Effects of soluble surfactant on lateral migration of a bubble in a pressure driven channel flow

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\section*{Abstract}

The effects of soluble surfactant on the lateral migration of a bubble in a pressure-driven channel flow are examined by interface-resolved numerical simulations. The interfacial and bulk surfactant concentration evolution equations are solved fully coupled with the incompressible Navier-Stokes equations. A nonlinear equation of state is used to relate interfacial surface tension to surfactant concentration at the interface. Extensive computations are performed to investigate the bubble dynamics for a wide range of parameters. It is found that surfactant dramatically changes the bubble dynamics. In the clean case, the bubble position depends on its deformability, characterized by the Eötvös (Eo) and the capillary (Co) numbers. The spherical bubble moves towards the wall, while the deformable one migrates away from it. On the other hand, in the presence of the surfactant, even the spherical bubble moves away from the wall. It is also found that the contaminated bubble stays away from the wall for Eo ≤ 0.1 and Eo ≥ 1.5 while it migrates towards the wall for 0.1 ≤ Eo < 1.5. Also, at high Eo, the onset of path instability is observed for both the clean and the contaminated cases. However, adding surfactant to the system triggers the path instability earlier and amplifies the oscillations afterwards.

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\section*{1. Introduction}

Surfactants are often naturally present or externally added into an interfacial system (Levich, 1962; Stone, 1994). It has been shown that even a small amount of surfactant can lead to dramatic structural changes of a bubbly flow, i.e., changes in bubble rise velocity, lateral migration, bubble-bubble interactions and mass transfer (Takagi \textit{et al.}, 2008). These structural changes can be utilized to perform useful functions in different applications such as microfluidics (Sjoblom, 2005; Tabeling, 2010; Anna and Mayer, 2006), medicine (Grotberg, 2001) and industrial processes (Takagi and Matsumoto, 2011). For instance, surfactants play a pivotal role in the safety and efficiency of the equipments in nuclear reactors and bubble columns. It is thus of significant importance to understand the effects of surfactants on the movement of a bubble.

The relevance to natural and many industrial applications has made the dynamics of an air bubble rising in a stagnant and flowing liquid an active research area. It is well known that surfactant causes a reduction in the rising velocity of a bubble (Clift \textit{et al.}, 2005), which is explained by the so-called Marangoni effect (Frumkin and Levich, 1947). A fully contaminated spherical bubble behaves as a rigid sphere in water, as the drag coefficient of a spherical bubble approaches that of a rigid sphere when a sufficient amount of surfactant is present. This phenomenological explanation was first given by Frumkin and Levich (1947). Cuenot \textit{et al.} (1997) has provided a comprehensive review regarding this subject until 1996. Later, Takemura and Yabe (1999) used the stagnant cap model to explain the mechanism of the transient process where the bubble behavior changes from that of a fluid sphere to that of a solid particle. Almatroshi and Borhan (2004) examined the effects of surfactant on the buoyancy-driven motion of bubbles and drops in a vertical tube experimentally. They observed the surfactant retardation effect for the cases of small air bubbles and viscous drops. Tasoglu \textit{et al.} (2008) numerically examined the effect of soluble surfactants on the unsteady motion and deformation of a bubble rising in an otherwise quiescent liquid contained in an axisymmetric tube by using a finite-difference/front-tracking method. They considered nearly spherical, ellipsoidal, and dimpled ellipsoidal-cap regimes of bubble motion. They have shown that...
the surfactants generally reduce the terminal velocity of the bubble, especially in the nearly spherical regime where bubble behaves similar to a solid sphere and its terminal velocity approaches that of an equivalent solid sphere. Tripathi et al. (2015a) examined the motion of a buoyancy-driven bubble in a so-called ‘self-rewetting fluid’. They showed that the motion of a spherical bubble in the Stokes flow limit can be arrested in a self-rewetting liquid due to thermocapillary Marangoni stresses.

Recent studies on bubble motion show that the presence of surfactant has drastic effect not only on the vertical motion but also on the lateral migration of a bubble (Fukuta et al., 2008). Fukuta et al. (2008) examined the behavior of the lift force acting on a contaminated bubble in a linear shear flow. They showed that the lift force acting on a spherical bubble decreases from the value of a clean bubble to near zero with an increase of Langmuir number. Takagi et al. (2009) showed that addition of surfactant drastically alters the behavior of an upward bubbly flow. They performed experiments to investigate the effects of species and concentration of surfactant on the flow structure. They found that in the absence of surfactant, some bubbles coalesced during the rising process and thus formed bubbles of larger size (deformable bubbles) which then moved toward the channel center instead of the channel wall. An addition of a small amount of 3-Pentanol surfactant prevented the bubble coalescence and bubbles remained nearly spherical, thus bubbles moved toward to the wall mainly due to aero-dynamic lift force. However, as the amount of 3-Pentanol surfactant is increased, nearly spherical bubbles moved toward the center of channel due to the Marangoni stresses. Muradoglu and Tryggvason (2014) developed a front-tracking method for simulation of soluble surfactant in 3D multiphase flows. They studied the effects of soluble surfactants on the motion of buoyancy-driven bubbles in a straight square channel in deformable (spherical) and deformable (ellipsoidal) regimes. They also simulated a few cases of lateral migration of bubbles in a pressure-driven channel flow with a soluble surfactant and found that the contaminated bubble drifts away from the channel wall and stabilizes at the center of the channel when the surfactant-induced Marangoni stresses are sufficiently large. However, their focus was on the numerical method rather than the flow physics. More recently, Lu et al. (2017) examined the effect of insoluble surfactant on the structure of turbulent bubbly upflow in a vertical channel by direct numerical simulations (DNS). They showed that, in the presence of surfactant, contaminated bubbles migrate to the center, whereas clean bubbles move towards the wall. Hayashi and Tomiyama (2018) investigated numerically the effect of surfactant on lift coefficients of bubbles in linear shear flows by using the interface tracking method. They showed that the lift coefficient of a bubble decreases as the Langmuir number increases. Therefore, surfactant decreases the critical values of $E_o$ and $Re$ at which the coefficient changes its sign.

