

## Evolution of bubble size distribution in particle stabilised bubble dispersions: Competition between particle adsorption and dissolution kinetics



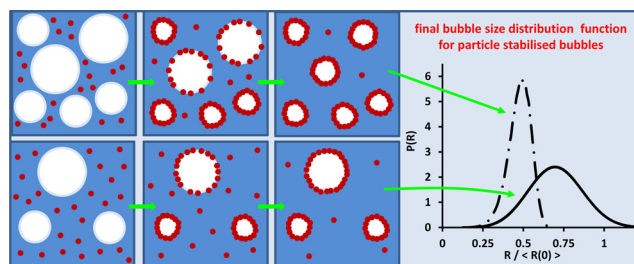
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### HIGHLIGHTS

- Evolution of particle stabilised bubble size distribution has been studied theoretically.
- Final size of bubbles can be “particle adsorption rate” or “particle number” limited.
- Final size distribution can be wider than original, in systems with an excess of particles.
- Final size distribution is narrower than original, for dispersions deficit in particles.
- Small bubbles influence the shrinkage of large bubbles, in particle poor dispersions.

### GRAPHICAL ABSTRACT



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### ABSTRACT

It is shown that in systems containing bubbles stabilised by nanoparticles, the time scales for the dissolution of small microbubbles can be comparable with those involving the transport and adsorption of the stabilising nanoparticles onto the surface of the bubbles. We have studied the evolution of model bubble size distribution functions in the light of this effect and also the competition between different sized bubbles for the finite number of available particles. It is found that for dispersions moderately rich in nanoparticles, the width of the final distribution function can become broader than the initial one, whereas for cases deficient in particles the reverse is observed. For each given bubble size, there exists a particle to bubble concentration ratio above which the final size of a bubble of this radius is no longer affected by the presence of other bubbles. In a system deficient in particles, this can still hold true for bubbles in the lower end of the size distribution range, but not the ones at the upper end. By considering simple cases consisting of just two bubble sizes, we show that the degree of shrinkage of the bigger bubbles is significantly increased in the presence of a small amount of gas in the form of smaller bubbles. In contrast, the final bubble size of the smaller bubbles is found to be largely insensitive to the amount of gas included within larger bubbles. The implications of these results for the final fraction of retained gas, in these types of particle stabilised bubble systems, are also discussed.

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## 1. Introduction

Many commonly encountered everyday foods owe their desirable texture and mouthfeel to the presence of bubbles and foams in

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these products. The size and spatial distributions, colloidal stability against coalescence, drainage and disproportionation, the life time and other similar physicochemical attributes of bubbles in food systems have been the subject of many research studies, both generally [1–5] and also in the context of more specific foods. Examples of the latter, underpinning the crucial role of gas bubbles in the quality of the final products, involve ice-creams [6,7], beers and sparkling wines [8,9], breads, cakes and dough [10,11] and beverages and coffee drinks [12]. Bubbles can also act as active or inactive filler particles in gel networks consisting of aggregates of emulsion droplets. By means of theoretical work, involving Brownian dynamic simulations, Wijmans and Dickinson [13] have shown that the presence of filler particles provides an interesting possibility for the control of mechanical strength and large deformation rheology of foods containing emulsion gels. Their findings have broadly been confirmed by experimental studies on such systems [14,15]. Indeed, one may extend the idea of using bubbles as particles even further by envisaging entire gel networks which consist of aggregates of bubbles, rather than emulsion droplets. Incorporating particulate gels of this type as food structuring agents has obvious distinct advantages. Not only can such an approach be very cost effective, it can also be useful in the design of healthier foods. Nonetheless, for the idea to be feasible in the first place, several conditions have to be met. Large bubbles are far too deformable to be used as substitutes for the emulsion droplets or replacements for colloidal solid particles. Thus, one needs to be able to generate rather small bubbles, say of the order of a few microns, in the system. Furthermore, these bubbles have to be highly stable against coalescence, given that in the gel network they remain very close to other bubbles throughout the shelf life of the product. Finally, they should also resist the process of disproportionation and subsequent dissolution. Interestingly, out of all these requirements, it is the latter that continues to pose the greatest challenge in realising small stable microbubbles in foods and related systems.

