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Effect of nanoscale hydrophobic droplet inclusions on the cavitation threshold in water

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The effect of hydrocarbon liquid nanoclusions on the cavitation threshold in water is considered, taking into account the dependence of surface tension on the radius of curvature. Analytical expressions for the cavitation threshold inside a water and inside a nanodroplets are obtained. It is shown that the Tolman effect increases the cavitation strength of water and hydrophobic nanodroplets decrease it several times.

Keywords: cavitation, nanoscale droplet, Tolman length, surface tension.

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Introduction

Water under negative tensile pressure in a meta-stable state transits into an equilibrium state during cavitation — generation of vapor bubbles [1–3]. A capability of the liquid to withstand mechanical stress and sustain negative pressure is induced by forces of adhesion between its molecules. Water exemplifies such a liquid due to its exceptionally large cohesion induced by hydrogen bonds between molecules.

Cavitation phenomena are everywhere ubiquitous. Collapse of cavitation bubbles is highly important for jet printing, ultrasound cleaning of surfaces, non-invasive destruction of kidney stones by means of acoustic shock waves, sonochemistry, sonoluminescence as well as when studying geothermal processes. On the contrary, destruction collapse of hydrodynamically-generated cavitation bubbles adversely affects hydraulic systems, valves and vanes of propulsion screws, causing wear and erosion, while generation of the cavitation bubbles can affect characteristics of hydrofoils. In biology, where the temperature is determined by an environment, meta-stable water is mainly formed due to pressure reduction. Collapse of the cavitation bubbles creates catapult-like mechanisms in ferns and allows caridean shrimps to stun their prey. On the contrary, cavitation makes obstacles when a juice is uplifted along high trees, where significant negative pressure is observed. In the latter case, negative pressure is formed as a result of evaporation of water from foil cell walls, and it is about (-10 bar) in plants and can be up to (-80 bar) in some desert species..

According to a classic theory of nucleation [4], a macroscopic volume of absolutely pure water at the room temperature shall withstand pressure between -120 and -160 MPa for quite long time. In a real situation, cavitation is always heterogeneous. Container walls, microscopic impurities, surface defects or slots, in which vapor or gas bubbles were previously present, function as cavitation nuclei. These nuclei contribute to heterogeneous cavitation and cause significant reduction of threshold cavitation pressure from values predicted by the classic theory of nucleation for ideally pure water in large volumes.

Sever reality is that it is almost impossible to get absolute purity of liquids. Even applying modern water purification methods, it is not possible ([5–7]) to get rid of micro- and nano-scale insoluble hydrophobic impurities that reduce the cavitation threshold down to -30 MPa. Even in the cleanest high-quality water the content of total organic carbon exceeds $1\,\mu$g/L [8,9]. Therefore, hydrocarbon aggregates are inevitably present in any macroscopic volume of water. Now there is a question — whether these tiny aggregates called droplets can affect, even in small quantities, tensile strength of the entire water system. It is expected that liquid organic nanoinclusions in water induce homogeneous cavitation inside the very droplets and heterogeneous cavitation at a droplet-water boundary. Due to smallness of surface tension of oil as compared to water, the value of threshold pressure (in modulus), at which homogeneous cavitation starts inside oil, shall be significantly reduced.

The present study considers an effect of nanoscale hydrocarbon (oil) inclusions as liquid droplets (even in very small quantities) on ultimate tensile strength (the cavitation threshold) in the macroscopic volume of pure water. Our calculation is based on the theory and numerical analysis given in the study [3]. According to the theory, cavitation takes place both in water and inside droplets. This effect can be one of the main reasons of reduction of the cavitation threshold. Unfortunately, the study [1,3] has mistakes: the well-known Tolman formula [10] of a dependence of surface tension at a boundary of two phases on a curvature radius was incorrectly written. As a result, many main results of the article turned out to be incorrect. The present study is aimed at calculating a dependence of threshold pressure on average cavitation time and answering the

main question — whether insoluble oil nanoinclusions can reduced the cavitation threshold in water in several times.