The effects of surfactant on lateral migration of a droplet and a bubble have been also studied in the creeping flow regime. Pak et al. (2014) used a reciprocal theorem to examine the effects of insoluble surfactant on lateral migration of a spherical droplet in an unbounded Poiseuille flow in the limit of vanishing Reynolds and surface Pelet common numbers. They showed that surfactant-induced Marangoni stresses cause cross-streamline migration of the droplet towards the centre of the Poiseuille flow and the magnitude of the lateral migration depends linearly on the distance of the drop from the centre of the Poiseuille flow. Das et al. (2018) studied analytically the combined effect of bulk-insoluble surfactant and a varying temperature on the motion of a viscous droplet in a shear flow. Neglecting the fluid inertia, they found that the droplet speed and direction of motion are strongly influenced by the interfacial transport of surfactant. Das and Chakraborty (2018) performed a numerical and analytical study to examine the effect of surface viscosity on the motion of a surfactant-laden droplet in a non-isothermal Poiseuille flow. Neglecting the fluid inertia, they found that the droplet migration velocity is notably affected by the di-latational viscosity, whereas the effects of the shear viscosity are negligible.

It is well known that the path of a rising bubble in a bulk liquid experiences a path instability, i.e., bubble motion changes from straight to zigzag/spiral when bubble size exceeds a critical value (Haberman and Morton, 1953). Kelley and Wu (1997) showed that the path of circular air bubble rising in a viscous fluid of a Hele-Shaw cell changes from a straight path to a zigzag one when the Reynolds number of the bubble exceeds a critical value. They observed that this path instability is a consequence of the vortex shedding behind the bubble. Zenit and Magnaudet (2008) experimentally studied the conditions for which the paths of freely rising bubbles become oscillatory. They argued that the dominant parameter to trigger the instability is the bubble shape rather than Reynolds number of the bubble and that the path instability is a direct consequence of the wake instability. Tripathi et al. (2015b) performed numerical simulations on a spherical bubble rising in a quiescent fluid. They observed five distinct regimes (namely, axisymmetric, skirted, zigzagging/spiralling, peripheral breakup, and central breakup) with sharply defined boundaries in the Galilei number (Ga)-Eötvös number (Eo) phase plot. Sharaf et al. (2017) performed extensive experiments over the entire range of $Ga$ and $Eo$ reported by Tripathi et al. (2015b) and provided a similar phase plot in the $Ga – Eo$ plane. However, they found one major difference, that is, unlike five different regions of bubble behavior in the numerical phase plot, only four regions were identified experimentally while a central breakup (region V) was not observed. Balla et al. (2018) have also observed path instability for a non-isothermal rising bubble in a so-called ‘self-rewetting’ fluid.

Surfactant affects the path instability of a rising bubble. Tagawa et al. (2014) experimentally examined the effect of surfactant on the path instability of a rising bubble. They categorized the bubble motion as straight/helical/zigzag on the basis of bubble Reynolds number and the normalized drag coefficient. Recently, Pesci et al. (2018) studied the effects of soluble surfactant on dynamics of a single bubble rising in a large spherical domain. They found that the bubble path depends both on the initial surface and bulk contamination. Another observation was that a quasi-steady state velocity of a bubble can be attained without reaching an equilibrium between ad- and desorption of surfactants.

In the present study, extensive numerical simulations are performed to examine the effects of surfactant on the lateral migration and the path instability of a bubble in a pressure driven flow at a finite Reynolds number. We consider both the soluble and insoluble surfactant cases. Simulations are performed to investigate bubble dynamics for a wide range of parameters including the capillary number ($Ca$), the elasticity number ($β_s$), the bulk surfactant concentration ($C_s$) and the Eötvös number ($Eo$).

2. Formulation and numerical method

Following Muradoglu and Tryggvason (2014), a single set of equations can be written for the entire computational domain provided that the jumps in the material properties such as density, viscosity and molecular diffusion coefficient are correctly treated and the effects of surface tension are included properly in the momentum equations. In the one-field formulation, the Navier-Stokes equations for the whole domain can be written as follows:

