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Understanding Nanobubble–Nanoparticle Systems: Formation, Nucleation, and Submicron Interactions

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Abstract:

Over the last twenty years, gas-saturated liquids have drawn increasing interest due to the unexpected stability of nanobubbles observed within them. This stability challenges conventional understanding and has prompted investigations into a wide array of potential applications, particularly those involving interactions between nanobubbles and nanoparticles. This review explores the current knowledge surrounding systems where both entities coexist, which is common in many nanobubble-related technologies. We discuss key processes such as the generation of nanoparticles from nanobubbles, the initiation of nanobubbles on nanoparticle surfaces, and the interactions that occur between them. While substantial progress has been made, critical aspects such as the precise mechanisms underlying nanobubble nucleation and the nature of nanoscale interactions remain poorly understood. Continued research in this area is vital to deepen our theoretical insight and to harness these phenomena for practical use.

Keywords: Nanoparticles, Nanobubbles, Submicron Interactions, Interface.

Introduction:

Under typical conditions, when a gas is present in excess within a liquid, it is expected to leave the supersaturated solution either by forming buoyant bubbles or through diffusion of gas molecules. However, research has shown that in such supersaturated systems, stable, nanoscale gas domains known as *bulk nanobubbles* can form instead. These are spherical gas-filled colloids with diameters less than 1 μm [1]. Bulk nanobubbles consistently exhibit a negative zeta potential, generally ranging from −15 to −60 mV, regardless of their size. It is widely hypothesized that this surface charge plays a key role in their remarkable stability. The negative charges at the gas–liquid interface are thought to generate electrostatic repulsion that counteracts the internal Laplace pressure, thereby stabilizing the nanobubbles. Furthermore, this charge repulsion also prevents coalescence, which helps maintain their small size and prevents them from rising due to buoyant forces [2-4].

Bulk nanobubbles can be produced through two primary mechanisms: gas supersaturation [1] and energy input methods. One of the earliest investigations, conducted by Sette and Wanderlingh, demonstrated that high-energy neutrons whether naturally occurring in cosmic rays or introduced artificially can generate stable cavities within bulk water [5].

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Subsequent experiments showed that shielding water from neutron exposure inhibits cavity formation, a finding that has been corroborated by several independent studies [6,7].

Nanobubble formation via supersaturation represents another widely used approach. A well-known method in this category is solvent exchange, where a water-miscible solvent (such as ethanol) is combined with water. The process results in a supersaturated environment because atmospheric gases like nitrogen and oxygen are less soluble in the mixed solvent system than in either pure component. This oversaturation leads to gas release in the form of bubbles, potentially initiating the formation of bulk nanobubbles [1].

A range of techniques have been developed to achieve supersaturation and generate bulk nanobubbles, including water electrolysis, controlled heating, ultrasonication, pressure fluctuation, and mechanical stirring or agitation[8-10].

The interaction between nanoparticles and nanobubbles is of particular interest due to its relevance in numerous applications where both entities coexist. This relationship can be explored from four main perspectives. First, nanoparticles can form following the complete dissolution of nanobubbles. Second, nanobubbles themselves can nucleate from the surfaces of nanoparticles. Third, the direct interactions between nanoparticles and nanobubbles have been studied in various systems. Finally, understanding these interactions has important implications for technologies and processes that rely on the presence and behavior of nanobubbles. This review aims to clarify these four aspects by analyzing the current literature on bulk nanobubbles[11-13].

Fomation of nanoparticles from nanobubbles:

Numerous studies on bulk nanobubbles have reported that gas supersaturation in solution can lead to the appearance of nanoparticles, typically around 100 nm in diameter. These particles have demonstrated notable stability, persisting for several hours or even days. Although they are frequently presumed to be nanobubbles, further analysis has revealed inconsistencies in their properties such as density , compressibility, and refractive index [14] that challenge this assumption.

