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Bubbles and Broth: A review on the impact of broth composition on bubble column bioreactor hydrodynamics

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ABSTRACT

The growing global population and heightened concern for climate change leads to increased interest in utilizing microbial fermentations to replace polluting production processes for e.g., plastics, fuels, and animal proteins. Computational fluid dynamics (CFD) is a valuable tool for accelerating the scale-up and optimization of large-scale bioprocesses. However, the design correlations underlying most of these CFD models are validated with air-water systems, not accounting for the distinct hydrodynamic properties of microbial fermentation broth. In this review, we provide an extensive overview of the current understanding of how various biotechnologically relevant solutes impact the hydrodynamics of bubble columns. We examine the effects of components found in fermentation broths, including salts, surfactants, viscoelastic solutes, alcohols, acids, ketones, sugars, biomass, and proteins, on mass transfer, bubble formation, bubble interactions, and flow regime transitions. These components all exhibit unique effects, yet their combined influences remain poorly understood. Future research should prioritize identifying the concentration at which coalescence inhibition occurs for different compounds, especially in mixtures, and exploring the role of proteins in bubble column hydrodynamics from micro- to macroscale.

1. Introduction

Industrial bioprocesses can play a key role in a carbon-neutral economy, for example through replacement of fossil fuel streams and replacement of animal-based meats. The power of microorganisms can be harnessed to create plastics and fuels from plant-based feedstock, CO2, or wastewater.

However, scale-up of novel processes and maximization of resource efficiency can prove challenging: changes in nutrient distribution and interphase mass transfer (e.g., local gas transfer) can change the performance of scaled up processes for better or worse, which can have severe economic consequences. While there is substantial historical research on the impact of broth composition on specific bioprocesses [1–3], classical 0-D and 1-D bioreactor models predicting e.g. gas holdup and mass transfer are typically based on air-water systems [4,5] or make broad generalization into 'coalescent' and 'non-coalescent' fluids [6,7] without detailed attention to specific compositional details. Such correlations often come with error margins of 15% or more when applied in their domain of validity [8–11]. More advanced modelling efforts such as computational fluid dynamics (CFD) suffer from the same ailments, where most physical models used in CFD are validated with air-water systems, for which an adequate amount of information and modelling approaches are available, or on heuristic assumptions/extrapolation of lab observations on key parameters such as the bubble size. These assumptions introduce substantial uncertainty in process design, especially for novel bulk bioprocesses with tight margins. In some cases, ignoring the impact of broth properties can result in much higher performance prediction differences: offsets of 300% in mass transfer capacity may result from application of models outside their domain of validity [12].

Aside from affecting bubble size and area through coalescence inhibition, the specific properties of broth and the solutes present in fermentation broths can impact the rise velocity of bubbles, especially of bubbles with an equivalent radius of around 1 mm. It was already proposed by Clift et al. in 1978 [13] to use different rise velocity correlations for pure and contaminated water (Fig. 1). Furthermore, larger variations in bulk liquid properties can lead to changes in bubble shapes as indicated in the Clift-Grace-Weber diagram (Fig. 2) [13]. Other hydrodynamic aspects, such as the flow regime and with that mixing efficiency, may also change because of broth property changes [14–16].

Improved insight in the impact of broth properties on gas-liquid

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Nomenclature

a	Interfacial area m^2/m^3	
C_D	Drag coefficient $[-]$	
$\overline{d_B}$	Mean bubble diameter m	
d_{32}	Sauter mean bubble diameter m	
d_B	Bubble diameter m	
D	Diffusivity m^2/s	
D_C	Column inner diameter m	
g	Gravitational constant m/s^2	
H_L	Unaerated liquid height m	
k_L	Liquid side mass transfer coefficient s^{-1}	
$u_{g,s}$	Superficial gas velocity m/s	
V_B	Bubble rise velocity m/s	
x	Mole fraction $[-]$	
ε_g	Gas holdup $[-]$	
ϵ	Turbulent energy dissipation rate W/kg	
μ_L	Liquid viscosity $Pa{\cdot}s$	
ρ	Density kg/m^3	
σ	Surface tension N/m^2	

hydrodynamics and mass transfer in bioprocesses is required for the reliable development, scale-up and optimization of novel bioprocesses. This is especially true considering the current attention for gas fermentation based on C_1 and H_2- substrates, and the ambition to replace low-cost, large-scale fossil commodities. Furthermore, these insights can help optimize more traditional aerobic fermentations, where the dissolved oxygen concentration is one of the limiting factors for further intensification. In this review, we aim to summarize the effects of common constituents of fermentation broth on bubble size, gas holdup, flow regime transitions and mass transfer in bubble column bioreactors.

2. Dimensionless numbers

Weber number:

$$We = \frac{\rho_L * v_s * d}{\sigma}$$

Reynolds number:

$$Re = \frac{\rho_L \cdot d_e \cdot v}{\mu_L}$$

Eötvös number:

$$Eo = \frac{g \cdot d_e^2}{\sigma} \cdot \Delta\rho$$

Morton number:

$$Mo = \frac{g \cdot \mu_L^4}{\rho_L^2 \cdot \sigma^3} \cdot \Delta\rho$$

3. Hydrodynamics in bubble columns

Bubble column reactors are simple and relatively cheap to construct and can operate at lower power input than stirred tanks for broths with a water-like viscosity (around $1 mPa{\cdot}s$) [5]. As such, we expect bubble columns and other gas-lift reactors to play an important role in bulk biobased processes. In a bubble column, the rising gas induces liquid circulation patterns facilitating liquid mixing, while also introducing gaseous substrate. While we focus on bubble columns, the impact of broth properties on micro- and mesoscale hydrodynamic processes will apply to other multiphase reactor configurations (including stirred tank bioreactors) and should be accounted for accordingly in modelling and design.

