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STUDY INTO CORRELATION BETWEEN THE ULTRASONIC CAPILLARY EFFECT AND SONOLUMINESCENCE

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The correlation between the phenomena of luminescence generation in the cavitation region (sonoluminescence) and increase of the height and velocity of liquid rise in a capillary under the action of ultrasound (ultrasonic capillary effect) has been investigated. It is shown that with small gaps between the capillary and emitter the thresholds of these effects, i.e., minimum values of the amplitude of oscillations at which they originate, virtually coincide. Variation of the parameters which leads to an increase in the intensity of sonoluminescence results in increase of the ultrasonic capillary effect. The results obtained confirm the hypothesis of the cavitation nature of the ultrasonic capillary effect and indicate the possibility of using a capillary as an indicator of activity of acoustic cavitation.

Introduction. The ultrasonic capillary effect (UCE) is an increase of the liquid rise height and velocity in a capillary tube under the action of ultrasound [1, 2]. It can be demonstrated by the following simple experiment (Fig. 1a). Place the capillary tube into a vessel equipped with an ultrasonic emitter.

Fill the vessel with a liquid. As a result of capillary forces the liquid will begin to rise [3]. The liquid–gas surface boundary in the capillary (meniscus) stops at the normal capillary rise height H_0. If ultrasonic vibrations are now applied, the liquid will tend to rise to a new height $H^* = H_0 + H_{us}$, where H_{us}, is the height of the rise under the action of ultrasound. The H_{us} value in some cases exceeds H_0 by orders of magnitude and is much higher than could be caused by radiation forces and acoustic streaming alone. Radiation forces and acoustic streaming (or hydrodynamic pressure of acoustic streaming) affect both the liquid in the capillary and the liquid near the capillary. In our experimental conditions, at maximum amplitude the height of the "hillock" formed on the surface of the liquid due to these forces was less than 1 cm, which is orders of magnitude less than the H_{us}.

The ultrasonic capillary effect can be characterized by the liquid height rise H_{us} or by the excess pressure ΔP_0 that must be applied over the meniscus in the capillary in order to keep the liquid level at height H_0 (Fig. 1b). Normally $\Delta P_0 = \rho g H_{us}$ [1, 2] with precision not less than the precision of measurements.

It has been shown that cavitation plays an important role in generation of the liquid flow directed into the capillary [2, 4]. Its dominance can be illustrated empirically by exploiting the threshold nature of cavitation. For example, in the pre-cavitation conditions no rise of a liquid in the capillary under ultrasound was recorded. As the amplitude A of vibration of the emitter face is gradually increased, the meniscus level increases abruptly at the moment a cavitation cloud appears at the capillary channel inlet.

Once a cavitation cloud is present at the capillary inlet (Fig. 2), the magnitude of the UCE is independent of the relative orientation of the capillary tube with respect to the transducer axis. In this work, new results have been obtained, which confirm the cavitational nature of the ultrasonic capillary effect and give an idea about using a capillary as a sensor of cavitation.

Experimental. Figure 3 shows the schematic diagram of the experimental setup. The test chamber is a glass cylinder with a diameter of 80 mm and a height of 210 mm (internal dimensions). It was equipped with a coiled glass tube through which a temperature-controlled liquid was pumped. Glass capillaries with inner diameter d_{in} = 0.15 mm and wall thickness 1.8 mm were used in the experiments. The source of ultrasound was a piezoceramic transducer with

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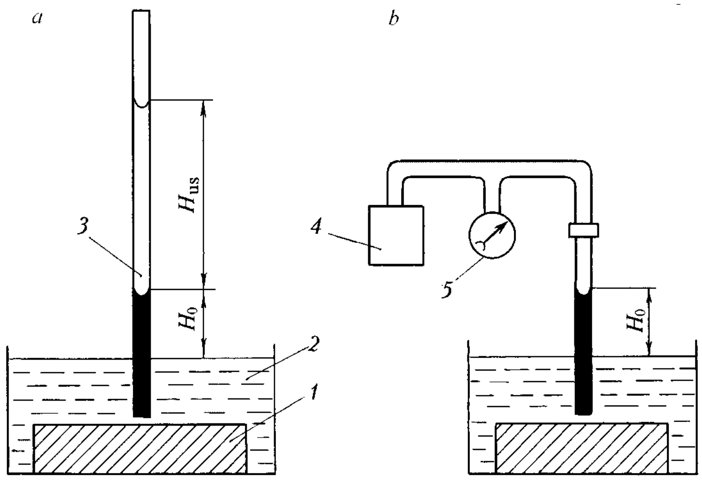