$$\frac{∂ρ\mathbf{u}}{∂t} + \nabla \cdot (ρ\mathbf{u}\mathbf{u}) = -\nabla p + \nabla \cdot (\tau) + gΔ ρ + \int_{\Gamma} [σ(\Gamma)\mathbf{n} + \nabla σ(\Gamma)] δ(\mathbf{x} - \mathbf{x}_i) dA, \quad (1)$$
where \( \mathbf{u}, p \) are the velocity vector and pressure fields, respectively, \( \mathbf{g} \) is the gravitational acceleration vector and \( \rho \) and \( \mu \) are the discontinuous density and viscosity fields, respectively. The effects of surface tension are included as a body force in the last term on the right hand side, where \( \sigma \) is the surface tension coefficient that is function of the surfactant concentration \( \Gamma \) at the interface, \( \kappa \) is twice the mean curvature, and \( \mathbf{n} \) is a unit vector normal to the interface. \( \nabla \sigma (\Gamma) \) represents the surfactant-induced Marangoni stresses where the surface gradient is defined as
\[
\nabla_{\delta} = \nabla - \mathbf{n} (\mathbf{n} \cdot \nabla).
\]
(2)
The surface tension acts only on the interface as indicated by the three-dimensional delta function \( \delta \) whose arguments \( \mathbf{x} \) and \( \mathbf{x}_I \) are the point at which the equation is evaluated and a point at the interface, respectively. The Navier-Stokes equations are supplemented by the incompressibility condition
\[
\nabla \cdot \mathbf{u} = 0.
\]
(3)
We also assume that the material properties remain constant following a fluid particle, i.e.,
\[
\frac{D\rho}{Dt} = 0; \quad \frac{D\mu}{Dt} = 0.
\]
(4)
where \( \frac{D}{Dt} \) is the material derivative. The density and viscosity vary discontinuously across the fluid interface and are given by
\[
\rho = \rho_o I(\mathbf{x}, t) + \rho_i \left( 1 - I(\mathbf{x}, t) \right),
\]
(5)
\[
\mu = \mu_o I(\mathbf{x}, t) + \mu_i \left( 1 - I(\mathbf{x}, t) \right),
\]
(6)
where the subscripts "o" and "i" denote the properties of the bubble and bulk fluid, respectively, and \( I \) is the indicator function defined as
\[
I(\mathbf{x}, t) = \begin{cases} 
1 & \text{in bulk fluid}, \\
0 & \text{in bubble fluid}. 
\end{cases}
\]
(7)
The surface tension is a function of the surfactant concentration and determined by the Langmuir equation of state
\[
\sigma = \sigma_o \left[ 1 + \beta_s \ln \left( 1 - \frac{\Gamma}{\Gamma_{\infty}} \right) \right],
\]
(8)
where \( \beta_s = \frac{\partial \sigma}{\partial \Gamma} \) is the elasticity number. Eq. (8) is slightly modified to avoid negative values of the surface tension (Muradoglu and Tryggvason, 2008; 2014)
\[
\sigma = \sigma_o \left[ \max \left( \epsilon_\sigma, 1 + \beta_s \ln \left( 1 - \frac{\Gamma}{\Gamma_{\infty}} \right) \right) \right],
\]
(9)
where \( \epsilon_\sigma \) is taken as 0.05 in the present study. The evolution equation for the interfacial surfactant concentration was derived by Stone (1990). Following Muradoglu and Tryggvason (2014) the interfacial surfactant is solved on the Lagrangian grid and can be expressed as
\[
\frac{D\Gamma}{Dt} = AD_{\Gamma} \nabla^2 \Gamma + AD_{\Gamma},
\]
(10)
where \( A \) is the surface area of an element of the interface, \( D_\Gamma \) is the diffusion coefficient along the interface and \( S_\Gamma \) is the source term given by
\[
S_\Gamma = k_{\text{a}} C_i (\Gamma_{\infty} - \Gamma) - k_{\text{d}} \Gamma,
\]
(11)
where \( k_{\text{a}} \) and \( k_{\text{d}} \) are adsorption and desorption coefficients, respectively, and \( C_i \) is the concentration of the surfactant in the bulk fluid immediately adjacent to the interface. The bulk surfactant concentration is governed by the advection-diffusion equation in the form
\[
\frac{\partial C}{\partial t} + \nabla \cdot (C \mathbf{u}) = \nabla \cdot (D_{\text{co}} \nabla C),
\]
(12)
where the coefficient \( D_{\text{co}} \) is related to the molecular diffusion coefficient \( D \), and the indicator function \( l \) as
\[
D_{\text{co}} = D_\Gamma l(\mathbf{x}, t).
\]
(13)
The source term in Eq. (11) is related to the bulk concentration by Eggleton and Stebe (1998)
\[
S_\Gamma = -D_{\text{co}} (\nabla C)_{\text{interface}}.
\]
(14)
The boundary condition at the interface given by Eq. (14) is first converted into a source term for the bulk surfactant evolution equation. In this approach it is assumed that all the mass transfer between the interface and the bulk takes place in a thin adsorption layer adjacent to the interface. Thus, the total amount of mass adsorbed on the interface is distributed over the adsorption layer and added to the bulk concentration evolution equation as a negative source term in a conservative manner. Eq. (12) thus takes the form as
\[
\frac{\partial C}{\partial t} + \nabla \cdot (C \mathbf{u}) = \nabla \cdot (D_{\text{co}} \nabla C) + S_\Gamma.
\]
(15)
where \( S_\Gamma \) is the source term evaluated at the interface and distributed onto the adsorption layer in a conservative manner. The details of this treatment can be found in Muradoglu and Tryggvason (2014, 2008). The flow Eqs. (1) and (3) are solved fully coupled with the bulk (15) and interfacial (10) surfactant concentration evolution equations using the finite-difference/front-tracking method (Muradoglu and Tryggvason, 2014; Tryggvason et al., 2001; 2011; Unverdi and Tryggvason, 1992). The momentum and the continuity equations are solved on a stationary staggered Eulerian grid and are discretized by second-order central difference approximations for the spatial derivatives except for the convective terms in the momentum and the bulk surfactant concentration equations that are discretized using the QUICK and the fifth order WENO-Z schemes, respectively. Time integration is done by a second order predictor-corrector method (Muradoglu and Tryggvason, 2014). The incompressibility condition leads to a non-separable Poisson equation for pressure which is solved using the parallel semi-coarsening multi-grid (PFMG) solver in the HYPRE library (Falgout et al., 2006). Detailed description of the numerical method can be found in Muradoglu and Tryggvason (2014, 2008).