To explain the unexpected formation of these nanoparticles during gas supersaturation processes, a mechanism has been proposed [11,12]. According to this model (Figure 1), supersaturated gas levels promote the emergence of transient gas bubbles. During their brief existence, impurities present in the solution migrate to and adhere to the bubble surfaces. These impurities may originate from the techniques employed to generate nanobubbles, although even ultrapure water has been shown to contain sufficient contamination to influence bubble behavior. When such impurities accumulate at the interface, they can alter its physical characteristics, notably shifting it from a slip to a no-slip boundary condition [15].

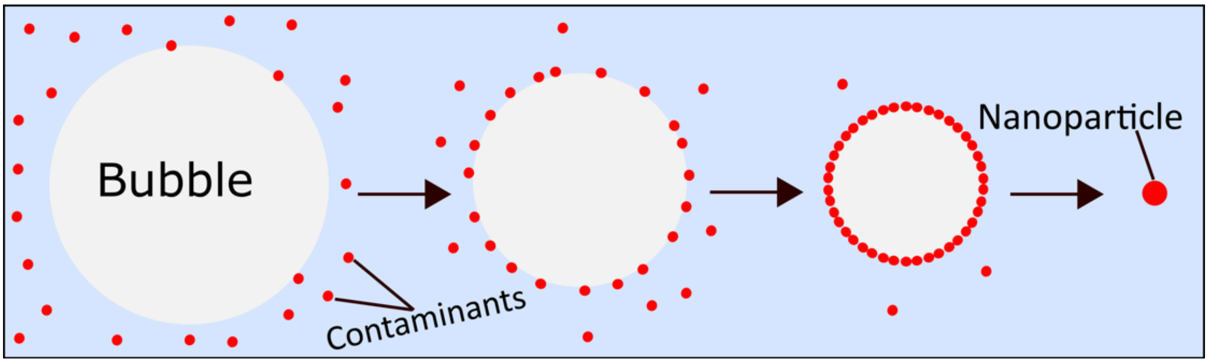

Figure 1. Schematic diagram of the mechanism of nanoparticles formation via supersaturation of gas and in the presence impurities. The formation of short-lived bubbles is initially enhanced by the supersaturation of dissolved gas. Later impurities that are already presented in the solution diffuse to the surface of these short-lived bubbles and get attached. As the bubble decrease in size, impurities will be concentrated, and this eventually leads to the generation of nanoparticles when the bubble dissolve. Adapted from Ref [12]

As these temporary bubbles fully dissolve, the material collected at their surfaces remains behind, forming nanoparticles. This phenomenon is plausible, given that even bubbles larger than 1 µm in diameter can accumulate enough surface contaminants to yield nanoparticles around 100 nm in size.

Nanobubbles nucleated from nanoparticles:

Several studies have suggested that combining nanomaterials with gas-saturated solutions can enhance the formation of nanobubbles. For instance, Xiao et al. observed that introducing gas into a solution already containing nanoparticles led to a notable increase in nanobubble concentration [13]. This enhancement was attributed to heterogeneous nucleation, facilitated by the presence of titanium dioxide nanoparticles, which requires less energy compared to homogeneous nucleation. Additionally, these nanoparticles contributed to improving the stability of the resulting nanobubbles. Key factors such as the surface wettability and texture (roughness) of the nanoparticles were identified as influential in promoting the formation of nanobubbles in bulk solutions.

Interactions between Nanobubbles and Negatively Charged Particles:

Zhang et al. discovered that the behavior of negatively charged gold nanoparticles in the presence of bulk nanobubbles varies depending on the size of the nanoparticles. Specifically, when the gold nanoparticles have diameters of 30 nm or less, the resulting mixture exhibits two distinct size distributions, as illustrated in Figure 2a-b. This suggests that the nanoparticles and nanobubbles remain separate entities with no significant interaction, maintaining individual stability. In contrast, gold nanoparticles with diameters of 40 nm or greater display different behavior compared to their smaller counterparts, indicating a potential interaction or altered stability in the mixture. For instance, when 60 nm gold nanoparticles were mixed with nanobubbles, as depicted in Figure 2c-d, a notable interaction was observed. The dynamic light scattering (DLS) analysis showed the disappearance of the characteristic peaks corresponding to both the original gold nanoparticles and the bulk nanobubbles. Instead, a new peak emerged, representing a size intermediate between the two original components. This suggests the formation of a hybrid complex that is larger than the nanoparticles but smaller than the nanobubbles. Remarkably, this newly formed nanobubble–nanoparticle complex remained stable for up to 8 hours, without significant changes in peak position or width. The authors hypothesized that this peak results from nanobubbles nucleating and growing directly on the surfaces of the gold nanoparticles. They noted that while the gold nanoparticles retain a fixed size, nanobubbles can expand or shrink through diffusion depending on their surrounding environment.