3.1. Flow regimes (macroscale)

The hydrodynamics in bubble columns are often described in terms of the flow regimes, typically the homogeneous flow regime, charac-
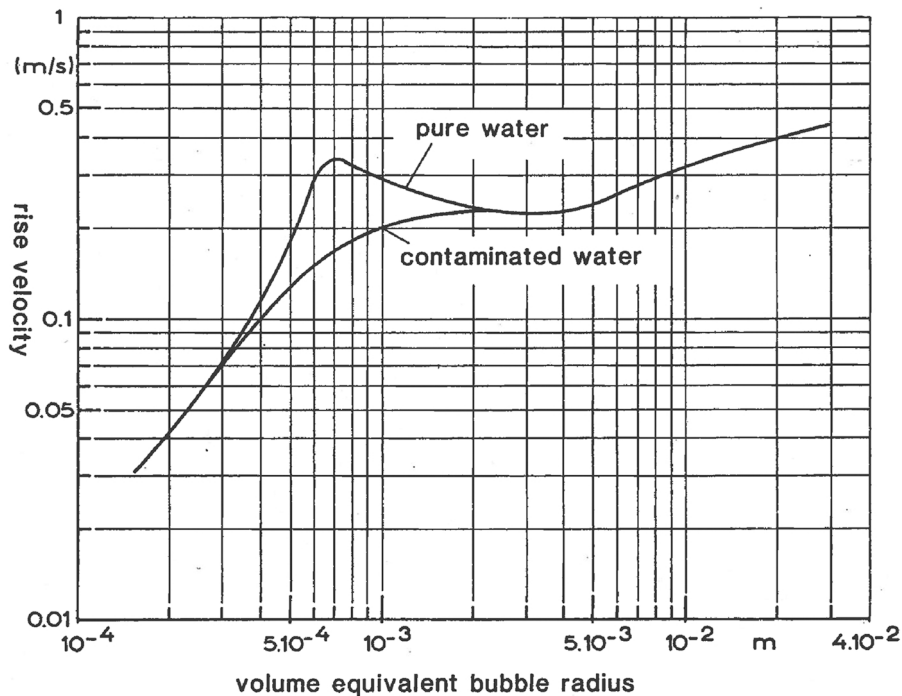

Fig. 1. Size-dependent rise velocities of bubbles in pure and contaminated water. Figure from [13].

terized by a narrow bubble size distribution (BSD) and well-defined flow patterns, and the heterogeneous regime, characterized by chaotic flow and a broad BSD. The transition from homogeneous to heterogeneous flow depends on several factors, most notably the superficial gas velocity ($u_{g,s}$); for air-water under standard conditions, a transition velocity of $0.04 m/s$ is typically reported [17]. The sparger geometry plays a role in the transition, with coarse spargers leading to heterogeneous flow at any $u_{g,s}$ [18,19], while with highly controllable spargers, homogeneous flow can be maintained at higher $u_{g,s}$ up to $0.07 cm/s$ in water [20]. Addition of surfactants can further increase the stability of the homogeneous flow regime [21]. While most studies report two flow regimes, more recently up to three extra flow regimes have been proposed to describe the transition from homogeneous to heterogeneous flow [22]. For columns with an internal diameter less than 15 cm, the slug flow regime can be observed at superficial gas velocities above 5 cm/s [23], and even lower velocities in the case a of highly viscous liquid phase [24]. However, for industrial bubble columns with diameters in the order of meters, this regime is irrelevant, and we will not discuss it in this review.

To understand the transition from the homo- to the heterogeneous flow regime, it is important to note that bigger bubbles induce a more chaotic flow pattern in their wake. This leads to more bubble collisions, which in turn increases coalescence. The resulting larger bubbles will then induce even more chaotic flow patterns, creating a self-reinforcing

system, up to the point where the local turbulence is strong enough to break up the biggest bubbles. If the initial formation of large bubbles can be suppressed, the homogeneous flow regime can be extended far beyond the normal transition velocity [20]. As initial bubble size and coalescence both play a critical role in the transition, it is not surprising that broth components and sparger design are observed to play a role in the transition to heterogeneous flow [1,25–30].

The flow regime has a substantial impact on bioprocess design and operation. In the homogeneous regime, bubbles are of uniform size and follow similar paths, leading to relatively few collisions and a BSD which is mainly dependent on initial bubble size at the sparger. The homogeneous regime is characterized by a gas holdup that is proportional to the superficial gas velocity; high overall gas holdup can be maintained due to the well-defined flow with a quasi-static liquid phase which is favorable maximizing the interfacial area concentration a. Robust baseline CFD models are available for homogeneous flow due to the well-defined flow patterns and easy experimental access [31,32]. In these models, the initial bubble size needs to be set accordingly, and correction factors for the drag force to account for the drag-modifying properties of surfactants may be required to obtain accurate results [25,33,34].

In the heterogeneous regime, the wide BSD induces a chaotic liquid flow pattern. The mutual interaction between the moving gas and liquid
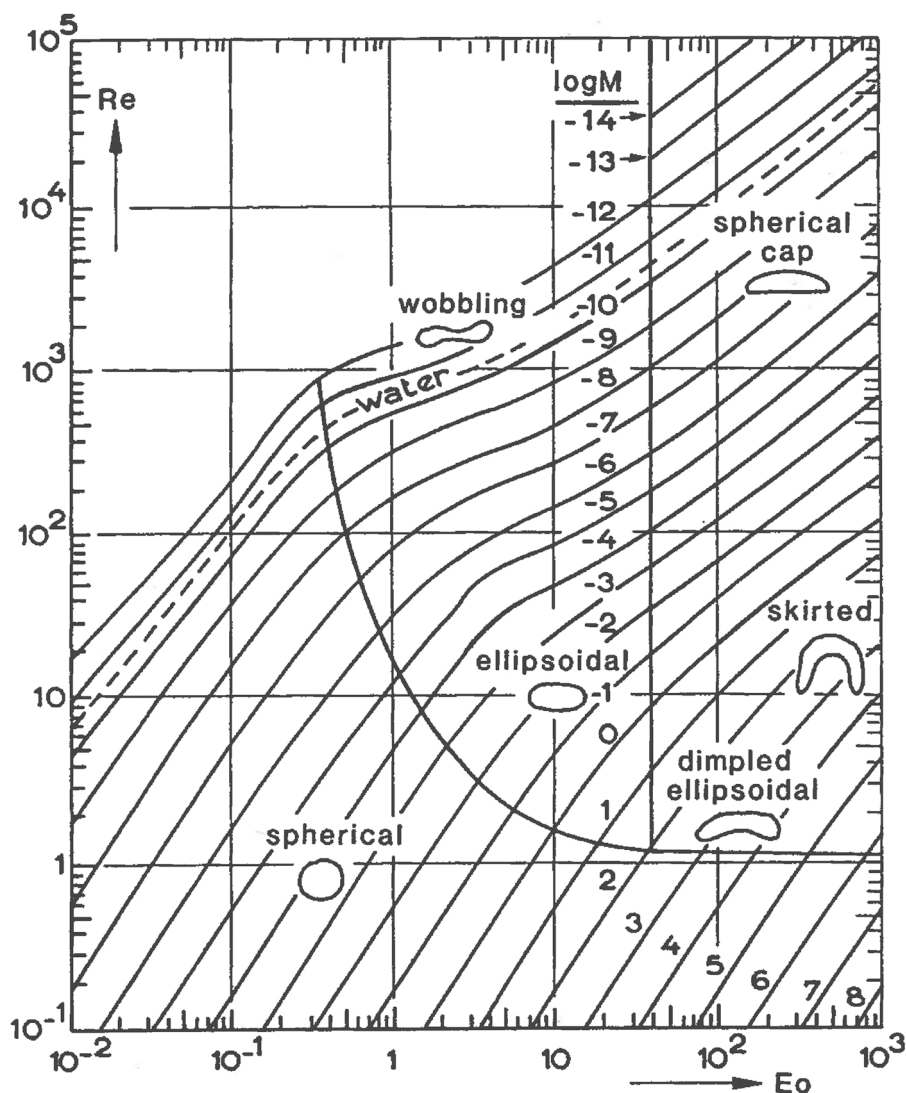