Unlike the study [3], expressions for all the basic characteristics (a critical radius, a threshold barrier, cavitation pressure) are obtained in an analytic form in an approximation that preserves first-order terms relative to a small parameter — a ration of the Tolman length to the bubble radius.

1. Theoretical model

Cavitation in our model proceeds in three independent channels: 1 — homogeneous cavitation in the macroscopic volume of water V_w; 2 — homogeneous cavitation inside the nanoscale volume $V_o = \frac{4}{3}\pi R_b^3$ of the oil droplet (R_o — the initial radius of the droplet); 3 — heterogeneous cavitation inside the droplet on its wall (Fig. 1).

First of all, we consider homogeneous cavitation in ideally pure water. It is a volume of water, which is far away from vessel walls and does not contain microbubbles, droplet and solid hydrophobic inclusions and everything that can contribute to heterogeneous nucleation of the cavitation bubbles. In such water, the cavitation bubbles can be formed only by means of homogeneous nucleation.

Tensile negative pressure $p < 0$ causes origination of vapor-filled bubbles (homogeneous cavitation) of the radius r. A change of Gibbs free energy (a bubble generation work) is

$$G_w(r) = 4\pi\gamma_w\left(1 + \frac{2\delta_w}{r}\right)^{-1} r^2 + \frac{4}{3}\pi r^3 p. \qquad (1)$$

Here γ_w is surface tension at the flat water-vapor boundary, $|\delta_w| \ll r$ is a Tolman parameter (length) for the water-vapor boundary [10]. Correspondingly, a ration of the Tolman length to the bubble radius is a theory smallness parameter. It is shown within the Gibbs theory in the paper [10] that surface tension of the small droplet of water (or any liquid) depends on its radius r (more exactly, a tension surface radius) according to the law $\gamma_{wr} = \gamma_w\left(1 + \frac{2\delta_d}{r}\right)^{-1}$. Based on experimental data, Tolman believed that for the droplet

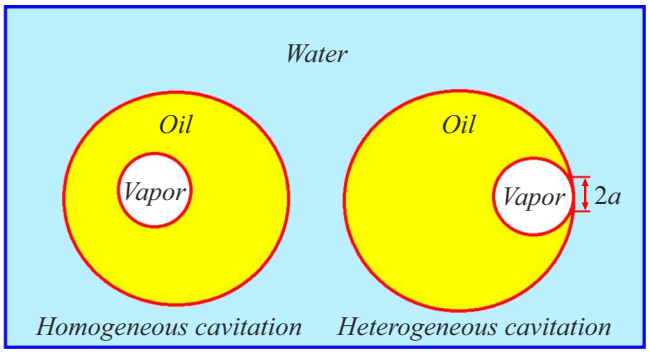

Figure 1. Image of homogenous and heterogeneous cavitation inside the nanoscale hydrophobic oil droplet.

δ_d is a positive number. When tracing calculations [10], one may be sure that for the nanobubble in liquid the condition $\delta_b < 0$ will be fulfilled. Indeed, it is written in the expression (3), (4) in the study [10]:

$$\frac{\Gamma}{\gamma' - \gamma''} = \delta_w\left(1 + \frac{\delta_d}{r} + \frac{1}{3}\cdot\frac{\delta_d^2}{r^2}\right).$$

Here γ' is a density of the liquid, γ'' is a density of its vapor. Γ — an excessive surface density of the mass (a single-component liquid–vapor system) on a mathematical tension surface of the droplet of the radius r. In a limit of the flat interface surface $\frac{\Gamma_d}{\gamma'-\gamma''} = \delta_d$, and for the droplet due to an inequality $\gamma'' < \gamma'$ we obtain that Γ_d and δ_d have the same signs and due to positivity of the Tolman parameter for the droplet the mass density for the flat liquid–vapor boundary is also positive $\Gamma_d > 0$. If the nanobubble is considered, then for its Tolman parameter δ_b at zero curvature the similar condition $\frac{\Gamma_b}{\gamma'-\gamma''} = \delta_b$ shall be fulfilled. For the flat boundary, $\Gamma_b = \Gamma_d$, therefore the Tolman parameter for the bubble shall be negative $\delta_b = -\delta_d$.