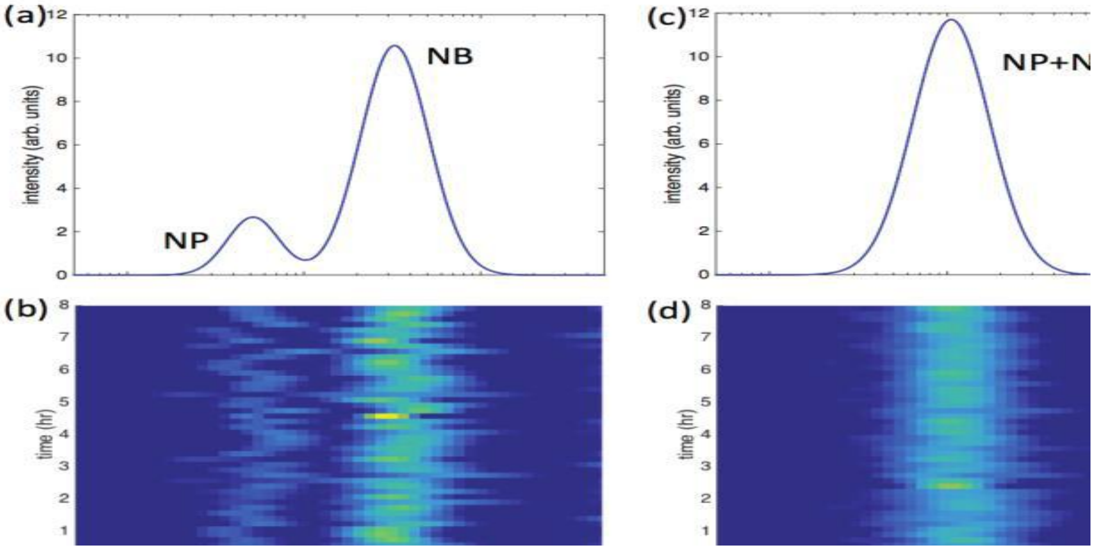

Figure 4. *Dynamic light scattering (DLS) analysis showing size distribution and temporal stability of negatively charged gold nanoparticles (20 nm and 60 nm) in a nanobubble-containing solution.*

Conclusion:

The study of bulk nanobubbles has revealed a complex yet fascinating landscape of behaviors and interactions that challenge classical expectations of gas behavior in liquids. Unlike macroscopic bubbles, bulk nanobubbles exhibit remarkable stability, largely attributed to their persistent negative surface charge, which provides electrostatic repulsion against coalescence and collapse. Two principal mechanisms gas supersaturation and energy input enable their formation, with a variety of methods such as solvent exchange, electrolysis, and ultrasonication contributing to their practical generation. An emerging area of interest lies in the interplay between nanobubbles and nanoparticles. Evidence shows that nanobubbles can lead to the formation of nanoparticles through impurity collection during transient bubble dissolution, while conversely, nanoparticles can serve as nucleation sites that facilitate nanobubble generation via heterogeneous nucleation. Furthermore, direct interactions between nanoparticles and nanobubbles depend significantly on particle size and surface properties, as demonstrated by gold nanoparticles, which form stable nanobubble–particle complexes when above a certain size threshold. These insights not only deepen our understanding of nanobubble behavior but also open new avenues for applications in areas such as drug delivery, water treatment, catalysis, and nanomaterials engineering. Continued investigation into the fundamental mechanisms governing nanobubble stability and nanoparticle interactions will be essential for optimizing their use across scientific and industrial domains

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