3. Problem statement and computational setup

Fig. 1 (a) schematically depicts the lateral migration of the contaminated bubble in a pressure driven flow and Fig. 1(b) shows the computational setup. As shown in Fig. 1(b), a spherical bubble of
diameter \( d_b \) is placed instantly near the left wall with the center initially located at \((x_c, y_c, z_c) = (2.615d_b, 1.29d_b, 1.38d_b)\). The interfacial surfactant concentration is initialized as clean (\( \Gamma = 0 \)) or contaminated (\( \Gamma = \Gamma_{eq} \)) for soluble and insoluble surfactants, respectively, whereas the bulk concentration is uniform at \( C = C_{eq} \). The flow is initialized assuming a pressure driven fully developed laminar channel flow with the average channel velocity given by

\[
V_0 = \frac{1}{12\mu_o} \left( - \frac{dp}{dz} \right) H^2, \tag{16}
\]

where \( H \) is the channel width and \((-dp/dz)\) is an applied constant pressure gradient. The computational domain is rectangular, bounded by two vertical walls at which the no-slip boundary conditions are applied. The periodic boundary conditions are used in the spanwise \((x)\) and the streamwise directions \((z)\). The domain size is \(5.236d_b \times 6.676d_b \times 10.472d_b\) and is resolved by a \(128 \times 192 \times 256\) grid in the spanwise \((x)\), wall-normal \((y)\) and streamwise \((z)\) direction, respectively. The grid is slightly stretched near the walls in the wall-normal \((y)\) direction. The average distance \(y_m\) also called the separation distance, is defined as the distance between the wall and the bubble center. The Reynolds numbers based on the bubble diameter and channel width are set to \(Re_x = \rho \nu d_b / \mu = 150 \) and \(Re_z = \rho \nu d_b / \Gamma = 1000\), respectively. For all the simulations the density \(\rho \) and viscosity \(\mu\) ratios are set to \(10\). Note that a typical liquid-air system has an order of magnitude larger density and viscosity ratios than used here. However, it has been previously demonstrated (Tasgolu et al., 2010; Muradoglu and Tasgolu, 2010; Olgac et al., 2013) that higher property ratios do not affect the results significantly. Therefore, the property ratios are kept small in order to enhance the numerical stability, relax the time-step restrictions and thus to reduce the computational cost.

In the present study, all simulations are performed using dimensional quantities but the results are expressed in terms of relevant non-dimensional quantities. The diameter of bubble \(d_b\) and average channel velocity \(V_0\) are used as length and velocity scales, respectively, and \(\Theta = d_b/V_0\) as the time scale to define various governing non-dimensional numbers as summarized below:

\[
Re_x = \frac{\rho_o V_0 d_b}{\mu_o}; \quad Re_z = \frac{\rho_o V_0 H}{\mu_o}; \quad Ca = \frac{\mu_o V_0}{\sigma_o}; \quad Eo = \frac{\sigma_o}{\rho_o \mu_o} d_b^2; \quad \rho_o = \frac{\rho}{\rho_o}; \quad \mu_o = \frac{\mu}{\mu_o}; \\
M = \frac{\mu_o}{\mu}; \quad \sigma_o = \frac{\sigma}{\sigma_o}; \quad Gd = \frac{\rho_o \mu_o \sigma_o}{\mu_o \sigma_o}; \quad \beta = \frac{\Gamma_{x\infty}}{\mu_o \sigma_o}; \quad \beta_s = \frac{RT \Gamma_\infty}{\sigma_o}, \tag{17}
\]

where \(Re_x, Re_z, Ca, Eo, \rho, \mu, \sigma, \beta, \beta_s\) are the Reynolds number based on the bubble diameter, the Reynolds number based on channel width, the capillary number, the Eötvös number, the Morton number, the Galilei number, the Peclet number based on bulk surfactant diffusivity, the Peclet number based on interface surfactant diffusivity, the Biot number, the Damköhler number, the dimensionless adsorption depth and the elasticity number, respectively.

Following Bunner and Tryggvason (2003), the bubble deformation is quantified by the deformation parameter defined as square root of the ratio of the largest and the smallest eigenvalues of the second moment of inertia tensor, i.e.,

\[
deformation = \sqrt{I_{\max}/I_{\min}}, \tag{18}
\]

where \(I_{\max}\) and \(I_{\min}\) are the maximum and minimum eigenvalues of the second moment of inertia tensor, given by

\[
l_{ij} = \frac{1}{V_b} \int_{V_b} (x_i - x_0)(x_j - x_0) \, dV. \tag{19}
\]

where \(V_b\) is the volume of the bubble and \(x_0\) and \(x_0\) are the coordinates of the bubble’s centroid in the \(i\)th and \(j\)th directions, respectively. Bunner and Tryggvason (2003) have shown that deformation quantification by this method is approximately equal to the ratio of the smallest to the longest axis for modestly deformed ellipsoids. For more complex bubble shapes, deformation quantification used here is a more general definition of the bubble deformation than the ratios of the longest and the shortest axis and eliminates any uncertainty in identifying those axis.

