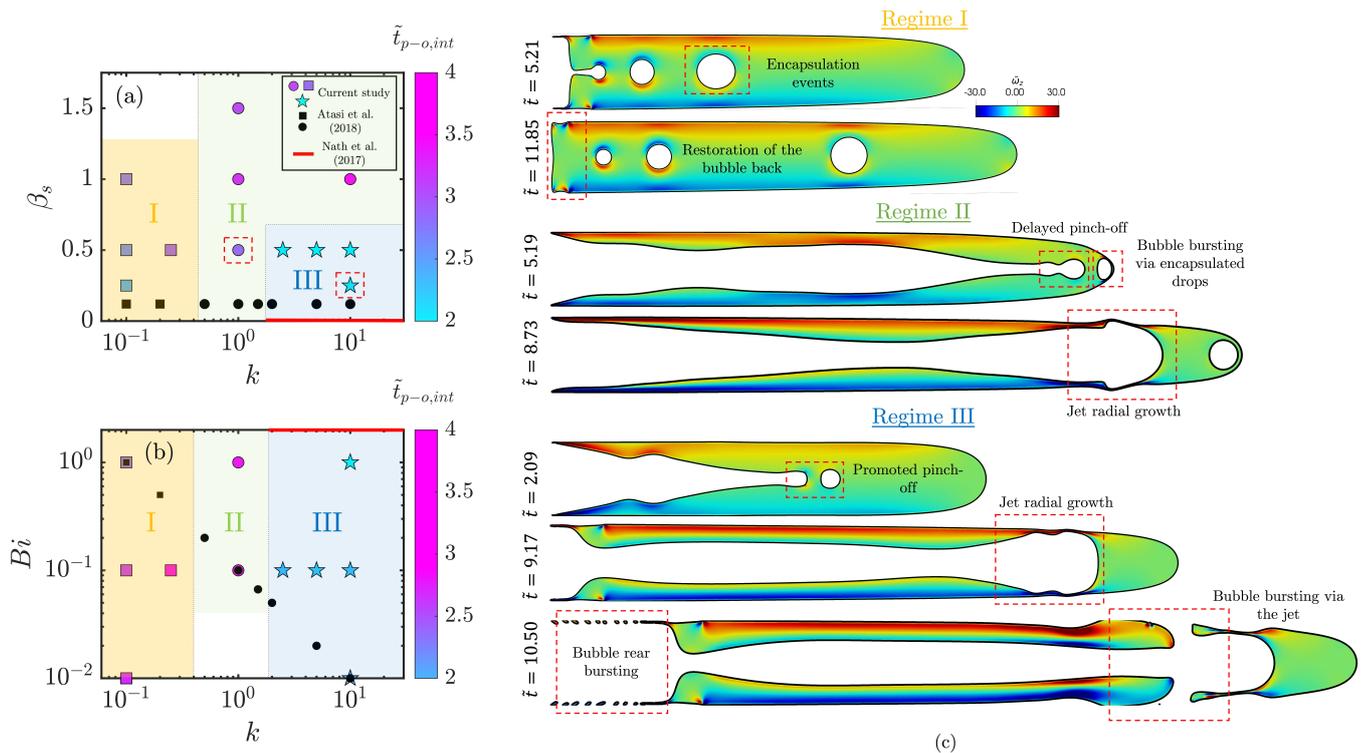


FIG. 11: Encapsulation and bursting regime maps in the (a)  $\beta_s$ - $k$  and (b)  $Bi$ - $k$  spaces.  $Bi = 0.10$  for (a) and  $\beta_s = 0.50$  for (b). Our results, shown as markers coloured by the first interior pinch-off event, reveal three distinct regimes: I (squares), II (circles), and III (stars). The black markers correspond to the data set of Atasi et al. [8], which does not differentiate regime II from III. (c) Schematic contour plots of normalised vorticity in  $z$ -direction and comparison for the three regimes identified. All other parameters remain unchanged from those specified in §II B for the base case. Regime maps developed for  $Bo \rightarrow 0$

our objectives. It is crucial to recognise, however, that the diminished influence of capillary forces—an essential condition for encapsulation, accentuated by surfactants—could potentially magnify the importance of gravitational forces, consequently impacting encapsulation dynamics.