When $\delta_d \equiv \delta_w < 0$, surface tension of the cavitation bubble shall increase with reduction of the radius. The studies [1,3] an incorrect expression $\gamma_{wr} = \gamma_w\left(1 - \frac{2\delta_w}{r}\right)^{-1}$ was used for surface tension, while the sign was correctly selected $\delta_w < 0$. It turns out that with reduction of the radius surface tension of the bubble drops, which contradicts to the study [10].

The critical radius r_w^*, which corresponds to a maximum of free energy G_w^*, is found from the condition

$$\frac{dG_w(r)}{dr} = 0 \qquad (2)$$

and with accuracy to the smallness first-order terms by δ_w it is equal to the magnitude

$$r_w^* = -\frac{2\gamma_w}{p} - \delta_w. \qquad (3)$$

Substituting the found value of r^* into (1), we obtain the dependence of the energy barrier magnitude G_w^* on pressure p in the same approximation:

$$G_w^* = \frac{16\pi\gamma_w^3}{3p^2} + \frac{16\pi\gamma_w^2\delta_w}{p}. \qquad (4)$$

Cavitation is a thermally-activated stochastic process, in which the bubble gets a critical size. Similar to a chemical reaction, a number of cavitation acts per unit time in a unit volume κ complies with the Arrhenius law:

$$\kappa = \kappa_w \exp\left(\frac{-G_w^*}{k_B T}\right),$$

where k_B is the Boltzmann constant, T is an absolute temperature. The magnitude κ_w^{-1} shall be understood as time, after which cavitation occurs in the unit volume of the liquid without an energy barrier at a given value of

negative pressure. Thus, an average expected time τ_w, after which the cavitation act will occur in meta-stable water of the volume V_w, is

$$\tau_w = \frac{1}{\kappa_w V_w} \exp\left(\frac{G_w^*}{k_B T}\right), \quad (5)$$

κ_w is referred to a frequency of „cavitation attempts“ per a unit water volume without the energy barrier. The expressions of the type (5) are common in many studies on cavitation and the magnitude κ_w is estimated by means of computer simulation of molecular water models [1–7,11]. Solving (4) and (5) and leaving only the smallness first-order terms by δ_w, we finally obtain that cavitation pressure p_w depends on the observation time τ_w in the following way:

$$p_w = -\sqrt{\frac{16\pi\gamma_w^3}{3k_B T \ln(\kappa_w V_w \tau)}} + \frac{8\pi\gamma_w^2}{k_B T \ln(\kappa_w V_w \tau_w)}\,\delta_w. \quad (6)$$

Now we consider homogeneous cavitation inside the nanoscale hydrophobic hydrocarbon (oil) droplet of the radius R_o in the macroscopic volume of pure water V_w at negative pressure $p < 0$. The change of Gibbs free energy of the system $G_o(r)$ during homogeneous formation of the cavitation bubble of the radius r inside the droplet is

$$G_o = 4\pi\gamma_o\left(1 + \frac{2\delta_o}{r}\right)^{-1} r^2 + 4\pi\gamma_{ow}R^2 - 4\pi\gamma_{ow}R_0^2 + \frac{4}{3}\pi r^3 p. \quad (7)$$