4. Results and discussion

Simulations are performed to investigate the effects of soluble surfactant on the lateral migration of a bubble by using fully parallelized three-dimensional front-tracking method (Farooqi et al., 2019). Each simulation is performed by using 24 and 8 cores for the domain and front, respectively. Muradoglu and Tryggvason (2014) performed an extensive grid convergence study of the present numerical method for a similar setting, therefore it is not repeated here. Following Muradoglu and Tryggvason (2014), the base line parameters are chosen as \(Re_x = 1000, Eo = 0, \beta = 0.5, \beta_s = 200, k = 0.125, Bi = 1.2\), and \(Da = 6.67\). The parameters are fixed to the base line values unless indicated otherwise. The results are first presented for the non-buoyant case to separate effects of gravity and surfactants. Then the results are presented for the buoyant case. We note that the Morton number is \(M = 1.85 \times 10^{-2}\) for \(Eo = 1.5\) in the buoyant case which is higher than the Morton number of \(M = 2.52 \times 10^{-11}\) for an air bubble in water at 20°C but could be matched by using a water-glycerin (W-G) solution (Legendre et al., 2012). For this water-glycerin solution, \(Eo = 1.5\) used here corresponds to a bubble with diameter of 0.9 mm. The channel width is 6.6 \(d_b\) or 6 mm and our channel Reynolds number corresponds to a bulk velocity of 2.7 m/s.

4.1. Non-buoyant case (\(Eo = 0\)).

Initially, simulations are performed to investigate the effects of the capillary number on the dynamics of a clean and a contaminated bubble (soluble surfactant) by varying \(Ca\) between 0.01 \(\leq Ca \leq 0.5\), while keeping all other parameters fixed at their values in the base line case. The non-dimensional average distance from the wall \(y_m/d_b\) and the bubble deformation are shown in Fig. 2 against the dimensionless time \(\Theta\).

A clean spherical bubble experiences a lift force in a direction perpendicular to both the direction of motion and the rotation axis. As shown in Fig. 1, the non-uniform fluid flow causes the bubble to rotate, resulting in a force in the negative y direction, i.e., towards the left wall. The resulting lift force on the bubble, also known as the Magnus effect, causes the lateral migration of the spherical bubble towards the left wall. Indeed, Fig. 2(a) shows that the bubble moves towards the wall for \(Ca \leq 0.2\), while it moves towards the channel center for \(Ca = 0.5\). The lateral migration of the bubble towards the wall is mainly due to the aerodynamic lift force. As the bubble becomes more deformable for \(Ca = 0.5\), the non axisymmetric shape of the bubble changes the direction of rotation of the bubble (Ervin and Tryggvason, 1997) and thus the bubble moves away from the wall in the direction of lift force maintaining higher separation distance. This steady separation distance is the function of the capillary number which in turn is a measure of the bubble deformation: the larger \(Ca\), the more the bubble deforms and the longer is the steady separation distance. Ervin and Tryggvason (1997) also supported our observation by reporting that the lift coefficient of a bubble becomes negative when the bubble deforms strongly. Similar observations have been also made recently by Aghalib et al. (2019) for hyperelastic fluid particles. Now we move on to consider the contaminated case, depicted by solid lines.
Fig. 2. Effect of capillary number (Ca) on the average distance from the wall (a) and the bubble deformation (b). $Re_c = 1000$, $\beta_i = 0.5$, $Pe_s = Pe_c = 200$, $k = 0.125$, $Bi = 1.2$, $Da = 6.667$.

Fig. 3. Effect of the soluble and insoluble surfactant on (a) the average distance from the wall (b) deformation of the bubble. $Re_c = 1000$, $\beta_i = 0.5$, $Pe_s = Pe_c = 200$, $k = 0.125$, $Bi = 1.2$, $Da = 6.667$.

Fig. 4. Evolution of contaminated bubble in the pressure-driven flow for $Ca = 0.5$: The insoluble surfactant case (top row) and the soluble surfactant cases with the initially clean ($\Gamma_{\text{initial}} = 0$) (middle row) and initially uniform at the equilibrium ($\Gamma_{\text{initial}} = \Gamma_{eq}$) (bottom row) distributions. $Re_c = 1000$, $\beta_i = 0.5$, $Pe_s = Pe_c = 200$, $k = 0.0$, $Bi = 0.0$, $Da = 6.667$. 
in Fig. 2(a). For the contaminated bubble, the presence of the surfactant changes the dynamics of the bubble completely. The non-uniform distribution of interfacial surfactant concentration leads to a variation of surface tension along the bubble surface, which causes tangential shear stress on the surface, known as Marangoni stresses. This Marangoni stressess counteract and dominate over the aerodynamic lift force for all Ca considered in this work. As a result, the contaminated bubble moves towards the channel center for all Ca.

The bubble deformation against time is shown in Fig. 2(b). As expected, the bubble deformation increases with Ca at all times for both clean and contaminated cases, except for the highest capillary number (Ca = 0.5) in the clean case. For all bubbles, the deformation first sharply increases due to high shear stresses near the wall, then relaxes to its steady state values. After the initial spike, the deformation of the contaminated bubble decreases with time due to the fact that the bubble moves towards the channel center, where the shear force is reduced. On the other hand, the clean bubble for Ca < 0.2 drifts towards the wall, where the higher shear forces deform it more until a steady state separation distance is achieved. At Ca = 0.5 however, the clean bubble moves towards the centerline and hence the deformation decreases with t∗, similarly to the contaminated case.