We have assessed the potential impact of gravity by comparing characteristic time scales within our system to the free-falling time of an encapsulated drop. Adopting  $Bo = 0.15$ , a moderate value of  $Bo$  that still induces noticeable gravitational effects on gas-liquid systems [61]), we find that the falling time of encapsulated drops ( $\approx 4.5 \times 10^{-2}$  s) largely exceeds the re-entrant jet’s pinch-off times under our tested conditions ( $\approx 5 - 7 \times 10^{-3}$  s) but is comparable to the total travel time of the bubble across our channel domain ( $\approx 3 - 6 \times 10^{-2}$  s). This suggests that gravitational effects are unlikely to significantly influence the curvature inversion, jet infiltration, and breakup processes elucidated in this study, or the impact of surfactants. Nevertheless, gravity is certain to disrupt the trajectory, velocity of entrapped drops, and thus the proposed regime maps illustrated in figure 11.

#### IV. CONCLUDING REMARKS

This work presents an in-depth characterisation of the unsteady mechanisms describing elongated bubbles flowing across liquid-filled channels via numerical simulations under negligible gravitational effects. These mechanisms arise in opposition to the well-known steady-state phenomena of Taylor bubbles under the traditional assumptions of negligible inertial/viscous effects, symmetric cross-section channels, and the absence of interfacial contaminants. As reported in limited instances (see Atasi et al. [8] and Sauzade and Cubaud [71]) and confirmed here, interesting dynamics emerge as  $We \rightarrow O(10)$ . Notably, the concomitant reduced capillary effects of high  $Ca$  (and  $We$ ) induce a loss of sphericity at the bubble’s front and back, triggering an elongation in the flow direction of the former and a flattening and subsequent curvature inversion of the latter. The underlying mechanisms by which this curvature inversion evolves into a re-entrant liquid jet infiltrating the bubble, how this liquid structure collapses into small liquid drops entrapped in the bubble domain, and the overall role of surfactants in these processes have been analysed in detail. We emphasise the multiple parallels that can be readily drawn between the various interfacial features and singularities that ensue in ours and other well-known systems involving capillary liquid breakup, such as inkjet printing,

two-phase microfluidics, contracting liquid filaments, and sheet cavitation.

By performing a systematic sweep of a number of surfactant parameters and characterising dimensionless groups, we have elucidated the coupled effect of lower surface tension and Marangoni stresses and their interactions with inertia and viscosity. Two distinct modes of liquid jet breakup/drop encapsulation were found for comparatively low rates of surfactant adsorption (low  $k$ ): one closure of the liquid jet at the bubble's rear and one end-pinching breakup mode at the interior of the jet. Our simulations have shown that the combined and individual effects of both surface tension and Marangoni stresses induce a delayed response in terms of pinch-off times, as well as an inversion of the sequence of breakup modes with respect to clean interfaces. Provided that  $k$  is low enough, these effects are maintained for increasingly strong (large  $\beta_s$ ) and less soluble (small  $Bi$ ) surfactants. Under the baseline conditions considered, a critical value of  $k = 1$  represents a phenomenological limit from which an increase of  $k$  represents an acceleration of the second mode of jet breakup when compared to an uncontaminated interface. This is due to the fast and deep liquid infiltration in the bubble at high  $k$ , which promotes capillary breakup.

A summarising map with three regimes of behaviour was constructed in the  $\beta_s$ - $k$  and  $Bi$ - $k$  ( $Da$ ) spaces, where regime I, limited by the critical  $k$ , allows for a semi-stable drop entrapment due to the eventual closure and restoration of the bubble back in the aftermath of pinch-off. Conversely, in regimes II (delayed pinch-off) and III (expedited pinch-off), the deep infiltration of the liquid jet that precedes end-pinching, together with the high relative velocity of the entrapped drops are responsible for the subsequent bursting of the bubble nose. These two regimes, in addition, display a multitude of rich interfacial events following the liquid jet end-pinching, including escapes from pinch-off, radial growth, and an eventual bubble bursting in the vicinity of the channel walls.

