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STABILIZATION OF BULK NANOBUBBLES WITH A HYDRATE LAYER

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Abstract. The stabilization of bulk nanobubbles is considered with the balance of the Laplace pressure at their boundary due to surface tension and electrostatic pressure due to Coulomb forces. The presence of a hydrate layer of thickness ~1 nm with a tangential orientation of water dipoles around it is taken into account, the low permittivity of which, approximately equal to 3, increases the pressure at the nanobubble boundary. The sizes and charge of a stable nanobubble are determined. It is shown that in salt water, the hydration layer, regardless of the charge of the nanobubble, increases the pressure at its boundary by almost 30 times, and in fresh water – several times less.

Keywords: *dielectric liquid medium, Coulomb and Laplace pressure, hydration layer, anomalous permittivity, size and charge of nanobubbles*

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INTRODUCTION

Interest in bulk nanobubbles (BNBs) in water is associated with the prospects of their application in various fields [1–30]. A distinction is made between surface nanobubbles (SNBs) and bulk nanobubbles (BNBs). The main difference between them is that the former is stationary and the latter is mobile. Nanobubble technologies are widely demanded to address climate change, environmental protection, cost and energy reduction in industrial processes such as flotation, aeration and ozonation, which can remove pollutants and color, disinfect water and oxidize organic pollutants. Also, BNBs are used to solve problems in ecology, agriculture, in medicine/biomedical, in optimizing therapeutic and diagnostic methods and other fields due to their small size, large specific surface area, long residence time in water, high mass transfer capacity, and high zeta potential [3]. The use of BNBs in the treatment of commercial and domestic wastewater instead of the previously used biological approaches reduces energy costs, increases efficiency, and eliminates expensive chemicals and multi-stage treatment [6]. Therefore, the study of the properties of BNBs, including their stability, is relevant, which explains the wide discussion of this issue in the literature.

Atomic force microscopy experiments for the detection of SNPs can be considered as already proven, but the existence of BNBs, which are attempted to be proven by dynamic light scattering techniques [1], is still open to debate; however, there are strong indications for their existence [19]. BNBs are generated using solvent replacement, temperature change, hydrodynamic cavitation, and electrolysis [4].

For practical application, BNBs should be stable, i.e., have long lifetimes. The stability mechanism has been widely discussed in the literature [21–24]. In the hydrodynamic approach, taking into account the possible coalescence of neighboring BNBs, it was found that gases dissolved in the electrolyte inhibit the coalescence of BNBs, taking into account the reduction of the force of attraction between them. Similarly, but to a lesser extent, the surface tension gradient at bubble fusion (Gibbs–Marangoni effect) has an effect [21, 22]. The influence of ion hydration has also been noted [23]. The electrostatic approach is more widespread [1, 2, 5, 7, 8, 10, 11,1 3,1 6, 17, 20, 24–31], in which it is usually assumed that the stability of the BNB is ensured by the fact that the Coulomb pressure P_0 and the Laplace pressure P_L acting on the boundary of the BNB balance each other. The gas pressure inside the BNB is not taken into account, since it is by orders of magnitude smaller than the pressures P_C and P_L in the whole range of BNB sizes. It is clear that the stability of BNBs has a thermodynamic character and cannot be considered in isolation from their homogeneous generation, fission, and coalescence. In [28], the regions of charge and radius values where BNBs are stable, coalesce and fission were determined. In [29] the thermodynamic analysis of their homogeneous generation BNBs in supersaturated aqueous solution was carried out, the minimum work of formation and the radius of critical nuclei were calculated, and the conditions of BNBs stability were derived. It was shown in [30] that homogeneous generation of BNBs is due to the minimization of the Gibbs energy of the gas-liquid disperse system. Note that the model of a single charged bubble in a homogeneous liquid dielectric was usually used, which was refined in [31] by the hypothesis of the presence of a hydrate shell of BNB with a thickness $\Delta \sim 1$ nm and relative permittivity (RP) of water $\varepsilon = 3$ with a tangent orientation of dipoles. This was explained by the conservation of hydrogen bonding energy of water dipoles in the “tangential” shell structure compared to the energy of their polarization orientation in the previously assumed “radial” structure. The basis for this hypothesis was the known results of the molecular dynamics method [34] where it is said that molecules tend to keep the maximum number of hydrogen bonds, “packing forces” create a dense layer in contact with the surface. The directions of dipole moments $\mathbf{p}$ are aligned in the near-surface layer in the tangential direction. Moreover, the orientation of the dipole polarization vector is determined by hydrogen bonding rather than the electric field. In addition, scanning dielectric microscopy of the local water capacitance between atomically flat walls [32] revealed an interfacial $\boldsymbol{\Delta}$-layer ($\Delta \sim 1$ nm, $\varepsilon = 3$) near the charged electrode. It is known from [33] that near the interface, water is ordered in layers extending several molecular diameters from the interface. It was also shown in [35] that water exhibits a clear layered structure near all surfaces, regardless of their hydrophilicity within $\Delta \approx 1$ nm from the electrode. Since the law of energy conservation is universal and does not depend on the shape of the surface near which it is realized, its effect at extended charged surfaces [32–35] can be fairly applied to the spherical BNBs surface [31]. Probably, an indirect experimental confirmation of the presence of an electrically “dead” shell of the BNBs can be considered as their increased stability. The calculations in [36] of the van der Waals forces at the registered stable bubbles lead to the conclusion about the peculiarity of interfacial properties at the BNB boundary, which also serves as an indirect support of the conclusion [31] about the presence of the BNB hydrate shell. The refined model was used in [37] in analyzing the characteristics of stable BNB, where the conditions for the existence of its Stern layer were found. In [38], an increase in stability is reported due to the reduction of the BNB capacitance by the Δ-layer.