Fig. 2. Clift-Grace-Weber diagram predicting bubble shapes from Reynolds, Eötvös and Morton numbers [13].

phase results in a drag reduction in the wake of larger bubbles [28,35], which combined with the higher rise velocity of the larger bubbles results in a reduced gas holdup compared to homogeneous flow. While this has a negative impact on a, heterogeneous flow strongly enhances liquid mixing. This is beneficial in reducing liquid-phase substrate heterogeneity when point-fed liquid phase substrates are involved in the process, although it has the drawback of a lower gas residence time, and therefore potentially lower utilization of gaseous substrates. For air-water systems, several studies provide experimental data on gas holdup [36–39], and provided the column diameter (D_C) > 0.15 m and H_L/D_C ratio > 5, it was observed that hydrodynamic observations, such as d_{32} and gas holdup, translate between scales [4,40,41]. Maximiano Raimundo et al. [41] did note some impact of column diameter on gas holdup, although we hypothesize this may be because they based their $u_{g,s}$ on standard pressure, while the effect of D_C vanishes if $u_{g,s}$ is based on the column-mean pressure, taking into account the reduced volume due to hydrostatic pressure. Under heterogeneous conditions, time-averaged data shows increased radial velocity and holdup in the center of the column, a profile which is typically fitted with higher-order polynomials [41–43]. This profile arises due to the induced liquid motion, and the resultant migration of larger, faster bubbles to the center of the column, while the smaller bubbles move towards the walls, an effect that is described by the lift force [44,45]. The hydrodynamic modelling of heterogeneous gas-liquid flow is considerably more complex than for homogeneous flow, due to the need to account for changes in bubble size, for example using population balances [46]. While the use of population balances in CFD is well-established [33,47,48], most approaches base gas-liquid interaction on the Sauter-mean diameter, which inherently does not account for the different impact of the lift force on different size classes [49]. Per-class accounting of forces is possible, albeit at great computational expense [50,51]. Consequently, most studies accounting for size-dependent force descriptions opt for simplified hydrodynamic descriptions. Besides these modelling complexities, experimental heterogeneous flow studies focus largely on air-water systems, and simulation studies similarly so.

The current models cannot be generalized to fermentation broths, where the impact of changes in the BSD and interphase forces on macroscopic flow behavior is largely unknown. Furthermore, the modifications of breakup and coalescence kernels to describe the effects of solutes on the BSD are still in the early phases of development [52]. Further experimental insight in the manifestation of macroscopic flow behavior in fermentation broths of various composition, as well as the micro- and mesoscopic phenomena giving rise to this, is required to account for these effects in bioreactor design and optimization, and in CFD models describing realistic bioreactor hydrodynamics.

3.2. Bubble formation and mass transfer (microscale)

Gas-liquid mass transfer in bubble columns is dependent on the mass transfer coefficient ($k_L a$), the maximum solubility of the gas (c_L^*) and the (local) dissolved concentration of the gas (c_L). The $k_L a$ is generally given as a single parameter but consists of two parts: k_L, the liquid side mass transfer coefficient, and a, the interfacial area. Many mass transfer studies focus on quantification of the global (reactor-level) mass transfer coefficient $k_L a$, in part due to the complexity of local quantification of $k_L a$, let alone k_L and a separately. The $k_L a$ is often characterized experimentally for oxygen transfer, but this can be used as a proxy for transfer rates of other gases [53]. Comprehensive lists of $k_L a$ correlations for different reactor types can be found in literature [10,17]. Recent works do show renewed interest in the disentanglement of these two parameters in attempts to facilitate more detailed process performance predictions considering the individual impacts of solutes on k_L and a [12,54,55]. Furthermore, as CFD studies allow for the local prediction of the separate parameters based on local bubble size, gas velocity and turbulence, accurate knowledge on local phenomena can facilitate

detailed process optimization, for example with internals or feed point positioning [56–59].

Equations to predict the local k_L can be separated in two categories: Theoretical and empirical. The basic assumptions for the theoretical models, are a stagnant film of liquid around each bubble (Whitman's film model [60]), the exchange of liquid elements between surface and bulk (Higbie's penetration model [61] & Danckwerts' surface renewal model [62]), or a combination of film and exchange (film-penetration model [63]). The first category considers the effect of local turbulence only in a penetration depth, while the Higbie and Danckwerts models both consider an exposure time of each element, either using the local turbulence or the slip velocity. These different models predict vastly different values of k_L, where predictions for the exchange models are 3- to 4-fold higher than those for the stagnant film models [64]. A more detailed overview of both theoretical and empirical models can be found in Garcia-Ochoa & Gomez, 2009 [10]. In bioprocesses, the presence of surface-active compounds to the liquid phase can make the bubble surface more rigid, resulting in a reduced k_L, closely resembling the predictions with stagnant film models [64]. In some cases, addition of just 10 ppm surfactant can lead to 75% reduction in k_L [65]. Furthermore, the k_L models are all proportional to the gas molecule diffusivity in the liquid phase D_g or $\sqrt{D_g}$ of the transported gas molecule, which is affected by temperature and the presence of solutes [66]. Recent computational work on direct numerical simulation (DNS) of mass transfer from single bubbles in the presence of surfactant captures the physical changes at the interface and opens the doors to continuous estimation of mass transfer from simple bubble rise velocity measurements [67].