Here γ_o is surface tension at the flat oil−vapor boundary, δ_o is the Tolman parameter (length) for the oil−vapor boundary, γ_{ow} is surface tension at the flat oil−oil boundary (the droplet size is also small, but it is much bigger than the nanobubble radius, so, we neglect the Tolman effect at the oil−water boundary); R is a finite radius of the droplet. Due to incompressibility of the droplet a volume conservation law is fulfilled:

$$R^3 = R_o^3 + r^3. \quad (8)$$

The finite radius of the droplet is excluded from (7), (8) and taking into account that $R_o \gg r$ we approximately obtain

$$G_o(r) = 4\pi\gamma_o\left(1 + \frac{2\delta_o}{r}\right)^{-1} r^2 + 4\pi\gamma_{ow}R_o^2 + \frac{8}{3}\pi\gamma_{ow}\frac{r^3}{R_o} - 4\pi\gamma_{ow}R_o^2 + \frac{4}{3}\pi r^3 p. \quad (9)$$

The equation $\frac{dG_o(r)}{dr} = 0$ is an forth-power algebraic equation with respect to r. After solving this equation and expanding the obtained expression into a series (details of quite long, but standard calculations are omitted), in a linear approximation relative to the Tolman length δ_o we obtain the simple expression for the critical radius r_o^*:

$$r_o^* = -\frac{2\gamma_o}{p + 2\gamma_{ow}/R_o} - \delta_o. \quad (10)$$

Then, by substituting the found value of r_o^* into (9), we obtain the dependence of the magnitude of free energy barrier G_o^* on pressure p in the same approximation:

$$G_o^* = \frac{16\pi\gamma_o^3}{3(p + 2\gamma_{ow}/R_o)^2} + \frac{16\pi\gamma_o^2\delta_o}{p + 2\gamma_{ow}/R_o}. \quad (11)$$

If the volume of water include N oil droplets, then the cavitation time decreases proportionally. The dependence of the cavitation time τ_o on the barrier magnitude G_o^* is written similar to (5):

$$\tau_o = \frac{1}{\kappa_o V_o N} \exp\left(\frac{G_o^*}{k_B T}\right), \quad (12)$$

κ_o — the frequency of „cavitation attempts“ per a unit volume of the oil droplet of the volume $V_o = \frac{4}{3}\pi R_o^3$ without the energy barrier. Solving the equation obtained from (11) and (12) relative to $p + \frac{2\gamma_{ow}}{R_o}$, we obtain the dependence of cavitation pressure p_o on the observation time τ_o in the linear approximation by the smallness parameter δ_o:

$$p_o = -\frac{2\gamma_{ow}}{R_o} - \sqrt{\frac{16\pi\gamma_o^3}{3k_B T \ln(\kappa_o V_o \tau_o N)}} + \frac{8\pi\gamma_o^2}{k_B T \ln(\kappa_o V_o \tau_o N)}\,\delta_o. \quad (13)$$

The third cavitation type, which can be observed in water with hydrophobic oil inclusions as nanodroplets is heterogeneous cavitation of the bubbles on the internal boundary of the droplet (Fig. 1). A circular portion of the radius a is an area of contact of the bubble with the internal surface of the oil droplet (Fig. 1). The magnitude a is directly proportional to the radius r of the formed bubble [3]:

$$a = \lambda r. \quad (14)$$

A coefficient of proportionality λ $(0 \le \lambda \le 1)$ is a unknown fitting parameter of the theory. The change of Gibbs free energy during heterogeneous cavitation $G_{ow}(r)$ is obtained by adding water−vapor surface energy of the (approximately) circular portion of the radius a to G_o and subtracting oil−vapor and water−vapor surface energies of the same portion:

$$G_{ow}(r) = G_o(r) + \pi\lambda^2 r^2\left[\gamma_w\left(1 + \frac{2\delta_w}{r}\right)^{-1} - \gamma_o\left(1 + \frac{2\delta_o}{r}\right)^{-1} - \gamma_{ow}\right]. \quad (15)$$

According to the Antonov rule, interphase tension at the boundary of two immiscible liquids is equal to a remainder of surface tensions of these liquids at the boundary with its own vapor [12]:

$$\gamma_w = \gamma_o + \gamma_{ow}. \quad (16)$$

Then, in the zero approximation, i.e. without taking into account the Tolman effect ($\delta_w = \delta_o = 0$), it turns out that works of formation of the cavitation bubble inside oil during heterogeneous and homogeneous cavitation are equal