Disproportionation is the process of diffusion of gas molecules from small bubbles to larger ones, leading to the continual coarsening of the bubbles via dissolution and disappearance of the smaller bubbles. The process is driven by the chemical potential gradients between the gas molecules in different sized bubbles, caused by the difference in the Laplace pressure inside them. Thus, in this sense disproportionation is very similar to Ostwald ripening in emulsion systems. However, there are several factors that make the time scales for disproportionation much shorter than those encountered during Ostwald ripening. Firstly, there is the higher solubility of gas molecules compared to lipids. Secondly, the interfacial tension of gas–water interfaces is somewhat higher than those for oil–water surfaces. This leads to higher Laplace pressures and larger chemical potential gradients. Finally, the molar volume of gas molecules is substantially higher than that of oils. These differences, coupled with the fact that often the bubble dispersions are open to an air surface at the top of the product effectively acting as an infinite sized bubble, all serve to make disproportionation a considerably faster process, not only limiting the long term shelf life of the bubble system, but also its short term stability. For an isolated bubble of size  $R$  in an open system, the life time of the bubble has been estimated to be  $\tau R^3$  [16,17], where

$$\tau = \frac{P_0}{6\gamma S D_g R_g T} \quad (1)$$

Under atmospheric pressure of  $P_0 = 100$  kPa and at room temperature  $T = 298$  K, the value of  $\tau$  is found to be  $6.872 \times 10^{15} \text{ s m}^{-3}$  if one takes the gas–water surface tension as  $\gamma = 0.07 \text{ N m}^{-1}$ , gas diffusion coefficient  $D_g = 2 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$ , Henry's constant for gas solubility  $S = 7 \times 10^{-6} \text{ mol N}^{-1} \text{ m}^{-1}$ . The quantity  $R_g$  appearing in Eq. (1) is the universal gas constant,  $8.31 \text{ J K}^{-1}$ . Thus, for a bubble of size  $R = 0.1 \text{ mm}$  the life time is estimated to be a couple of hours,

while for a bubble of size  $2 \mu\text{m}$  ( $R = 1 \mu\text{m}$ ) the dissolution time can be as short as a few milliseconds.

A few techniques for stabilising small gas bubbles have been explored in the past. One such technique involves the use of highly insoluble gases such as decafluorobutane. The technique is widely used in medical imaging, with gas microbubbles of typical size  $3\text{--}4 \mu\text{m}$  acting as the ultrasound contrast agents [18]. For use in food systems, such a method is not available, both due to safety related issues and also the high associated cost. Another technique is to cover the surface of the bubbles with surfactants that tend to crystallize on the surface of the bubbles once they are adsorbed [19]. Microbubbles surviving up to a year have been reported using this method [20], although the limitation in the type of surfactant used and the inevitable competitive adsorption with other amphiphilic molecules always present in foods, has meant that the method has not been pursued much further. Yet another approach to stabilising very small bubbles is the so called Pickering route [21,22]. In this approach it is the adsorption of small nanoparticles, as oppose to molecular species, onto the surface of droplets or bubbles that is responsible for their colloidal stabilisation. Although known for quite some time, the first systematic studies of Pickering emulsions were carried out by Binks and Lumsdon in a series of studies [23–26], and later extended to bubbles by the same and other researchers [27–29]. As with any other type of emulsion or foam, strictly speaking, droplets and bubbles stabilised by particles remain thermodynamically unstable. However, due to the considerably higher energies involved in the displacement of suitably adsorbed particles from air–water or oil–water interfaces [21,30], for all intents and purposes the bubbles or emulsion droplets are indefinitely stable. For example, for nanoparticles of size  $20 \text{ nm}$ , possessing the appropriate surface chemistry to give a contact angle of around  $90^\circ$ , the adsorption energy can easily be thousands of  $k_B T$ . For this reason, interfacial networks of such particles provide an excellent means for controlling the dissolution of microbubbles.

Experimental work involving a variety of proteins, including strong and weak film forming proteins such as  $\beta$ -lactoglobulin and gelatin, has shown that such biomacromolecular based interfacial films do not possess sufficient strength to withstand the Laplace pressures inside the small bubbles [16]. Consequently, the macromolecules either desorb from the surface as the bubble shrinks or, where they form cross-linked films, the interfacial layers buckle and crinkle until only a shrivelled up shell of protein is left behind. In contrast, particle stabilised bubbles, once they are generated, retain their size for periods of days with no appreciable shrinkage [29,31] or sign of dissolution.

Whilst the adsorption of nanoparticles onto the surface of bubbles is certainly capable of arresting disproportionation, there are a few factors that have limited the use of this approach in the food industry. Prominent amongst these is the availability of edible grade nanoparticles of the right size, with suitable contact angles to adsorb at the interfaces. Fat crystals are often considered as one possibility, but seem to be better suited to the stabilisation of W/O rather than O/W or bubbles [32]. Indeed in food systems, such fat particles are often thought to contribute to the destabilisation and coalescence of bubbles, as is demonstrated by the poor foaming ability of whole milk when compared to skim milk [33]. Additionally, inclusion of saturated solid fat in foods is not particularly encouraged. A different possibility involves particles of ethyl cellulose [34] or indeed complexes of ethyl cellulose with cellulose [35]. Although promising results have been reported using this method, the smallest reported bubble sizes are of the order of a few tens of microns. Given that the size of the prepared ethyl cellulose particles tends to be  $\sim 100\text{--}200 \text{ nm}$  [34], it will be difficult to envisage stabilisation of bubbles much smaller than those already reported.

Yet another type of edible particle with potential for use in foods is based on the use of the fungal proteins called hydrophobins.

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