The interfacial area a inside the bubble column is dependent on the sparger configuration, and then on breakup and coalescence. The sparger type affects the initial bubble size distribution, which further impacts hydrodynamics and mass transfer along the column in both coalescing and coalescence-inhibiting systems [19,22,29,68,69]. In coalescence inhibiting solutions, porous plate spargers lead to higher interfacial area than perforated plates [1]. Capillary banks can be used to fine-tune bubble sizes [20,55], but are generally not used beyond lab-scale experiments.

Bubble breakup was studied extensively in the 1950's by Hinze [70], and a binary model for breakup is available, based on the critical Weber number (We_{crit}). Bubbles with a Weber number much higher than the We_{crit} will break up, while bubbles with a Weber number much lower than We_{crit} remain stable. The fate of bubbles with a Weber number right around We_{crit} remains hard to predict with this model. Recent investigation through experimental work and DNS have found a description of bubble breakage probability around We_{crit} that describes the transition between stable and unstable bubbles with a smooth function, which considers local fluctuations in turbulence intensity [71]. Breakup is then dependent on bubble size, flow characteristics and physicochemical properties of the gas and liquid [52]. As broth components impact viscosity, surface tension and general flow regime, it can reasonably be expected that they will impact breakup rates. These effects will be described in detail below.

Bubble coalescence starts with bubble collisions; whether or not colliding bubbles coalesce is dependent on contact time and film drainage time [72]. These, in turn, are dependent on bubble rigidity (surface tension), viscosity, density of both gas and liquid phase, and surface tension gradient. These properties are clearly influenced by broth composition which will be discussed below.

3.3. Impact of broth components

As noted in the prior section, broth composition affects gas-liquid hydrodynamics and interphase mass transfer on various levels. On the microscale, mass transfer is affected through changes in both interfacial area a and liquid side mass transfer coefficient k_L [33,64,73,74]. On the

mesoscale, broth composition (including rheology) will affect bubble drag due to modification of bubble surface properties [25], as well as swarm behavior through e.g. alteration of the lift force [75] and changes in bubble drag coefficients [34]. The impact of different categories of broth components will be discussed at the bubble interface level, looking at their interaction with gas-liquid mass transfer, then their effects on single bubbles and bubble-bubble interactions. These results will then be linked to the flow regime of the reactor, and all of these will be combined into their impact on process performance. The effects are summarized qualitatively in a reference table for easy lookup (Table 1).

3.3.1. Salts

Salts are added to fermentation broths as sources of key nutrients for the cells in the fermentation and to assure a suitable osmotic pressure of the broth. In defined broths, they are typically present in concentrations of around 10 g per liter, or an ionic strength of 0.01 – 0.3 M. Specific fermentation setups may lead to more extreme ionic strengths. For example, in the case of fermentation of molasses, assuming 50% molasses in the fermentation broth [76], the salt contents (generally reported as ash) could reach ionic strengths of 0.5–1M [77].

The presence of high concentrations of salt in the gas-liquid interface hinders molecular diffusion, reducing k_L in single bubbles [66], but an overall increase in $k_L a$ is observed in larger systems [78,79]. The implied, and observed, main effect of the presence of electrolytes is inhibition of coalescence, leading to a strong increase of interfacial area [14,79–82]. As the presence of salts also increases surface tension, at higher salt concentrations (>25 g/L) a stabilization of larger bubbles is observed [80]. However, in fermentation broths, concentrations generally remain below this level and thus mainly an increase in mass transfer is expected. The reduction of coalescence leads to more stable small bubbles, and therefore delays the transition to the heterogeneous regime to higher superficial gas velocities [26]. This increases a, but reduces liquid mixing performance [83]. The ionic strength of a solution is often proposed as a potential proxy for the concentration at which coalescence inhibition reaches its maximum [79,84,85]. While the conversion to ionic strength can collapse d_{32} or coalescence graphs of most salts onto a single curve, the cases of potassium iodide and sodium iodide show the limits of ionic strength as a proxy [82,86,87]. As can be seen in Fig. 1, the coalescence curve for sodium iodide is shifted to higher concentration and thus to higher ionic strengths compared to the other sodium halides in the graph [87].

Each specific salt has a transition concentration: a representative concentration for the coalescence inhibition effect by that salt. Several definitions of transition concentration circulate in literature. Dependent on author preference and the experimental method used to obtain them, they provide a concentration at which 50%, 75%, 90% or 95% coalescence inhibition is reached, measured either by coalescence events in the case of bubble contact studies or bubble size (d_{32} or $\overline{d_B}$) in the case of BSD measurements [88]. The transition concentration is not just a property of the fluid, it also depends on hydrodynamic conditions, specifically the approach velocity of two bubbles [89,90]. With increased approach velocity, coalescence efficiency decreases, and the transition concentration will be lower.

The coalescence inhibition effect is dependent on the surface localization of the ions composing the salt. If both the positive and negative ion migrate to the surface, or if both preferentially stay in the bulk, coalescence inhibition is strong. If one of the two is surface-enriched, while the other is surface-depleted, coalescence inhibition is much weaker (Fig. 2). Craig et al. [91] first proposed this model, and it was further worked out by Henry & Craig [87], who linked it to the Surface Partitioning Model from Pegram & Record [92]. Recently, these works were extended on, and a method to predict relative coalescence inhibitory strength of ionic solutions was developed by Duignan [93], taking into account surface tension effects of the individual ions. Most of the experimental and modelling efforts to date focus on pure salt solutions; the work of Duignan opens the doors to estimating transition concentrations of mixed salt solutions, which is of great relevance to bioprocesses.

3.3.2. Surfactants

Surfactants encompass all molecules that are surface-enriched. These are generally the hydrophobic and amphiphilic molecules in solution. In bioprocesses, these molecules are introduced through the addition of antifoam, as byproducts of metabolism and lysed cells, and sometimes the fermentation product of interest has surfactant properties itself, with ethanol as a key example [95,96]. Surfactant molecules migrate to the air-water surface and form a layer there; this layer reduces the liquid film mass transfer coefficient k_L by reducing the local diffusion coefficient of gases [33,97,98]. At the same time, the presence of surfactants generally reduces bubble sizes by inhibiting coalescence and promoting breakup [33]. There are exceptions such as Disfoam, which broadens the bubble size distribution and stabilizes large bubbles [25]. Additionally, the presence of surfactants at the bubble surface leads to an alteration of the single bubble rise velocity [97], which will impact gas distribution in the column.