$(G_{ow}(r) \approx G_o(r))$. In the expression for we neglected the effect of linear tension at the boundary of the three phases [12]. Substituting (9) into (15), after standard procedures and quite long calculations, for the critical nucleus in the linear approximation by the small parameters δ_w and δ_o we obtain the expression

$$r^*_{ow} = -\frac{2\gamma_o}{p + 2\gamma_{ow}/R_o} - \frac{\lambda^2\gamma_w}{4\gamma_o}\delta_w + \frac{\lambda^2 - 4}{4}\delta_o. \qquad (17)$$

When $\lambda = 0$, heterogeneous cavitation becomes homogeneous and (17) transits into (10). Substituting (17) into (15) and expanding the expression into a series, we obtain the dependence of the energy barrier G^*_{ow} on pressure:

$$G^*_{ow} = \frac{16\pi\gamma_o^3}{3(p + 2\gamma_{ow}/R_o)^2} - \frac{4\pi\gamma_o^2(\lambda^2 - 4)}{p + 2\gamma_{ow}/R_o}\delta_o + \frac{4\pi\gamma_o\gamma_w\lambda^2}{p + 2\gamma_{ow}/R_o}\delta_w. \qquad (18)$$

When $\lambda = 0$, the magnitude G^*_{ow} is transformed into G^*_o (see (11)). The formula for the observation time of heterogeneous cavitation is written as

$$\tau_{ow} = \frac{1}{\kappa_{ow}A_oN}\exp\left(\frac{G^*_{ow}}{k_BT}\right), \qquad (19)$$

where $A_o = 4\pi R_o^2$ is an area of the droplet surface, κ_{ow} is the frequency of „cavitation attempts “ per a unit are of the droplet without the barrier. As a result, after solving the algebraic equation and expanding into a series, we obtain the dependence of heterogeneous cavitation pressure p_{ow} on the observation time τ_{ow} from (18), (19):

$$p_{ow} = -\frac{2\gamma_{ow}}{R_o} - \sqrt{\frac{16\pi\gamma_o^3}{3k_BT\ln(\kappa_{ow}A_o\tau_{ow}N)}} - \frac{2\pi\gamma_o^2(\lambda^2 - 4)}{k_BT\ln(\kappa_{ow}A_o\tau_{ow}N)}\delta_o + \frac{2\pi\lambda^2\gamma_o\gamma_w}{k_BT\ln(\kappa_{ow}A_o\tau_{ow}N)}\delta_w. \qquad (20)$$

The expressions (6), (13) and (20) are the basic final equations of the present study. When assuming that cavitation proceeds only in these three above-said channels, then the resultant time of cavitation expectation will be calculated by the formula

$$\frac{1}{\tau} = \frac{1}{\tau_w} + \frac{1}{\tau_o} + \frac{1}{\tau_{ow}}, \qquad (21)$$

whence, by means of (5), (6), (11), (12), (18) and (19) one can find the functional dependence $\tau(p)$.

2. Calculation results and conclusions

Fig. 2 shows results of calculation of threshold pressure on time for pure water by the formula (6). Our calculations show that taking into account the Tolman effect increases cavitation threshold pressure. It is expected, since with taking into account that $\delta_w < 0$, in this case the critical radius and the energy barrier increase according to (3) and (4). Due to the error in the expression for surface tension, the study [3] has obtained an incorrect result — the Tolman effect reduces the modulus of cavitation threshold pressure.