Next, simulations are performed to examine combined effects of the solubility of the surfactant and the capillary number. The interfacial surfactant concentration is initially set to the equilibrium concentration, \( \Gamma_0 = \frac{1}{\Gamma_{\infty}} \Gamma_{\infty} \), for the insoluble case while the bubble is initially clean in the soluble case. In the insoluble case, the source term is also switched off to avoid mass exchange by setting the adsorption \( k_a \) and the desorption \( k_d \) coefficients to zero in the source term given in Eq. (11). The evolution of the non-dimensional separation distance \( y_0/d_0 \) and the bubble deformation are depicted against the normalized time \( t' \) for both soluble and insoluble cases as shown in Fig. 3. Note that, although not shown in Fig. 3, simulations are also performed for the soluble cases with the interfacial surfactant concentration initialized uniformly at \( \Gamma_{\infty} \) and the results are found to be essentially the same as the initially clean cases except for the very early transient period of time, i.e., approximately for \( t' < 20 \). As can be seen in Fig. 3, the bubble moves towards the channel center for both cases, but the separation distance in the case of insoluble surfactant is larger. The difference in the separation distance is negligible at \( Ca = 0.01 \) and \( Ca = 0.05 \), while it increases at higher \( Ca \). We observe that the time evolution of the deformation parameter remains very similar for both soluble and insoluble cases. To explain the differences in wall separation, the evolution of interfacial insoluble and soluble surfactant concentration for the most deformable bubble (Ca = 0.5) is shown in Fig. 4. Snapshots are taken at the non-dimensional times of \( t' = 0, 0.0, 16.6, 66.6 \) and 116.6. Initially at \( t' = 0 \), the interfacial surfactant concentration is uniformly distributed over the surface of the bubble for the insoluble case, whereas, for the soluble one, the bubble surface is either clean (the second row) or uniform at \( \Gamma_{eq} \) (the third row). At \( t' = 16.6 \), the contaminated bubble strongly deforms for all the cases as it moves away from the wall. It can be seen that the amount of surfactant advected to the nose of the bubble is higher for the insoluble case. This leads to larger Marangoni stresses and thus faster transient lateral migration for the insoluble case at intermediate times, leading to a larger wall separation distance. As the bubble approaches an equilibrium condition at later times, the interfacial surfactant concentration distribution becomes similar for both insoluble and soluble cases. Fig. 4 shows that the initial interfacial
surfactant concentration in the soluble case has a negligible effect on the surfactant distribution at the interface and thus on the lateral migration of the bubble except for the very early transient period of approximately $t^* < 20$.

Then, the effect of the elasticity number $\beta_s$, is considered in the surfactant concentration as shown in Fig. 5. The physico-chemical parameter $\beta_s$ determines the sensitivity of interfacial tension to variations in surfactant concentration, i.e., the larger $\beta_s$, the stronger the surfactant. Fig. 5 shows that for $\beta_s = 0$ (clean), the bubble moves towards the channel wall and maintains a certain steady separation distance. Increasing the elasticity number to $\beta_s = 0.1$ triggers a change in bubble dynamics. At early times, the bubble maintains the same separation distance, but eventually migrates to the center of the channel. Increasing further the elasticity number to $\beta_s = 0.25$, the direction of lateral migration completely changes at initial times and the bubble migrates away from the wall. For $\beta_s > 0.25$, we observe similar results. The contours of the interfacial surfactant for $\beta_s = 0.1$ and $\beta_s = 0.25$ are also shown in the inset of Fig. 5 at $t^* = 50$. It can be seen that the interfacial surfactant distribution at $\beta_s = 0.25$ is more uniform than that at $\beta_s = 0.1$. This is due to the rigidifying effect of surfactant, that is, increasing $\beta_s$ increases the
Marangoni stresses which causes the reduction in surface mobility of the bubble. A similar trend is observed for the deformation of the bubble. For the clean bubble ($\beta_s = 0.0$), the deformation increases due to higher shear stresses at the wall. At $\beta_s = 0.1$, the deformation is initially nearly constant in time, until the bubble starts drifting to the channel center, where the deformation decreases. Further increasing the elasticity number ($\beta_s \geq 0.25$), the deformation of the contaminated bubble first oscillates and then decreases, as the bubble moves towards the channel center.

Additionally, the effect of the bulk surfactant concentration in the range of $0 \leq C_\infty \leq 1$ is examined for a nearly spherical bubble ($Ca = 0.05$ and $\beta_s = 0.5$), as shown in Fig. 6. In the clean case ($C_\infty = 0$), the bubble moves towards the wall in agreement with the earlier observations. It is worth noting that at $C_\infty = 0.05$, the direction of the migration remains the same, although the final separation distance is larger and consequently it takes longer to reach the steady state. At $C_\infty = 0.1$, the bubble maintains nearly the same separation distance until $t^* < 70$ while it starts drifting to the channel center at later times. Further increasing the concentration to $C_\infty \geq 0.25$, the contaminated bubble moves towards the channel center and maintains a certain steady separation distance. To gain further insight, the bubble with the interfacial surfactant concentration contours are plotted in the inset of Fig. 6 for three values of $C_\infty = 0.05, 0.1$ and $0.25$ at $t^* = 50$. In the case of $C_\infty = 0.05$, the amount of the surfactant adsorbed on the surface is negligible thus the aerodynamic lift force dominates over the Marangoni stresses. As a result, the direction of the migration is the same as the clean bubble. For $C_\infty = 0.1$, the surface is more contaminated leading to larger Marangoni stresses which grow in time. The contaminated bubble maintains the same separation distance at early times and then eventually, migrates to the center of the channel at later times. Furthermore, in the presence of sufficient amount of the surfactant at $C_\infty = 0.25$, the surface of the bubble is contaminated significantly, and the Marangoni force dominates the aerodynamic lift force; hence, the contaminated bubble drifts away from the wall. These results are consistent with the experimental findings of Takagi et al. (2009) on the effect of surfactants on the bubbly flow. Evolution of the deformation in the range of $0 \leq C_\infty \leq 1$ is also shown in Fig. 6 (right). The deformation initially oscillates at high shear stresses near the wall, then increases for $C_\infty \leq 0.05$ and decreases for $C_\infty \geq 0.1$ before reaching a steady value. The deformation decreases with $C_\infty$, as the bubble migrates towards the region of low shear stresses with $C_\infty$.