While the most obvious effect of surfactants is a change in the gas-liquid surface tension, this alone cannot explain all observations. For example, solutions of Triton X-100, SDS and CTAB in water with similar surface tension show vastly different effect on gas holdup in a 9.5 cm column, with superficial gas velocities up to 3.5 cm/s [99]: at the same surface tension, SDS and CTAB solutions exhibit a larger increase in gas holdup than Triton X-100. In viscous and non-Newtonian fluids, Triton X-100 is also observed to increase gas holdup; Passos et al. [100] observed that an addition of 1 g/L of Triton X-100 leads to approximately a 35% increase in holdup at a $u_{g,s}$ of 3 cm/s in Xanthan solutions. In the aforementioned study by Anastasiou et al. [99], a big difference is observed between the behavior of Triton X-100 solution and the other two surfactants. It appears that there is a distinct difference between ionic surfactants (SDS and CTAB) and non-ionic surfactants (Triton X-100). Investigation into the effects of 0.01 M of 2-propanol or D-mannitol, or 0.01 v/v % Antifoam Y-30 shows a narrowing of the bubble size distribution upon addition, at both 4 cm/s and 11 cm/s superficial gas velocity [97]. It should be noted that in this study, between 0.3 and 0.5 M of sulfite is always present for oxygen transfer rate measurements. As this affects the bubble size, these results might be skewed, but the general trend fits with other data. In related work [101], the effect of 2-propanol on gas holdup in a system without sulfite is measured. It is observed that addition of 0.0025 M 2-propanol leads to an increase in gas holdup of about 40% at superficial gas velocities up to 30 cm/s.

The effect of several antifoams is investigated in work by McClure [96], where it is clear that Antifoam Y-30, Antifoam O-30, Disfoam and simple vegetable oil all result in a reduction of gas holdup in the regime up to 12 cm/s [96]. The effect is more pronounced at higher gas flow rates, but hardly any difference between the different antifoams is

Table 1
Effects of the studied substances on multiphase flow characteristics in bubble columns.

Substance category		d_B	C_D	V_B	ε	k_L	a	$k_L a$
Salts	αα & ββ	↓	?	↓	↑	↓	↑	↑
	αβ & βα	–	?	–	–	↓	–	–
Surfactants		↓↑	↑↓	↑↓	↑↓	↓	↑↓	↑↓
Viscoelastic solutes	Viscosity	↑	?	↓	↓	?	↓	↓
	Elasticity	↓	?	↓	↑	?	↑	?
Alcohols		↓	↑	↓	↑	↓	↑	↑
Acids		↓	?	↓	↑	?	↑	?
Ketones		↓	?	↓	↑	?	↑	?
Sugars		– /	?	?	?	?	– /	– /
		↓					↑	↑
Biomass		?	?	↑↓	?	↑	?	?/↓
Proteins		↓	?	?	?	?	?	?

noted. Important to note that in these systems, the presence of Na$_2$SO$_3$ also inhibits coalescence.

A potential predictive property for the effect of surfactants on holdup and bubble size, could be the partition coefficient between 1-octanol and water (measured as its logarithm log$_{10}$P). In a single-bubble study [25], it is observed that surfactants with a log$_{10}$P around 0 (equally distributed between octanol and water) result in a reduction of mean bubble size from 6 to 4 mm and increase in gas holdup of up to 30%. Surfactants with a log$_{10}$P below -1 or above 3 result in an increase in mean bubble size and a decrease in holdup. It should be noted that these conclusions are drawn from a small sample of 6 different surfactants, 2 in each category (low, neutral, high), at a single concentration each (0.02 M or 0.01 vol%). Results from [99] with Triton X-100 (log$_{10}$P of the monomer is 4.6) contradict these rules, as addition of Triton X-100 leads to increased holdup.

Another available model for surfactant-containing solutions considers the $\left(\frac{d\sigma}{dx}\right)^2$, as originally proposed in the coalescence theory by Marrucci [104]. These are optimized for binary solutions where the solute has strong effects on surface tension [103,105], and can be used to predict holdup with reasonable accuracy (Fig. 3). Still, as bioprocesses don't contain binary mixtures, more comprehensive relations are required to predict coalescence in complex solutions.

3.3.3. Viscoelastic solutes

The presence of cells themselves can lead to increased viscosity and non-Newtonian behavior in bioprocesses. Especially filamentous fungi increase broth viscosity, which can result in a strong reduction of gas liquid mass transfer [106,107]. Besides cells, substrates (such as glycerol) and products (such as xanthan or similar polymers) can affect liquid viscosity of fermentation systems. .

The effects of viscous solutes on bubble columns are generally investigated with model fluids, such as carboxymethylcellulose (CMC), Xanthan gum, glycerol, saccharose, glucose, Boger solution and monoethylene glycol (MEG). Of these, glycerol, saccharose, glucose and Boger solution exhibit (almost) pure viscous behavior, while CMC, Xanthan gum and MEG exhibit viscoelastic behavior. The effects on bubble size and gas holdup can be split up in both a viscous and an elastic part, with viscous forces increasing bubble size and decreasing gas holdup, and elastic forces having the opposite effect [24]. However, in both viscous and viscoelastic solutions, gas holdup decreased compared to that of pure water, and bubble sizes increased [24,108]. .

Generally, a 4- to 5-fold decrease in k_La is observed in viscous systems, with a clear dependence on the concentration of the viscous solute [106,109,110]. With an increase in viscosity, k_L relations based on surface renewal due to turbulent eddies predict that k_L decreases due to their dependence on $\left(\frac{\epsilon}{\mu_L}\right)^{1/4}$. With an increase in viscosity (μ_L), k_L decreases. Following the relation by Park et al. [111], bubble rise velocity decreases with increasing viscosity, which on its own might imply a
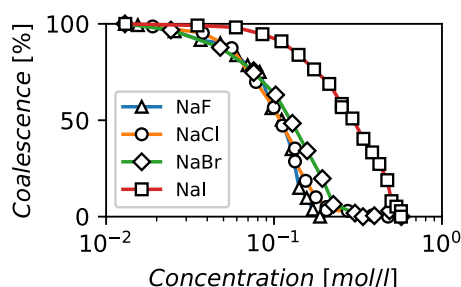
Fig. 3. Typical coalescence inhibition behavior of salt solutions. Here, a series of sodium halides was investigated: sodium fluoride ($\triangle$), chloride ($\circ$), bromide ($\diamond$), iodide ($\square$).
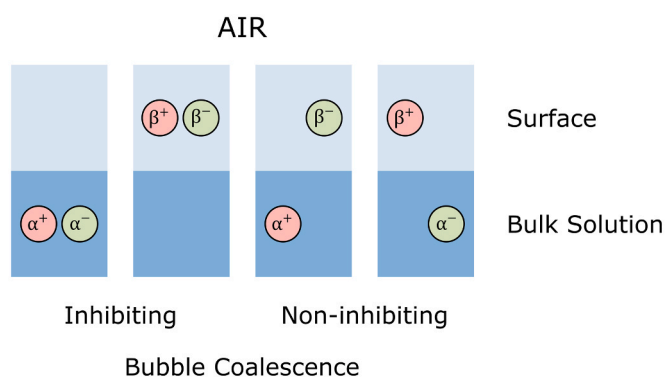Reproduced with permission from [87]. Copyright 2010 American Chemical Society.