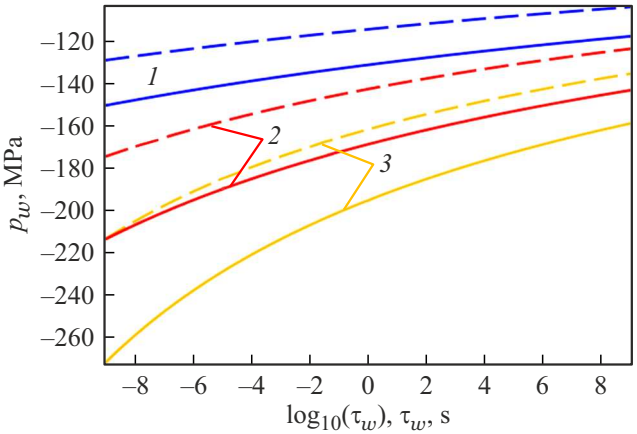

Figure 2. Dependence of cavitation threshold pressure on expected time in ideally pure water (that does not contain hydrophobic inclusions) at the three different volumes of water (calculated by the formula (6)). The dashed lines mark the calculation without the Tolman effect ($\delta_w = 0$), while the solid lines mark the same taking into account the Tolman effect. The data [3]: $T = 300\,\mathrm{K}$, $\gamma_w = 68\,\mathrm{mNm}^{-1}$, $\delta_w = -0.058\,\mathrm{nm}$, $\kappa_w = 1.25 \cdot 10^{18}\,\mathrm{s}^{-1}\cdot\mathrm{nm}^{-3}$. *1* — $V_w = 1L$; *2* — $V_w = 1\,\mu\mathrm{m}^3$; *3* — $V_w = 1000\,\mathrm{nm}^3$.

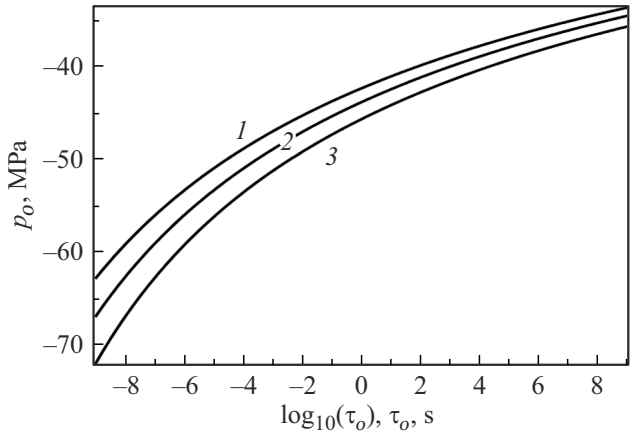

Figure 3. Dependence of threshold pressure of homogeneous cavitation inside the oil droplet on expectation time at the three different droplet sizes (calculated by the study (13)). The calculated data [3]: $T = 300\,\mathrm{K}$, $\gamma_o = 23\,\mathrm{mNm}^{-1}$, $\gamma_{ow} = 45\,\mathrm{mNm}^{-1}$, $\delta_o = -0.116\,\mathrm{nm}$, $\kappa_o = 5.4 \cdot 10^9\,\mathrm{s}^{-1}\cdot\mathrm{nm}^{-3}$, $N = 50$. *1* — $R_o = 200$; *2* — 100; *3* — 50 nm.

Results of calculation of homogeneous cavitation inside the droplets are shown in Fig. 3, 4. It is compared with calculation in Fig. 2 that the hydrophobic inclusions quite significantly reduce cavitation pressure. According to these