Fig. 1. Setup for measurements of H_{us} (a) and excess pressure ΔP_0 (b): 1) ultrasound transducer; 2) liquid; 3) capillary tube; 4) compressor; 5) manometer.

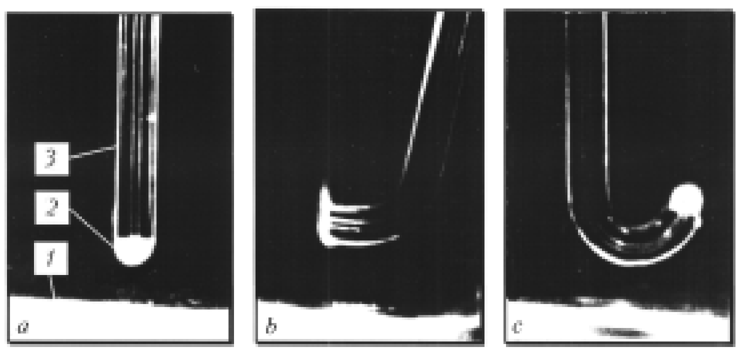

Fig. 2. Cavitation cloud at the butt-end of straight (a) and bent (b, c) capillary tubes in water (f = 41.9 kHz; A = 6 μm): 1) emitter; 2) cavitation cloud; 3) capillary. Distance d between the capillary butt-end and the emitter is 4.5 mm, inner diameter of the capillary tube is 0.15 mm, outer diameter is 3.9 mm.

a waveguide, mounted at the bottom of the chamber. The vibrating surface (i.e., ultrasound emitter) of the waveguide was 15 mm in diameter and its resonance frequency was 41.9 kHz. A UVM-3M noncontact vibrometer was used to measure the vibration amplitude of the emitter surface and to calibrate the amplitude sensor. All amplitudes cited in this paper are zero-to-peak, the standard error in the measurement of A being 4%.

The test chamber was filled with a liquid. The liquid was kept at the chosen temperature for 5 h and subjected to degassing by ultrasound for 20 min at the maximum transducer amplitude — 22 μm. In so doing, the gas content decreased by 20–25% compared with the equilibrium value [5, 6]. The gas content (mm^3/cm^3) was estimated by gas chromatography. Preliminary partial degassing of the liquid considerably increases the reproducibility of the results, since after this treatment the gas content remains essentially unchanged under the influence of ultrasound during measurements [5]. The test time between two successive measurements was chosen to be 5 min on the basis of the results of Ciuti et al. [6].

The procedure for the measurements was as follows. The capillary tube was immersed in the liquid and fixed in the prescribed position along the central axis of the chamber by means of a coordinate positioning mechanism. (The central axis of the chamber and of the waveguide of the transducer coincide to within ±0.1 mm.) The valve connecting the capillary–manometer–compressor system with the atmosphere was opened.

Under the capillary forces, the liquid in the capillary tube rose to the height H_0. The valve was then closed and the ultrasound generator was turned on. Under the action of ultrasound within a suitable regime of sonification, the liquid tended to rise again. The liquid was restored to its original position H_0 by using a compressor, which increased the pressure over the meniscus in the capillary. The excess pressure ΔP_0 over the meniscus necessary to keep