To the best of our knowledge, this is the first thorough and detailed characterisation of the phenomenon of drop encapsulation in moving bubbles in the presence of surfactants. There are, however, a few avenues worthy of further pursuit in this topic. For instance, an examination of the influence of surfactant parameters,  $\beta_s$  for example, on the critical  $k$  value. Based on our numerical results, it is likely that this critical  $k$  will decrease with increasing  $\beta_s$ , but additional evidence is required. A deeper exploration of the stability of the encapsulated drops in regime I may also be of interest to determine if there exists a set of conditions that allow for a perpetual entrapment of these drops, and the potential applications of such systems in the context of emulsification. [Finally, an expansion of our proposed regime maps to include the effects of gravitational forces represents an interesting avenue for further research.](#)

**Declaration of Interests.** The authors report no conflict of interest.

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## Appendix A: Surfactant properties and timescales

For typical ionic and non-ionic surfactants in water, such as sodium dodecyl sulphate (SDS), N-dodecyl-N,N-dimethylammonio-3-propane sulfonate, and Triton X-100 (TX100), the value of  $\Gamma_\infty$  ranges between  $O(10^{-6} - 10^{-5})$  mol/m<sup>2</sup> and  $D_s$  between  $O(10^{-12} - 10^{-8})$  m<sup>2</sup>/s, which results in  $Pe_s = O(10^3 - 10^6)$  [22, 44]. As noted previously in Constante-Amores et al. [20] and Batchvarov et al. [10], saturation of the system's dynamics is reached above  $Pe_s \sim 100$ .  $Pe_c$  was set to be equal to  $Pe_s$ , following the study of Agrawal and Neuman [2].

Based on Refs. [20, 46] and our range of characterising  $Oh$ , we define an inertial-capillary or Rayleigh time scale as  $t_{in-cap} = \sqrt{\rho_l D^3 / \sigma_s}$  for  $Oh \ll 1$  and a Marangoni time scale as  $t_m = \sqrt{(\mu_l D) / \Delta\sigma}$ , where  $\Delta\sigma = \sigma_s - 0.05\sigma_s$  is a measurement of the surface tension gradients. The ranges for the characteristic time scales of the system are estimated to be:  $t_{ad} \sim O(10^{-4} - 10^0)$  s,  $t_{des} \sim O(10^{-3} - 10^{-1})$  s,  $t_{in-cap} \sim O(10^{-3} - 10^{-2})$  s,  $t_{conv} \sim O(10^{-3} - 10^{-2})$  s, and  $t_m \sim O(10^{-4})$  s (for surfactant-laden cases). These time scales ensure that sorptive/desorptive, inertial, capillary, and Marangoni phenomena will play an important role in the dynamics of the system.