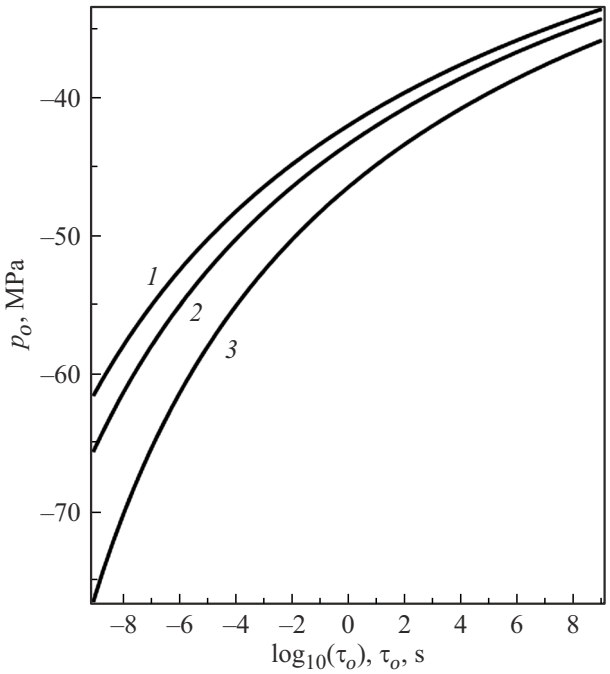

Figure 4. Dependence of threshold pressure of homogeneous cavitation inside the oil droplet on expectation time with the three different quantities of oils droplets in water (calculated by the study (13)). The calculated data [3]: $T = 300$ K; $\gamma_o = 23\,\mathrm{mNm}^{-1}$, $\gamma_{ow} = 45\,\mathrm{mNm}^{-1}$, $\delta_o = -0.116$ nm, $\kappa_o = 5.4 \cdot 10^9\,\mathrm{s}^{-1}{\cdot}\mathrm{nm}^{-3}$, $R_o = 100$ nm. *1* — $N = 100$; *2* — 100; *3* — 1.

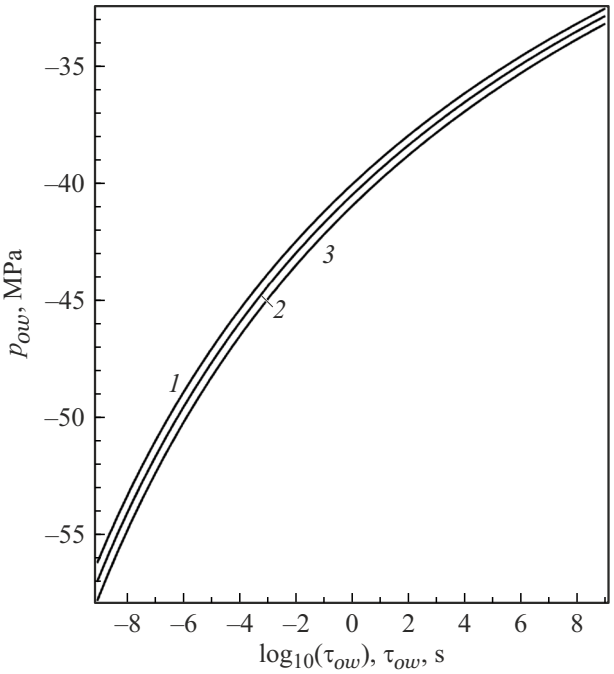

Figure 5. Dependence of threshold pressure of heterogeneous cavitation inside the oil droplet on expectation time at the three different values of the parameter λ, which determines the area of contact of the cavitation bubble with the wall (calculated by the formula (20)). The data [3]: $T = 300$ K; $\gamma_o = 23\,\mathrm{mNm}^{-1}$, $\gamma_w = 68\,\mathrm{mNm}^{-1}$, $\gamma_{ow} = 45\,\mathrm{mNm}^{-1}$, $\delta_w = -0.058$ nm, $\delta_o = -0.116$ nm, $R_o = 100$ nm, $\kappa_{ow} = 7.8 \cdot 10^{13}\,\mathrm{s}^{-1}{\cdot}\mathrm{nm}^{-2}$, $N = 100$. *1* — $\lambda = 0$; *2* — 0.7; *3* — 1. $R_o = 50$ nm.

calculations, with an increase of a number of the droplets and their volume threshold pressure drops.