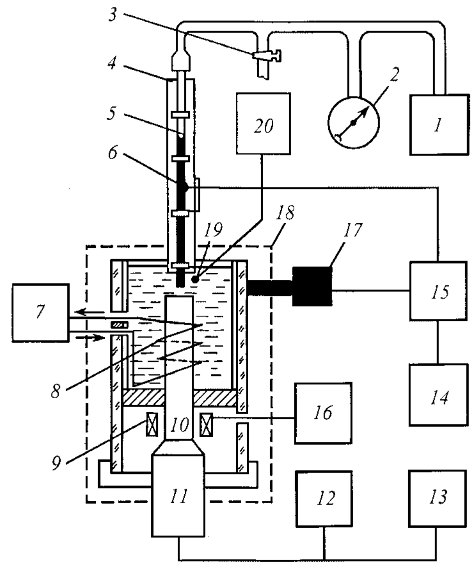

Fig. 3. Schematic of the experimental setup: 1) compressor; 2) manometer; 3) valve; 4) guide plate of coordinate positioning mechanism; 5) capillary; 6) piezoelectric sensor; 7) thermostat; 8) coil; 9) amplitude sensor; 10) wave guide; 11) transducer; 12) frequency meter; 13) generator; 14) computer; 15) oscilloscope; 16) voltmeter; 17) photomultiplier; 18) light-tight box; 19) thermocouple; 20) galvanometer.

TABLE 1. Vibration Amplitude Thresholds for the Appearance of SL and UCE for Different Liquids.

Parameter	d	Liquid					
		1	2	3	4	5	6
$A_{SL,th}$	0.05	7.0	2.5	1.5	0.7	0.5	0.4
$A_{UCE,th}$		8.5	3.0	1.5	0.9	0.5	0.5
$A_{SL,th}$	5.0	8.0	3.5	2.3	1.1	0.7	0.4
$A_{UCE,th}$		–	5.0	3.0	1.6	1.2	0.7

Note: 1 — glycerin; 2 — water–glycerin mixture with 60% (weight) of glycerin and 40% of water; 3 — water, 4 — chlorobenzene, 5 — isoamyl alcohol, 6 — acetone, $T = 25^{o}$C. Values of A are for single measurements taken to ±4% precision.

it at the level H_0 was measured by a manometer. Simultaneously the intensity L of sonoluminescence (SL) was recorded. The measurements of ΔP_0 and L in these experiments were accomplished after 2 min of sonification at the chosen amplitude.

The temperature of the liquid was monitored by a Chromel–Copel thermocouple, which was placed at a distance of 5 mm from the capillary entrance. The temperature was maintained constant within $\pm 1^{o}$C error limits in experiments with water and other low-viscosity liquids and $\pm 3^{o}$C in experiments with glycerin and water–glycerin mixture.

A computer-controlled digital signal generated a stepped ramp signal, which, by controlling the function generator, was used in experiments devoted to measurements of the threshold amplitudes of the phenomena studied. The emitter vibration amplitude could be held constant for a certain period and then increased to the next highest value by stepped ramp. In our experiments, the ramping sequence consisted of increasing the amplitude A to a set value to 5 sec, then to a higher value for 5 sec, etc., until sonoluminescence or the ultrasonic capillary effect manifested itself. The step size was 0.1 μm for amplitudes in the range 0–2 μm, and 0.5 μm for amplitudes in the range 2–15 μm.

Results. The main result of our experiments is that at ultrasound intensities lower than the SL threshold, no increase of the capillary meniscus level has been registered.