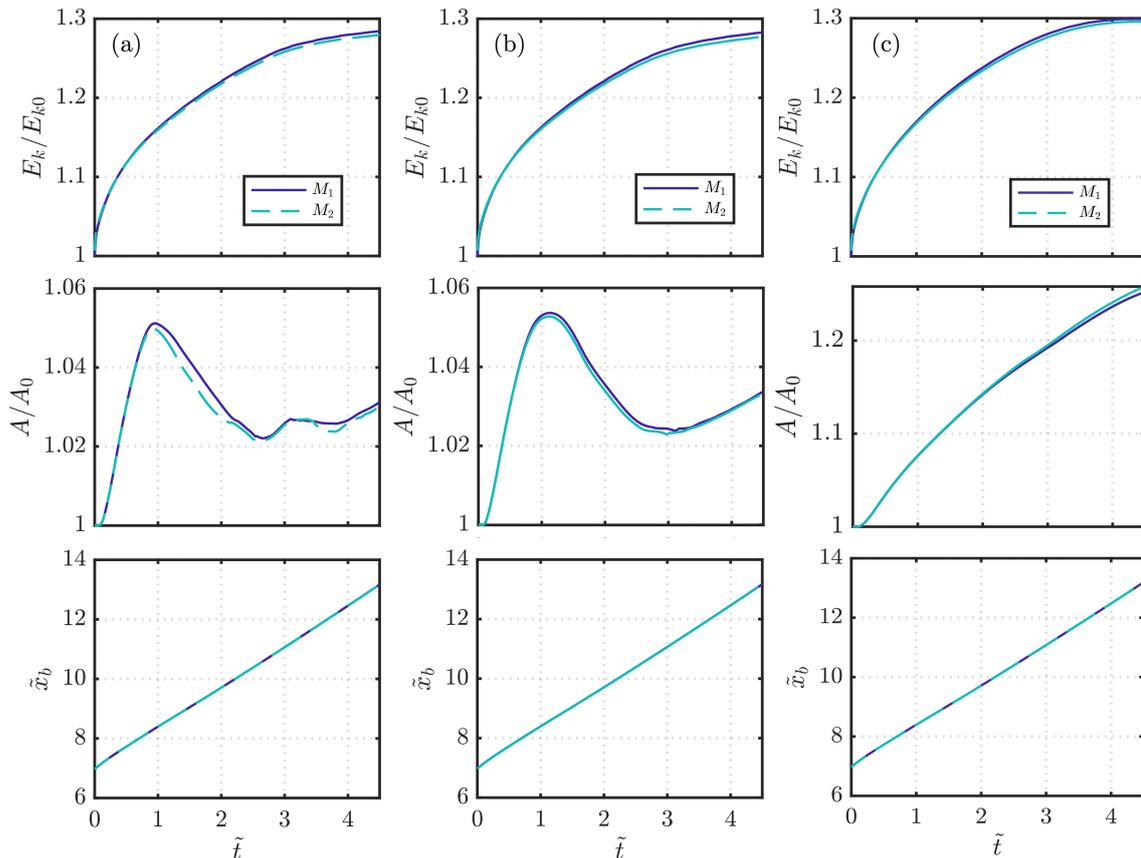


FIG. 12: Results of mesh independence study for surfactant-free and surfactant-laden cases. (a) Case 0 (surfactant-free). (b) Case 1. (c) Case 2

### Appendix B: Mesh independence and resolution analysis

For the assessment of our results' (in)dependency on the mesh, we have tested two uniform Cartesian grids with varying resolutions on both surfactant-free and surfactant-laden cases. The operating conditions for all testing cases are those that allow the development of the liquid jet and drop encapsulation phenomenon, thus evaluating the resolution on small velocity and time scales. Figure 12 depicts the temporal evolution of the kinetic energy,  $E_k = \int_V (\rho \mathbf{u}^2)/2 dV$ , the interfacial area, and the position of the bubble nose,  $\tilde{x}_b$ , the former two being normalised by their values at  $\tilde{t} = 0$ . The tested mesh resolutions correspond to  $M1 = 3456 \times 128 \times 128$  and  $M2 = 6912 \times 256 \times 256$ , and the cases to surfactant-free for 'Case 0' and surfactant-laden with  $Da = 1$ ,  $k = 0.1$  for 'Case 1' and  $Da = 0.1$ ,  $k = 1$  for 'Case 2'. All other parameters remain unchanged from those specified in §II B. Table I reports a few additional features of the tested mesh resolutions, including the minimum cell size,  $\Delta x_{min}$ , the maximum relative variation of the gas phase volume,  $\Delta V_g = (V_g - V_{g0})/V_{g0}$ , and that of the liquid phase,  $\Delta V_l = (V_l - V_{l0})/V_{l0}$ . Considering the adaptive nature of our time-step treatment (see §II A), we highlight that modifying the mesh resolution alters the simulation time-step in accordance to the cell size. For Case 1, we observed  $\Delta t \approx 8 \times 10^{-7}$  s and  $\Delta t \approx 4 \times 10^{-7}$  s for M1 and M2, respectively. The almost complete overlapping of the plots displayed in figure 12 for the grids considered and the three sets of operating conditions suggests that increasing the resolution by twofold on each direction does not lead to the development of significantly different dynamics. Unless stated otherwise, all simulations have been run with M1 given that it provides enough resolution to ensure mesh-independent results, while also requiring substantially less computational resources, as seen in table I. This table also indicates that M1 allows us to achieve volume conservation for both phases with a deviation of  $8 \times 10^{-3}$  % or lower from the onset of the simulations.