4.2 Buoyant case ($E_0 \neq 0$).

Here, the effect of the Eötvös number is examined for clean and contaminated bubbles. Simulations are performed for $E_0$ ranging from 0 to 3, as shown in Fig. 7. For the clean case, it is observed
that the bubble moves towards the wall for all $E_o$, except for the lightest (most deformable) bubble at $E_o = 3$, as shown in Fig. 7(a).

It is interesting to observe that the effect of $E_o$ on the steady separation distance is less than effect of $Ca$. At $E_o = 3$, the aerodynamic lift force reverses its direction and thus, bubble migrates towards the center of the channel. It starts oscillating called path instability (Pesci et al. (2018)) as it approaches to the center of the channel, which will be discussed in more detail below. On the other hand, the effect of $E_o$ is more significant on the contaminated case as shown in Fig. 7(c). At $E_o = 0$, the contaminated bubble migrates towards the centerline. However, the behavior of the contaminated bubble changes completely with an increase of $E_o$. For $E_o < 1$, the bubble migrates towards the wall, maintaining a slightly larger separation distance than in the corresponding clean case. This is due to the buoyancy-induced increase in the velocity of the bubble that results in a higher aerodynamic lift force. At $E_o = 1.5$, the contaminated bubble initially moves towards the wall but then eventually migrates to the center of the channel, where it starts oscillating. At $E_o = 2.0$, the direction of the lateral migration is reversed and hence the bubble migrates towards the center and maintains a certain steady separation distance. Path instability is observed for the cases of $E_o = 2.5$ and $E_o = 3.0$ as the bubble approaches the center of the channel as seen in Fig. 7(a). Deformation of the clean and the contaminated bubbles is shown in Fig. 7(b) and (d), respectively. As expected, the deformation increases with $E_o$ for both cases. Also, it can be seen that the deformation of the contaminated bubbles is lower than that of the clean bubbles irrespective of their average distance from the wall. This is attributed to the rigidifying effect of surfactant on the contaminated bubbles.

To understand the distinct behavior of two contaminated bubbles at $E_o = 0.5$ and 1.5, bubbles with contours of interfacial surfactant are plotted in Fig. 8 at $t^* = 100$. Note that the bubble dynamics is critical beyond this time as shown in Fig. 7(c). Indeed, as can be seen in Fig. 8a, the contaminated bubble at $E_o = 0.5$ is nearly spherical and the surfactant concentration is almost uniform leading to small Marangoni stresses. The aerodynamic lift force dominates the force due to surfactants-induced Marangoni force, as a result the bubble maintains the same separation distance in time. On the other hand at $E_o = 1.5$, for the contaminated bubble the surfactant concentration is more non-uniform, that is, more surfactant is advected to the rear end leading to larger Marangoni stresses. Thus, the Marangoni stresses overcome the lift force and contaminated bubble migrates to the channel center as the time progresses. To give more insight on the effects of $E_o$, bubbles at $E_o = 0.1, 2$ and 3 with constant contours of interfacial surfactant concentration are shown in Fig. 9 at initial, intermediate and later times (at approximate steady state). Initially at $t^* = 8.3$, the adsorption of surfactant at $E_o = 0$ is much higher than the other cases, therefore the contaminated bubble instantly migrates towards the center unlike in other cases where it first migrates towards the wall. At intermediate time, the bubble at $E_o = 0$ and $t^* = 83.3$ stays spherical but with high concentration at the front, drifting the bubble away from the wall. On the other hand, the bubble at $E_o = 1$ and $t^* = 83.3$ is deformable and the distr-
bution of surfactant concentration is almost uniform, maintaining the bubble close to the wall. On the contrary, for the bubbles at $Eo = 2$ ($t^* = 89.4$) and $Eo = 3$ ($t^* = 75$), the surfactant has accumulated at the bottom of the bubbles due to strong convection, and approaches an axisymmetric distribution as the contaminated bubbles migrate to the center of channel. Note that the bubble behaves similarly at later times. The contaminated bubble at $Eo = 0$ also migrates to the center but the interfacial surfactant concentration is different than the $Eo = 2$ and $Eo = 3$ cases, that is, surfactant is advected to the nose of the bubble rather than the bottom of the bubble.