Fig. 4. Schematic of ion partitioning and ion assignments from bubble coalescence. α ions are excluded from the surface; β ions are accumulated at the interface. $\alpha\alpha$ and $\beta\beta$ electrolytes inhibit coalescence; $\alpha\beta$ & $\beta\alpha$ electrolytes are less inhibiting. The surface layer width is $\sim$ 6Å based on calculations [94].
Figure reproduced with permission from [87]. Copyright 2010 American Chemical Society.
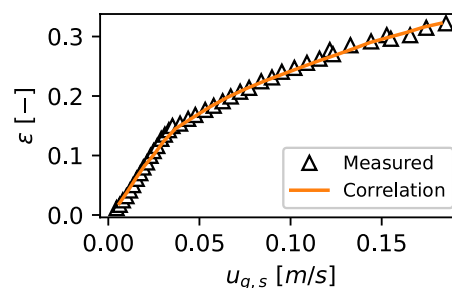
Fig. 5. Gas holdup in 1 wt% MEG solution as a function of superficial gas velocity.
Measured data (triangles) from [102], correlation values by [103]. Figure reproduced from [103], Copyright (2017), with permission from Elsevier.

higher gas holdup would be observed in viscous liquids. Experimental results support the increase in gas holdup for lower viscosities (up to 3 $*10^{-3}$ Pa*s) with glycerol, MEG and sucrose [102,112,113]. However, in the case of glycerol, this could also be linked to coalescence inhibition, as Keitel & Onken [114] reported that glycols inhibit coalescence as well, at concentrations about 10-fold higher than for alcohols. Glycerol was observed to start inhibiting coalescence at a concentration of 2 mmol/L, which is roughly three orders of magnitude below the amount needed to increase viscosity to $3*10^{-3}Pa*s$; the resulting smaller bubbles could explain the increased gas holdup.

More viscous systems ($0.01 - 3Pa*s$) show a clear decrease in gas holdup [112,113]. At these viscosities, the homogeneous flow regime is destabilized [113,115,116]. This leads to formation of larger bubbles with higher rise velocities – offsetting the aforementioned effect of a lower rise velocity. The reduction of holdup, combined with the increase in bubble size results in a drastically reduced interfacial area. Another effect of high viscosity is that the scalability criteria from Wilkinson [4] concerning column size no longer hold in systems of viscous oils ($\mu_L = 75mPa\cdot s$, $\sigma_L = 28$ $mN\cdot m^{-1}$), as in these systems holdup and bubble swarm velocity change with column diameter [117].

3.3.4. Alcohols

Alcohols are some of the main industrial products of bioprocesses, with bio-ethanol production a well-known example. In more recent years, the fermentation of syngas to various products has also seen commercial application [95]. The impact of ethanol on the mass transfer rate in such processes is substantial [53], and strongly contributes to the

feasibility of such processes [12]. In air-water systems, alcohols function as surfactants, migrating to the air-water interface and reducing surface tension. The presence of the alcohol molecules at the air-water interface then results in a reduction of k_L and an increase in the drag coefficient [118].

The presence of the alcohol molecules inhibits bubble coalescence [19,119], but only when the concentration of the alcohol exceeds a certain transition concentration [82,114]. These concentrations are generally below 10^{-2} mol/L [114]. In a clean water-ethanol system, coalescence behavior experiences a sharp 'step' at the transition concentration; addition of viscous solutes (xanthan or saccharose) smoothens the transition over a larger concentration range, while 50% coalescence rate is achieved at approximately the same concentration as in water-ethanol systems [82]. This could mean that, in more viscous systems, application of the binary design correlations for coalescent/non-coalescent systems is increasingly difficult. The reduction in coalescence explains the decrease in bubble size (d_{32} or $\overline{d_B}$) observed with the addition of alcohol [19,80,114,120]. Interestingly, the addition a small amount of ethanol (0.05% by volume) does not seem to affect the rate of bubble breakup, as pointed out by Gemello et al. [120]. It would be interesting to see if this holds for other alcohols and higher concentrations.

The combination of increased drag coefficient and smaller bubble sizes lead to an increase in gas holdup, as observed in both low superficial gas velocities (homogeneous regime) [1,80,114,121–123] and high superficial gas velocities (heterogeneous regime) [27,123–125], with the stabilized bubbles also delaying the transition to heterogeneous flow [34,126]. Both these effects are illustrated very well by Krishna et al. [125] (Fig. 6).

The effects on coalescence, bubble size and gas holdup are stronger for alcohols with a longer carbon chain [80,82,114,121,127], leading to a lower transition concentration, down to concentrations below 10^{-5} mol/L for alcohols with more than 6 carbon atoms [114]. This effect can be fitted with an exponential relation with the shape $c_t = a n_c^{-b}$ where a and b are fitted coefficients and n_c is the number of carbon atoms in the chain [82]. When fitted for solutions with different viscosities, values for a vary between 10 and 200, while values for b always fall between 7 and 8 [82]. When alcohols with $n_c > 8$ are considered, a very low transition concentration is predicted, in line with experimental results [114,119]. These results contradict the $\log_{10}P$ theory of McClure et al., as the $\log_{10}P$ of these alcohols rises above 3 and they are therefore expected to decrease holdup.

In the case of ethanol, there are hints that an optimum concentration exists above which the gas holdup decreases with further addition of
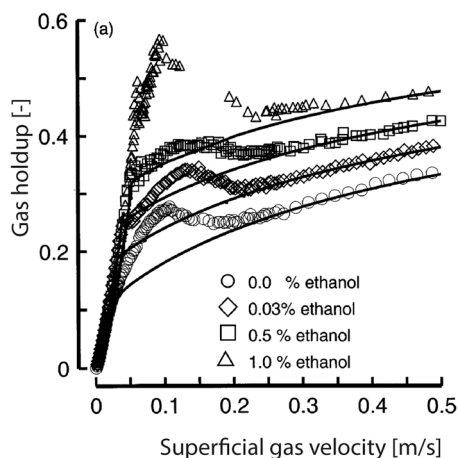

Fig. 6. Impact of ethanol addition on the gas holdup profile over a wide range of superficial gas velocity.
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ethanol [1,73]. It appears that simulations taking into account both the effect of surfactant and swarm effects on bubble drag, can reproduce this optimum concentration computationally [34].