An additional mesh independence test was carried out to specifically evaluate the pinch-off dynamics' sensitivity to the mesh resolution. For this test, we contrast the time taken to reach the curvature inversion,  $t_{inv}$  (described in §III A), and the back and interior pinch-off times,  $\tilde{t}_{p-o,bk}$  and  $\tilde{t}_{p-o,int}$ , respectively, for Case 0 and Case 1. Table II depicts these values for the two mesh resolutions considered. We note that both  $\tilde{t}_{p-o}$  are reported with  $\tilde{t}_{inv}$  as the reference frame. As a complement to the pinch-off times, we compare in table III the size of the entrapped drops at selected times after the pinch-off event and before their stabilisation (see figures 7-8(b)). These results indicate that increasing the mesh resolution by a factor of 8 does not alter

TABLE I: Additional features of the mesh resolutions tested

Case name	Mesh	Computing time/CPU to reach 10 ms (h)	$\Delta x_{min}$ ( $\mu\text{m}$ )	$\max(\Delta V_g)$ (%)	$\max(\Delta V_l)$ (%)
Case 0	M1	$\sim 0.60$	7.81	$7.70 \times 10^{-3}$	$1.04 \times 10^{-3}$
	M2	$\sim 8.46$	3.91	$-6.13 \times 10^{-3}$	$8.27 \times 10^{-4}$
Case 1	M1	$\sim 0.78$	7.81	$7.65 \times 10^{-3}$	$3.02 \times 10^{-4}$
	M2	$\sim 12.71$	3.91	$3.84 \times 10^{-3}$	$5.19 \times 10^{-4}$
Case 2	M1	$\sim 2.21$	7.81	$8.00 \times 10^{-3}$	$2.61 \times 10^{-4}$
	M2	$\sim 7.34$	3.91	$7.28 \times 10^{-3}$	$9.83 \times 10^{-4}$

TABLE II: Recorded  $\tilde{t}_{inv}$ ,  $\tilde{t}_{p-o,bk}$ , and  $\tilde{t}_{p-o,int}$  for the two mesh resolutions tested. The values of  $\Delta\tilde{t}$  represent the percent change of  $\tilde{t}$  going from M1 to M2

	$\tilde{t}_{inv}$			$\tilde{t}_{p-o,bk}$			$\tilde{t}_{p-o,int}$		
	M1	M2	$\Delta\tilde{t}_{inv}$	M1	M2	$\Delta\tilde{t}_{p-o,bk}$	M1	M2	$\Delta\tilde{t}_{p-o,int}$
Case 0	0.122	0.120	-1.964%	2.158	2.098	-2.762%	2.328	2.265	-2.696%
Case 1	0.117	0.120	2.389%	2.904	2.829	-2.593%	2.647	2.563	-3.129%

the dynamics at the locality of the pinch-off more than 3.5% for surfactant-free and surfactant-laden cases.

The mesh size selected also allowed to capture accurately the thin liquid film surrounding the bubble, which, according to previous works [38, 55], requires a minimum of 5-10 computational cells covering its domain for a correct development of the liquid velocity profile. In our simulations, we have ensured the placement of at least 5-6 cells in the liquid film section for the cases that exhibit encapsulation ( $We \sim 30$ ), for which  $h(x)/D \sim 5^{-2}$  (see figure 11 of Magnini and Matar [55]). This condition is further achieved in the surfactant-laden simulations, which develop a thicker liquid film. We make the note, however, that the size of the entire domain, as well as the comprehensive sweep over dimensionless numbers presented, make it prohibitively expensive in terms of computation to achieve the refinement levels reported in works such as Castrejón-Pita et al. [16], Li and Sprittles [51] to fully capture all regime transitions leading to the interfacial singularities of pinch-off.