As noted above, BNB stability is usually considered to be ensured by the balance of Coulomb pressure at the BNB boundary (hereinafter referred to as BNB pressure):

$$P_{\mathrm{C}}(r_0) = \frac{{q_0}^2}{32\pi^2 \varepsilon_0 \varepsilon {r_0}^4} \tag{1}$$

and Laplace pressure:

$$P_{\mathrm{L}}(r_0) = 2\alpha / r_0, \tag{2}$$

where r_0 and q_0 are the radius and charge of BNB, respectively; $\varepsilon_0 = 8.85 \cdot 10^{-12}\,\frac{\mathrm{F}}{\mathrm{m}}$ is the dielectric constant of vacuum; $\alpha = 0.072$ N/m is the surface tension coefficient of water.

1. PROBLEM STATEMENT

Note that formula (1) is valid for a homogeneous dielectric medium. However, as noted above, a hydrate Δ-layer ($\Delta \approx 1$ nm, $\varepsilon_1 \approx 3$) of polar water molecules with a tangent orientation of the polarization vector is formed at the boundary of the BNB. Outside the Δ-layer, the RP of water returns to the usual value of $\varepsilon_2 \approx 80$ (Fig. 1). Therefore, let us refine the formula (1) taking into account the influence of the hydrate Δ-layer and the concentration of dissolved c salts.

2. ANALYZING THE STRUCTURE OF NANOBUBBLES IN WATER

A spherical system with a double-layer dielectric is similar to two capacitors of capacitance C_1 and C_2 [39]. For the BNB with radius r_0, the inner capacitor has the shell radii r_0 and $r_0+\Delta$, RP $\varepsilon_1 \approx 3$, and the outer capacitor parameters are $(r_0+\Delta)$, $(r_0+\Delta+L)$ and RP $\varepsilon_2 \approx 80$, where: $L = \sqrt{\varepsilon_0 \varepsilon_2 k_{\mathrm{B}} T / 2e^2 N_{\mathrm{A}} c}$ – Debye length, m; $k_{\mathrm{B}} = 1.38 \cdot 10^{-23}$ J/K is the Boltzmann constant; $N_{\mathrm{A}} = 6 \cdot 10^{23}$ mol^{-1} is Avogadro’s number; $T = 293$ K is the temperature, c is the concentration of the salt solution, mol/m^3; $e = 1.6 \cdot 10^{-19}$ C is the elementary charge.

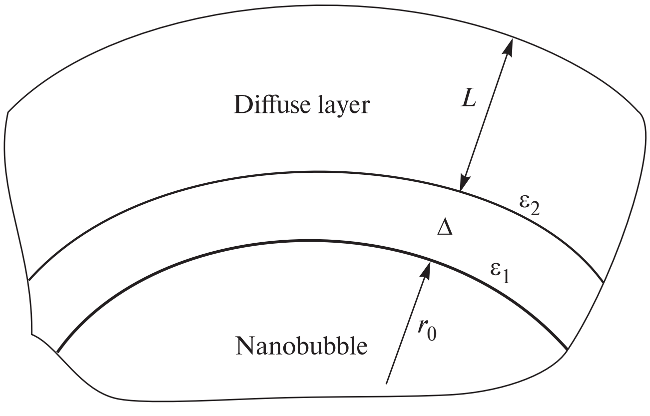

Fig. 1. Structure of the shell of the bulk nanobubble.