3.3.5. Acids

Carboxylic acids are another relevant class of possible fermentation products, for which gas fermentation production routes are available [95].

The effect of C_1-C_3 monocarboxylic acids on d_{32} is highlighted by Keitel & Onken, who report a more pronounced decrease in bubble diameter than with n-alcohols [114]. This effect is confirmed with optical techniques [80] by Jamialahmadi & Müller-Steinhagen, who extend the analysis to gas holdup, comparing the holdup in 1% carboxylic acid solution with that of water. They present a small, but notable increase in gas holdup for formic acid solution, and strong increase for acetic and propionic acid, all at superficial gas velocities below 11 cm/s. Considering the $\log_{10}P$ values for these acids (-0.54, -0.17 and 0.33 for formic-, acetic- and propionic acid, respectively), these results fit within the $\log_{10}P$ theory by McClure et al. There is not much further work available on the impact of acids in aerated bioprocesses; further study may be worthwhile considering their potential commercial relevance.

3.3.6. Ketones

Another promising product from gas fermentation is acetone. Production of this ketone is currently in the scale-up or commercial phase [95]. However, research into the effect of ketones on bubble column hydrodynamics is limited. According to the available research, the effect of ketones is similar to that of alcohols, but less pronounced. Generally, an order magnitude higher concentrations of ketones are required to have a similar effect on bubble size and gas holdup [114]. In a comparison between acetone and ethanol, in a 1 vol.-% solution, ethanol results in a strong increase in gas holdup at superficial gas velocities above 3 cm/s, which is not the case for acetone [128]. These results go against the $\log_{10}P$-theory posed by McClure [25], as the $\log_{10}P$ values for ethanol and acetone (-0.31 & -0.24, respectively) are close to 0 and very similar to each other, which should lead to similar effects on gas holdup. Considering the importance of ethanol-based mass transfer enhancement in current syngas fermentation processes, a better understanding of the mechanisms at play and their impact on gas holdup and mass transfer is deemed highly relevant for the prospected diversification of the syngas-fermentation product portfolio.

3.3.7. Sugars

There's a myriad of bioprocesses using sugars (e.g. glucose, xylose, sucrose) as main carbon and energy source. Sugar concentration in continuous processes is very low in general, but high concentrations can be observed close to the feeding inlet. Batch processes start with a high sugar concentration which decreases over time. Not much data is available on the impact of sugars on hydrodynamics and process performance, considering the often-low concentrations, and low importance of rapid liquid mixing in batch processes. A study by Rivas-Interián et al. [79] reports that glucose has a statistically significant impact on $k_L a$, increasing it slightly. This effect correlates well with their reported increase in interfacial area and reduced d_{32}. Sucrose is noted to have a similar effect, but this effect is not statistically significant.

Reports on the effect of sugars on the surface tension of solutions vary, with most experimental work indicating an increase in surface tension with increasing sugar concentration, but some also reporting a decrease [129,130]. Several sugars are known to have a coalescence inhibiting effect, but there's no clear way to link this to molecular weight or surface tension gradients yet [129]. Deeper research on the relation between sugar concentration and bubble coalescence might not be worthwhile from a biotechnological perspective, as most fed-batch and continuous processes operate at very low bulk sugar concentrations (< 1 mM).

3.3.8. Biomass

A study on bubble coalescence in broth reports a clear reduction in coalescence rate for broth compared to mineral medium, but hardly a difference between filtered broth (cell-free) and unfiltered broth, indicating a minor role for cells themselves in the coalescence process [131]. Most of the effects of cells likely follow from surface active excreted metabolites and biopolymers, as well as lysis products.

The effect of cells themselves is noted to reduce oxygen mass transfer, with higher concentrations of cells leading to bigger reduction [2]. These experiments are carried out in a stirred tank with superficial gas velocities up to 5 mm/s, which is far from industrial conditions.

Some authors propose the presence of cells to enhance mass transfer, the so-called enhancement factor [10,74,132]. Due to the cells reacting with the gas in the transfer layer, the mass transfer rate can be improved. However, the effect of this factor is often found to be negligible for systems with internal aeration and mixing [133,134]. In some literature, the biological enhancement factor is taken to contain all (in)direct effects of the present biomass, including the presence of surfactant layers, which sometimes results in enhancement factors < 1. In this review, we aim to separate these factors into their own, separate effects to improve the modelability of novel bioprocesses.

Prakash et al. consider yeast suspensions at $u_{g,s}$ up to 0.3 m/s in a 0.28 m diameter column [135]. The gas velocity for small and large bubbles is estimated via the gas disengagement technique. Upon addition of biomass, an increase in velocity for large bubbles is observed, while the small bubble velocity is reduced.

The effect of cells is not studied extensively, but the literature available indicates that the cells themselves probably have only a small effect on coalescence rate. The enhancement factor can speed up mass transfer but might not be relevant in bubble column reactors. The effect on bubble drag and swarm effects is not yet reported, while the effects on bubble rise velocity are contradictory. The main opportunities for future studies lie in investigation of bubble drag and swarm effects, and investigation of the gas holdup in bubble columns with biomass.

3.3.9. Proteins

Proteins can be found in fermentation broths either as the main product of the process, or as a byproduct resulting from cell lysis. Research into foam stability for the food industry indicates significant effects of proteins [136,137]; it can thus be expected that bubble column performance can be impacted by the presence of proteins. Furthermore, the presence of proteins in solution tends to impact surface tension [138], which is of key importance for bubble formation, coalescence, and breakup mechanisms.

The scarce experimental results in literature confirm the decrease of bubble size (d_{32} or $\overline{d_B}$) with protein concentration [139,140]. However, the data is very limited and more detailed studies of the impact of protein concentration on bubble column hydrodynamics and mass transfer could prove important for predicting the performance of bioprocesses at high biomass concentrations, especially given the current interest in precision fermentation.