### Appendix C: Validation in square capillaries

A supplementary validation for square capillary channels is conducted using the numerical data and conditions described in Magnini and Matar [55] for surfactant-free scenarios under non-negligible inertia. Satisfactory quantitative agreement between our results and those of Magnini and Matar [55] is achieved in terms of bubble-to-liquid velocity ratio,  $U_b/U_l$ , and gas area fraction,  $\varepsilon$  (see figure 13(a)-(b)), with maximum deviations of 2.7% and 3.1%, respectively. We underline that the cases that do not have a direct counterpart from the above mentioned reference (i.e.,  $Ca = 0.0089$ ,  $Ca = 0.0377$ , and  $Ca = 0.068$  for  $Re = 443$ ) adequately follow the qualitative trend of increasing  $U_b/U_l$  and decreasing  $\varepsilon$  with  $Ca$  and  $Re$ .

Figures 13(c)-(d) exemplify the well-known bubble morphological regimes found in non-circular channels, as well as our model's correct capturing of the non-axisymmetric bubble shapes that arise below a threshold  $Ca$  [17, 40, 50]. In accordance with Ferrari et al. [32], the corner flow effects exhibited in  $Ca \lesssim 0.01$  promote the development of a saddle-shaped interface nearby the channel centre lines (see outermost series in figures 13(c)-(d)), which brings about the formation of four lobes projecting towards the channel corners. As portrayed in figure 13(c), these lobes are located furthest away from the channel

TABLE III: Interfacial area ( $A_d$  in  $\text{mm}^2$ ) of encapsulated drop post pinch-off. The values of  $\Delta A_d$  represent the percent change of  $A_d$  going from M1 to M2

	Case 0				Case 1			
	$\tilde{t}$	2.979	3.422	3.865	4.308	2.984	3.427	3.870
$A_d$ M1	0.800	0.762	0.755	0.754	0.713	0.709	0.709	0.709
$A_d$ M2	0.777	0.747	0.739	0.741	0.693	0.693	0.697	0.694
$\Delta A_d$	-2.943%	-1.898%	-2.242%	-1.714%	-2.805%	-2.257%	-1.693%	-2.116%