Finally, the effects of the $Eötvös$ number on the path instability of the clean and the contaminated bubbles are studied. The three-dimensional trajectory of the bubble for $0 < Eo \leq 3$ is shown in Fig. 10. Note that for the clean case, the path instability is only observed at highest $Eötvös$ number considered in this work, i.e., at $Eo = 3$. As can be seen, after the clean bubble reaches the center of the channel, its motion changes from straight to zigzag path. Zenit and Magnaudet (2008) experimentally studied the conditions for which the path of freely rising bubble becomes oscillatory. They found that the criteria of path instability of a bubble with a free-slip surface is not the Reynolds number but the aspect ratio $A > 2.0$ of the bubble shape. It is interesting to observe that the deformation parameter for $Eo = 3$ is less than $Eo = 2$ as shown in Fig. 7(b). There are several possible reasons for this discrepancy. Firstly, the present work considers a rising bubble in a pressure driven flow rather than the freely rising bubble considered by Zenit and Magnaudet (2008). Secondly, there is a clear difference in the definition of the deformation parameter (see Eq. (19)). Tripathi et al. (2015b) and Sharaf et al. (2017) performed numerical and experimental studies, respectively, to generate a phase plot in the Galilei and $Eötvös$ numbers plane, which separates distinct regimes in terms of bubble behavior. According to their phase plots, the clean case at $Eo = 3$ also lies in the oscillatory regime.
(region III). In the presence of surfactant, the critical \( E^{\#}_{\text{St}} \) number \( (E_{\text{St}}) \) at which the path instability is triggered is affected significantly. In the current work, addition of surfactant leads to decrease in the critical \( E^{\#}_{\text{St}} \) number from \( E_{\text{St}} \approx 3 \) for the clean case to \( E_{\text{St}} \approx 1.5 \) for the contaminated case. Another difference is that the contaminated bubble at \( E_{\text{St}} = 1.5 \) is in a spiral motion rather than the zigzag motion observed for the clean case. Further increasing the \( E^{\#}_{\text{St}} \) number to \( E_{\text{St}} = 2.0 \) and \( E_{\text{St}} = 2.5 \) leads to a zigzag path instability. Finally at \( E_{\text{St}} = 3 \), the path initially changes from straight to zigzag and later to helical/spiral. Note that, this zigzag-helical transition is also observed for freely rising clean bubbles in the experimental study of Shew et al. (2006) and numerical study of Mougin and Magnaudet (2001). The bubble velocity at \( E_{\text{St}} = 3 \) for both clean and contaminated cases along the rising direction \( z \) and \( x - y \) plane is shown in the Fig. 11. As can be seen, the quasi-steady state terminal velocity of the contaminated bubble along rising \( z \) direction is lower than that of the clean bubble due to the Marangoni stresses which increase the drag force of the rising bubble. On the contrary, the terminal velocity of contaminated bubble along \( x - y \) plane is much higher than that of the clean bubble due to the Marangoni stresses. Thus, the increase in the magnitude of oscillation and reduction of the critical parameters at which path instability is triggered can be attributed to the effect of Marangoni stresses. Fig. 12 shows the vortical structures for the clean \( (E_{\text{St}} = 3) \) and the contaminated \( (E_{\text{St}} = 2.5 \) and \( E_{\text{St}} = 3) \) cases, at different time instants. As can be seen, for the zigzag motion of the bubbles the wake structure consists of two counter-rotating vortices with a symmetry plane and at each cycle (from one peak of \( y_{\mu} \) to another) the two vortices interchange their signs. Note that the zigzag motion is observed for both cases \( (E_{\text{St}} = 2.5 \) and \( E_{\text{St}} = 3) \) at all time instances, except for \( t^* \geq 489.4 \) at \( E_{\text{St}} = 3 \) where the contaminated bubble follows helical trajectory. For the clean case at \( E_{\text{St}} = 3 \), the vortical structures decay with time and almost vanish at \( t^* = 381.2 \), as the bubble stabilizes at later times. On the other hand, for the contaminated case at \( E_{\text{St}} = 2.5 \), the vortical structures do not vanish with time, while the bubble continues to oscillate indefinitely. For the contaminated bubble \( (E_{\text{St}} = 3) \), the vortical structure changes when its motion changes from zigzag to helical at non-dimensional time \( t^* = 489.4 \). For the helical motion, two counter-rotating vortices of opposite sign are formed that wrap around each other as shown in Fig. 12. Fig. 13 shows the interfacial surfactant concentration and orientation of the contaminated bubble at the same non-dimensional times at which the vortical structures are shown. As can be seen the orientation of the bubbles is tilted towards the direction of motion for zigzag motion and at such instances the Marangoni-induced force would act in the same direction resulting in the enhancement of horizontal oscillation magnitude as observed in Fig. 11(a).

5. Conclusions

A finite-difference/front-tracking (FD/FT) method is used to examine the effects of soluble surfactant on the lateral migration of nearly spherical and deformable bubbles in a pressure-driven channel flow. It is found that a small amount of surfactant can alter direction of lateral migration of a bubble. The clean bubble moves toward the wall and maintains a steady separation distance depending on the deformability of the bubble. On the contrary, the contaminated bubble migrates toward the center of the channel and its steady separation distance is nearly independent of the deformability of the bubble. By comparing soluble and insoluble surfactant cases for different capillary numbers, it is observed that the lateral migration of the bubble is faster in case of soluble surfactant than the soluble surfactant during the transient period and this difference in both cases increases with the capillary number.

The effects of the bulk surfactant concentration \( (C_{\infty}) \) and the elasticity number \( (\beta_i) \) on the lateral migration of the bubble are examined by extensive simulations. It is found that in both cases, the bubble migrates toward the center of channel after a critical value of \( C_{\infty} = 0.25 \) and \( \beta_i = 0.25 \).

Finally the effect of \( E^{\#}_{\text{St}} \) number on the lateral migration is examined. For the clean case, the bubble moves away from the wall only at high \( E_{\text{St}} = 3 \), whereas it migrates towards it at smaller values. In the presence of surfactant, the bubble reverses the direction of lateral migration for \( E_{\text{St}} \leq 0.1 \) and \( E_{\text{St}} \geq 1.5 \), while it moves towards the wall for the values in between. Path instability is observed for rising bubbles in both clean and contaminated cases. It is observed that the surfactant reduces the critical values at which a path instability is triggered.

Declaration of Competing Interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

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Supplementary material

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