3.3.10. Combined solutes

Schügerl et al. [1] examined the impact of a *Candida boidinii* medium and biomass in combination with several alcohols on the gas holdup in bubble column reactors. It is often hard to distinguish the effects of the medium and yeast from the effects of the alcohol, as the alcohol effects seem to dominate. In the same series of experiments, a clear increase of volume-specific area is observed with the addition of 10 g/L salt to a methanol solution. Salt addition to an ethanol solution has a similar effect, though much less pronounced. No effect on gas holdup is seen with the addition of phosphate-containing medium to methanol or ethanol.

Addition of ethanol to syngas fermentation medium reduces k_L, but due to decreasing bubble size and increasing holdup, $k_L a$ still increases

[53]. In experiments with different broths from similar processes, these effects follow the same trend, but with different and unpredictable magnitude. It appears that the presence of cells and fermentation products confounds the effects of alcohol addition.

4. Future perspectives and conclusion

The effects of all solutes on the specific phenomena giving rise to the whole hydrodynamics will be succinctly listed below, starting at microscale, and focusing on ever larger phenomena, while we indicate the current knowledge gaps.

For the solute groups considered in this study, the effects on liquid side mass transfer (k_L) is only known for alcohols, salts, surfactants and biomass. For the other solutes, no studies concerning their effect on k_L could be found. For the four known ones, alcohols, salts, and surfactants reduce the k_L, while the presence of biomass increases the effective k_L through the enhancement factor. The effects of surfactants can be estimated through the impact on gas diffusion [98], so for single surfactants a simple study of gas diffusion in the pure surfactant can yield enough information to predict its impact on k_L. The effect of salts on k_L can also be directly correlated to the change in diffusion coefficient [66]. For combined surfactants and salts, elucidation of the additive nature of this effect would be an interesting and valuable avenue of research.

In the case of single bubble drag, there is literature data available only for surfactants, including alcohols. For surfactants, both increasing and decreasing drag coefficients are reported, depending on the nature of the surfactant. The distinction between drag-reducing and drag-increasing surfactants can perhaps be made with the $\log_{10} P$ theory proposed by McClure [25], but more data is needed to make this a robust predictor, and some deviations from this behavior have been observed.

Most of the studied solute groups have a clear impact on bubble coalescence, and most of these effects are inhibitory. Only viscous solutes and certain surfactants promote bubble coalescence. It is interesting to note that the effect of viscosity is somewhat counteracted by the elastic forces in viscoelastic solutions. For the coalescence inhibiting compounds, most of them exhibit a sharp transition concentration, below which their effects are negligible. The additive nature of compounds within and across separate groups of solutes remains an open area of research. If it turns out that transition concentrations are additive across groups, this will prove an essential tool for prediction of the coalescent nature of complex solutions. Determination of single-compound transition concentrations and their additive behavior is crucial for accurate prediction of bubble column behavior during fermentations.

Of the currently considered solute groups, surfactants can have a significant effect on the transversal lift force, the force that pushes larger bubbles away from the vessel wall and smaller bubbles toward the wall. The presence of surfactants can increase the critical size for this effect [75], meaning that fewer large bubbles will concentrate in the center of the column and the bubble rise velocity profile might be somewhat flatter. Further verification of this effect would require experiments in larger columns ($D_C > 0.15$ m). No clear impact on the lift force was described for the other solute groups.

All solutes that inhibit coalescence can delay the transition from the homogeneous to the heterogeneous flow regime. With that, almost all currently considered solute groups will impact the flow regime, generally delaying the onset of the heterogeneous regime. This effect is concentration-dependent, and higher concentrations of the same solute can further delay the onset of the heterogeneous regime [34]. However, there is also a maximum to this effect, as observed for ethanol solutions. The delayed onset of the heterogeneous regime is paired with increased holdup at higher superficial gas velocities. For solutes that promote bubble coalescence (viscous solutes and select surfactants), an inverse effect on the macroscale operation of the bubble column is observed, with earlier transition to the heterogeneous regime and decreased holdup. Future study on the impact of combined solutes on the transition

point would be very valuable, considering both combinations of multiple coalescence inhibiting compounds, and combinations of coalescence inhibiting and coalescence promoting solutes.

The overall mass transfer of bubble columns is reduced by viscous solutes and select surfactants, while it is increased by other surfactants, including alcohols, and salts. There is a small but uncertain effect of sugars, which implies an increase in mass transfer. However, this effect is likely irrelevant to most chemostat and fed-batch bioprocesses due to the low sugar concentrations.

In all above considerations, data for proteins is lacking. For this group of solutes, it is only known that they generally reduce bubble size. No effects of the type of protein have been reported, while it is expected that a rich series of effects is hidden underneath the surface. Comprehensive investigation of the impact of proteins on all different considered scales is an important avenue of research for improved prediction of bubble columns in bioprocesses.

Industrial and scientific practitioners are encouraged to assess the expected concentrations of the relevant solutes and, if these are above or around the transition concentration, to experimentally determine a correction factor f_{broth}, where $k_L a_{broth} = f_{broth} \cdot k_L a_{water}$, as is customary in wastewater treatment [141].

In conclusion, the performance of bubble columns for bioprocesses represents a complex interplay of forces and solutes, and while current modeling efforts are primarily focused on water-air systems, the broader landscape remains largely uncharted. We have summarized the influence of various biotechnologically relevant solutes on bubble column behavior, shedding light on the critical role they play in shaping the dynamics of these systems. Based on this, interesting paths for further research were identified to improve the modelling performance of bubbles in broth.

Key publications

There are a few key publications we recommend to the interested reader to further their understanding on the effects of solutes on bubble column performance:

[114] Keitel & Onken – A comprehensive experimental work considering the impact of multiple groups of organic solutes and salts on holdup, bubble size and coalescence.

[10] Garcia-Ochoa & Gomez – A clear and complete overview of design correlations for bubble columns and stirred tanks.

[93] Duignan – Salts & coalescence on the microscale, diving deep into the mechanisms behind coalescence inhibition by salts.

[24] Esmaeili, Guy & Chaouki – Comprehensive overview of effects of viscous and viscoelastic solutes on bubble columns.

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CRediT authorship contribution statement

RV: Conceptualization, Writing – original draft, Writing – review & editing, Visualization. **Lp:** Conceptualization, Writing – review & editing. **Ch:** Conceptualization, Writing – review & editing.

Declaration of Generative AI and AI-assisted technologies in the writing process

During the preparation of this work the authors used ChatGPT 3.5 in order to improve the readability of certain sections. After using this tool/ service, the authors reviewed and edited the content as needed and take full responsibility for the content of the publication.

Declaration of Competing Interest

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.

Data Availability

The data and code to reproduce the figures are included in the uploaded files.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.bej.2023.109124.

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