Capacitor capacities, $C_1 = 4\pi\varepsilon_0\varepsilon_1\left(\frac{1}{r_0}-\frac{1}{r_0+\Delta}\right)^{-1}$, $C_2 = 4\pi\varepsilon_0\varepsilon_2\left(\frac{1}{r_0+\Delta}-\frac{1}{r_0+\Delta+L}\right)^{-1}$, respectively. The total capacitance is determined by the well-known formula:

$$C(r_0) = 4\pi\varepsilon_0\left[\begin{array}{l}\frac{1}{\varepsilon_1}\left(\frac{1}{r_0}-\frac{1}{r_0+\Delta}\right)+\\ +\frac{1}{\varepsilon_2}\left(\frac{1}{r_0+\Delta}-\frac{1}{r_0+\Delta+L}\right)\end{array}\right]^{-1}. \quad (3)$$

Let's determine the electrostatic pressure of the BNB $P_C(r_0)$ in a liquid dielectric ($\varepsilon_2 = 80$) without Δ-layer and the pressure $P(r_0)$ for the BNB with Δ-layer. The energy of each of the BNB variants is $U = q_0^2/2C$. The pressure for the BNB at $\Delta = 0$ is [40]:

$$P_C(r_0) = -\frac{\partial U}{\partial r_0}\frac{1}{4\pi r_0^2} = -\frac{q_0^2}{8\pi r_0^2}\frac{\partial}{\partial r_0}\left(\frac{1}{C}\right) = \\ = \frac{q_0^2(L+2r_0)L}{32\pi^2\varepsilon_0\varepsilon_2 r_0^4(L+r_0)^2}, \quad (4)$$

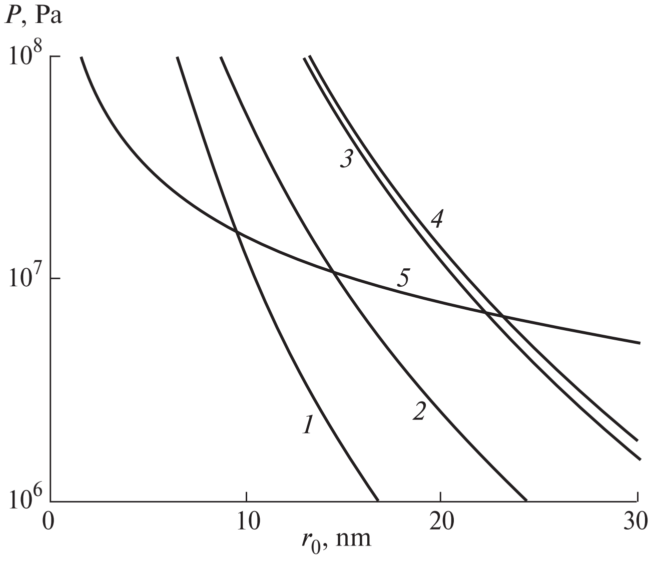

Fig. 2. Plots of dependence of pressures $P_i(r_0)$ at the nanobubble boundary ($q_0 = 4\cdot10^{-16}$ C) as a function of its radius: curves *1*, *2* – pressures $P_1(r_0)$ and $P_2(r_0)$ for BNB without Δ-layer in salt (c = 100 mol/m^3) and pure (c = 1 mol/m^3), water, respectively; curves *3*, *4* – pressures $P_3(r_0)$ and $P_4(r_0)$, for BNB with Δ-layer in salt (c = 100 mol/m^3) and pure (c = 1 mol/m^3) water, respectively; curve *5* – Laplace pressure $P_5(r_0) = P_L(r_0)$ according to formula (2).

where the capacitance $C = 4\pi\varepsilon_0\varepsilon_2 r_0(1 + r_0 / L)$. Similarly, taking into account (3), we find for BNB with Δ-layer:

$$P(r_0) = \frac{q_0^2}{32\pi^2\varepsilon_0 r_0^2}\times \\ \times\left[\frac{\Delta(2r_0+\Delta)}{\varepsilon_1(r_0+\Delta)^2 r_0^2}+\frac{L(2r_0+2\Delta+L)}{\varepsilon_2(r_0+\Delta)^2(r_0+\Delta+L)^2}\right], \quad (5)$$

which refines (1) taking into account the Debye length L and Δ-layer. As one would expect, formulas (4) and (5) at $\Delta = 0$, $L \to \infty$ come to the form (1). Note that the Debye length $L = 9.6$ nm in pure water and $L = 0.96$ nm in salt water.