As noted above, the energy barriers of homogeneous and heterogeneous cavitation inside the droplet without the Tolman effect (in the zero approximation) coincide (see the text after the formula (16)). In the linear approximation from (15) we obtain that $G_{ow}(r) - G_o(r) \approx 2\pi\lambda^2 r(\gamma_o\delta_o - \gamma_w\delta_w)$. If substituting numerical values of the Tolman lengths and surface tension (see a footnote to Fig. 5), we obtain that $G_{ow}(r) > G_o(r)$, i.e. heterogeneous cavitation on the droplet walls is not more energetically beneficial than homogeneous cavitation. This fact is illustrated by precise calculation in Fig. 5: with an increase of the parameter λ that determines the area of contact of the bubble with the wall cavitation threshold pressure p_{ow} increases.

We also note that the magnitude of the heterogeneous cavitation threshold p_{ow} does not transfer into the homogeneous cavitation magnitude p_o when $\lambda = 0$, which is due to different expressions for observation time in these two cases (see (12) and (19)). However, this inconsistence is insignificant in the theory, makes up a pair of percent and does not affect conclusions of the present study.

Fig. 6 shows calculation of the cavitation time by the formula (21) at the various values of tensile pressure. This calculation is useful by the following reason. If an

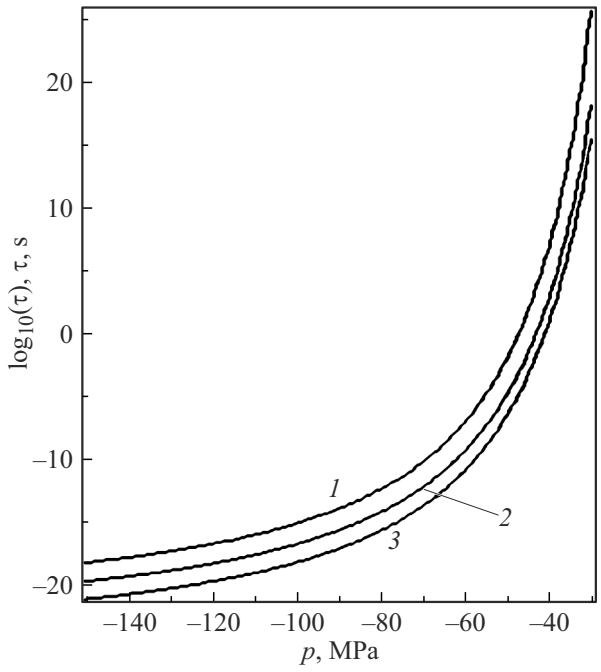

Figure 6. Dependence of cavitation expectation time on tensile pressure in liquid p, which is calculated by the formula (21) at the various values of the oil droplet radius R_o. The calculated data: $N = 1$, $\lambda = 0.7$, $V_w = 1$ L. The other parameters are the same. *1* — $R_o = 30$, *2* — 150, *3* — 750 nm.

acoustic wave occurs in water, then a pressure amplitude, at which cavitation originates, is called a Blake threshold [13]. Cavitation will occur, if a wave frequency satisfies the condition $f < \frac{1}{\tau}$. The higher the frequency, the lower the cavitation expectation time τ shall be, the higher the modulus of threshold pressure shall be and, correspondingly, the higher the Blake threshold shall be.

Conclusion

The present study was the first to obtain analytical expression for cavitation threshold pressure for the three cases: homogeneous cavitation within the volume of „pure water“ (that does not contain hydrophobic nanoscale liquid inclusions) and homogeneous and heterogeneous cavitation inside the oil nanodroplet. We have calculated values of cavitation pressure in these cases and confirmed a hypothesis that hydrocarbon hydrophobic nanoinclusions reduced cavitation pressure in several times.

The incorrect expressions [3] have been rectified, from which it followed that the Tolman effect reduced the cavitation threshold. It is also shown that homogeneous cavitation inside the hydrocarbon droplet occurs with smaller negative pressures than heterogeneous one, although the difference is very small (Fig. 5). For the case of acoustic cavitation, the method allows calculating the so-called Blake threshold.

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Conflict of interest

The author declares that he has no conflict of interest.

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