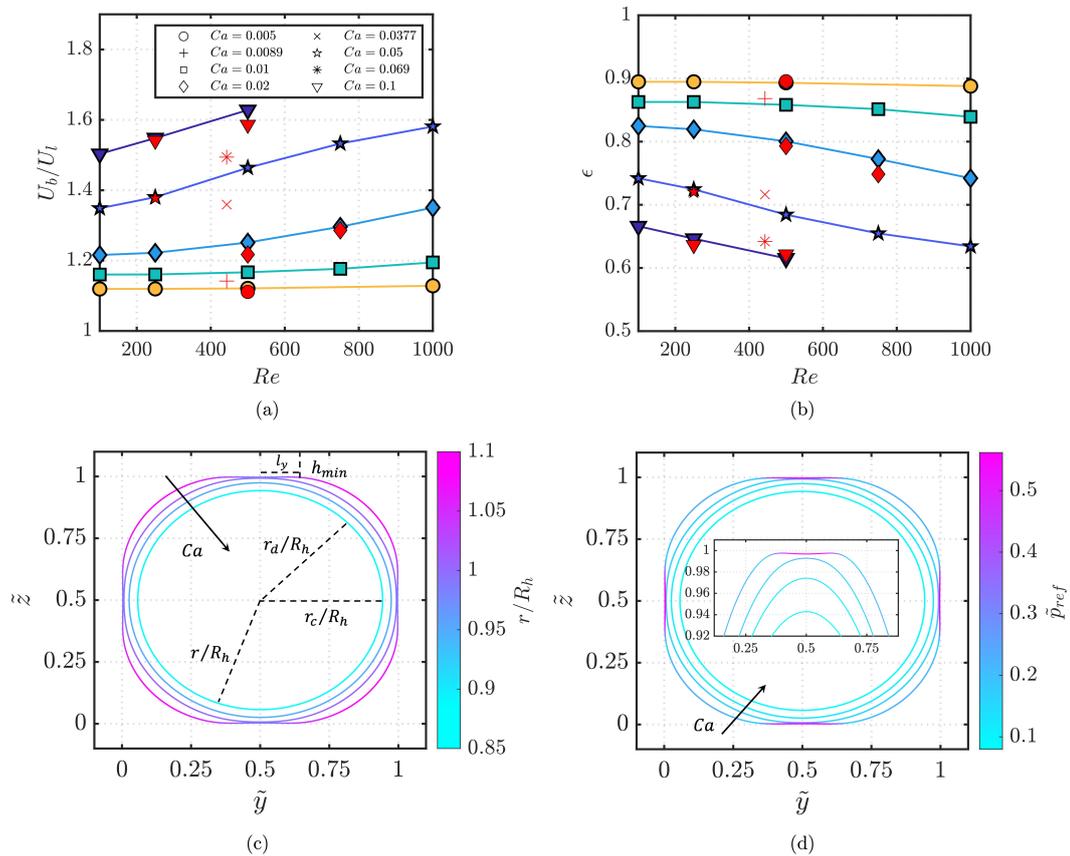


FIG. 13: Validation for square capillary channels and  $We > 1$  in terms of (a) bubble nose velocity relative to average liquid phase velocity, and (b) gas phase area fraction. Each marker corresponds to a different  $Ca$  (as seen from the legend). The colour red represents our numerical results and the remaining colours Magnini and Matar [55]’s results. (c)-(d) Replication of the bubble morphological structures proposed in the same reference with our numerical simulations in terms of (c) distance from channel centre to any point of the interface normalised by the channel’s hydraulic radius,  $R_h$ , and (d) normalised relative pressure,  $\tilde{p}_{ref} = (p - p_b)/\rho_l U_a^2$ . From outermost to innermost series, we show our results for:  $Ca = 0.0089$ ,  $Ca = 0.02$ ,  $Ca = 0.0377$ , and  $Ca = 0.1$  at  $Re = 443 - 500$ . Results in (b)-(d) are measured at a cross-sectional plane normal to the stream-wise direction located at a distance of  $5.5D$  behind the bubble nose, as reported in Magnini and Matar [55]

centre-point (highest  $r/R_h$ ) and will continue to grow as the saddle deepens ( $r_d > r_c$ ) with decreasing  $Ca$ . The presence of the saddle shifts the minimum liquid film thickness,  $h_{min}$ , from the centre-line of the channel to a position,  $l_y$ , that varies as a function of  $Ca$ . We compute  $l_y/R_h = 0.2$  for the case highlighted in the figure, which matches remarkably well with the value reported by Magnini and Matar [55] for  $We \ll 1$  ( $l_y/R_h \approx 0.195$ , see figure 7(b) of the reference). The high pressure zones at the saddles showcased in figure 13(d) are evidence of significant capillary flow drainage away from the thin-liquid film region, as reported in Magnini and Matar [55].

The other morphological regimes represented in figures 13(c)-(d) correspond to an additional non-axisymmetric cross-section regime for  $0.01 \lesssim Ca \lesssim 0.05$ , characterised by  $r_d > r_c$  and the absence of the saddle ( $l_y = 0$ , see  $Ca = 0.02$  in the figure), and a fully symmetric bubble topology ( $r_d = r_c$ , see  $Ca = 0.1$  in the figure) for the final regime. We draw attention to the almost perfect alignment of these regimes and the ranges of  $Ca$  in which they occur between our numerical predictions and those of the literature, notwithstanding that the regimes documented in the literature were observed under conditions of vanishing inertia.

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