Fig. 2 shows the dependences of pressure $P_i(r_0)$ on the BNB radius according to formulas (4) and (5) in pure (c = 1 mol/m^3) and salt (c = 100 mol/m^3) water, as well as the Laplace pressure $P_L(r_0)$, where the pressure index $P_i(r_0)$ corresponds to the number of the curve in the graph of Fig. 2. The increase in Coulomb pressure due to Δ-layer of BNB can be seen, which increases its stability and is particularly strong in salt water. Note that when $P_i(r_0)>P_L(r_0)$, it is the charge that makes the BNB stable, especially in its smaller size region. This points to erroneous conclusion [41] about the unattainability of BNB stability by fulfilling the condition $P(r_0) = P_L(r_0)$.

Let us compare the effectiveness of the influence of Δ-layer in salt and pure water by determining the pressure ratio $k_0(r_0)=P_4(r_0)/P_2(r_0)$ in pure and $k_c(r_0)=P_3(r_0)/P_1(r_0)$ in salt water for BNB with and without Δ-layer, which is shown in Fig. 3.

Let us analyze the obtained results. The increase in pressure $P(r_0)$ at the BNB with Δ-layer is due to the fact that the capacitance $C(r_0)$ of the series connected capacitors C_1 and C_2 is determined by the smallest of these values – the capacitance C_1 of the BNB Δ-layer, taking into account $\varepsilon_1 << \varepsilon_2$. This, according to (5), increases $P(r_0)$. It can also be seen in Fig. 2 that as the radius of the BNB increases, its pressure $P(r_0)$ becomes lower than the Laplace pressure $P_L(r_0)$, since $P_L(r_0)/P(r_0) \sim r_0^3$, subject to (1), (2). In addition, the size of the stable BNB due to Δ-layer increases with increasing its radius, but decreases with increasing salinity of c solution. This can be explained by the decrease in the Debye length L and the capacity of the BNB, considering (5). It can also be seen that Δ-layer weakens the effect of salinity on pressure and on BNB stability. This is due to the fact that the influence of the capacitance C_2 is suppressed by the first summand in square brackets (5), which is relatively large for the above reason $\varepsilon_1 << \varepsilon_2$.

Fig. 3 shows that in salt water Δ-layer increases the pressure of BNB almost 30 times, and in fresh water – from 10 to 4 times in the range of BNB sizes of 5–100 nm. This is also explained by the competition of the above-mentioned summands of formula (5). Moreover, in salt water, the summand dependent on L is so small that its change almost does not affect the value of $k_c(r_0)$, in contrast to the dependence of $k_0(r_0)$ in pure water. It is

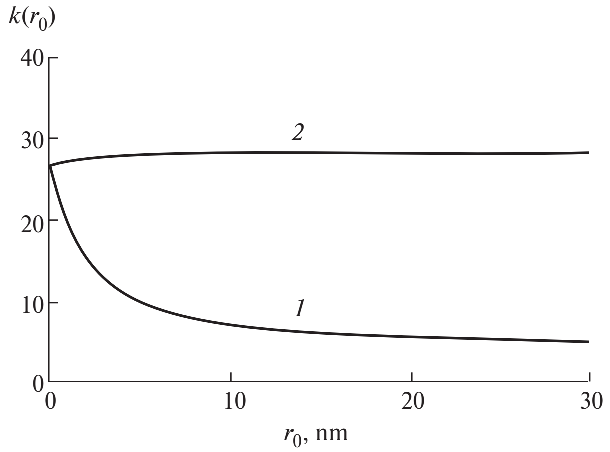

Fig. 3. Graph of dependence of the ratio $P(r_0)/P_C(r_0)$ on the BNB radius with and without Δ-layer: curve *1* – pressure ratio $k_0(r_0) = P_4(r_0)/P_2(r_0)$ in pure water ($c = 1$ mol/m^3); Curve *2* – pressure ratio $k_c(r_0) = P_3(r_0)/P_1(r_0)$ in salt water ($c = 100$ mol/m^3).

also important to note that the plots of Fig. 2 depend on the value of the BNB charge q_0, while the plots of Fig. 3 do not, since q_0 is a factor in expressions (4) and (5).

CONCLUSION

— The stability of the BNB is provided by Δ-layer with an RP of $\varepsilon_1 = 3$;

— the multiplicity of BNB pressure rise due to Δ-layer is independent of its charge;

— the BNB pressure due to salinity drops markedly for BNB without Δ-layer and slightly for BNB with Δ-layer.

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ETHICS DECLARATION

There are no human or animal studies in this paper.

CONFLICT OF INTERESTS

The author declares that he has no conflict of interests.

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