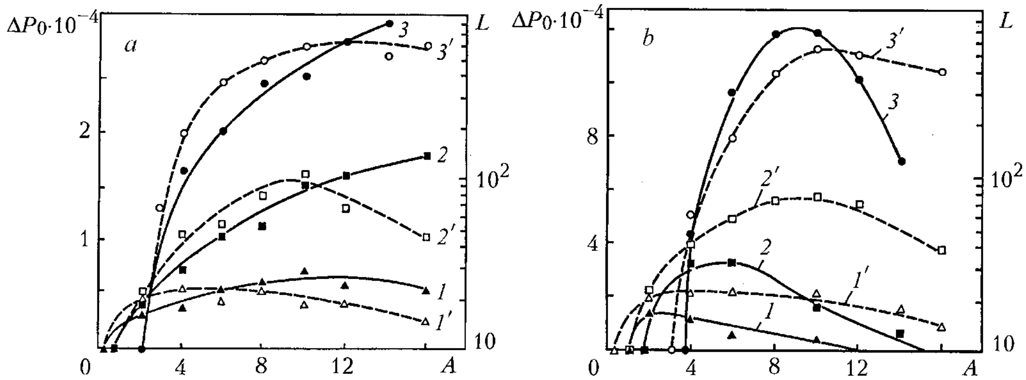

Fig. 4. SL intensity L (dashed lines) and pressure ΔP_0 (solid lines) as functions of the amplitude A of emitter oscillations for different liquids: 1, 1′) acetone; 2, 2′) water; 3, 3′) water–glycerin mixture with 60% (weight) of glycerin and 40% of water (T = 23°C); a) d = 0.05 mm, b) 5.

Table 1 gives single measurements of the threshold amplitude for the appearance of SL $A_{\mathrm{SL,th}}$ and for the beginning of the capillary rise under ultrasound, i.e., for the UCE appearance $A_{\mathrm{UCE,th}}$. Thresholds have been measured for two conditions: (1) when the butt-end of the capillary was at a very small distance d from the radiating surface (d = 0.05 mm) and (2) when the butt-end of the capillary was at a large distance (d = 5 mm). For each liquid and value of d, A was slowly increased, and the values at which UCE and SL appeared are noted below.

At small distances d (d = 0.05 mm), the SL and UCE thresholds are close, either occurring simultaneously (i.e., to within the resolution of the incremental increase of A) or with $A_{\mathrm{UCE,th}}$ tending to exceed $A_{\mathrm{SL,th}}$. If the capillary entrance is at the larger distance d (d = 5 mm), $A_{\mathrm{UCE,th}}$ exceeds $A_{\mathrm{SL,th}}$ generally to a greater degree than was seen for d = 0.05 mm. This is to be expected, since the acoustic field is unfocused. As the faceplate vibration amplitude increases, cavitation will first occur closest to the transducer faceplate and only later in the acoustic far field. The field of view of the photomultiplier covers both locations (d = 0.05 and 5.0 mm), while a capillary is a local sensor.

It is interesting that with the exception of acetone, both thresholds are lower with a capillary positioned close to the surface of the emitter, the ratio of the $A_{\mathrm{SL,th}}$ values being 0.6–0.9 and the ratio of the $A_{\mathrm{UCE,th}}$ values being 0.4–0.6. This suggests that the proximity of the capillary tube plays a part in nucleating inertial cavitation. (Note also that in Fig. 2 a cavitation cloud nucleated at the butt-end of the capillary.)

Hence, as is often the case with multibubble cavitation, it appears that the sonoluminescent threshold being measured here is one relating to the nucleation of inertial cavitation. This can be compared to two other thresholds. First, there is the threshold for detection of the effect (SL), which is sometimes crucial in experiment [7] and, as illustrated above, must also take into account the volume of liquid to which the detector is sensitive. Second, there is the threshold which is most commonly considered in theoretical attempts to predict the "threshold for cavitation," which pertains to the achievement within the collapsing bubble of some conditions (of, say, internal temperature [8–10] or wall velocity [11]) which are taken to characterize inertial cavitation. In such theoretical treatments it is usual to assume the pre-existence of a suitable nucleus to nucleate cavitation. Clearly, the actual observation of cavitation requires all three thresholds (nucleation, collapse, and detection) to be exceeded, such that the most stringent threshold becomes the important one. The evidence here is that the nucleation threshold is critical.

The measurements presented in Fig. 4a were done with the capillary positioned at a small distance from the radiating surface (d = 0.05 mm) and in Fig. 4b with the capillary positioned at a large distance d (d = 5 mm).

A linear scale has been chosen for ΔP_0 and a logarithmic one for L. Every point is the averaged result of three independent measurements, and the uncertainty associated with each point is explained later.

From Fig. 4 it is seen that, once the driving amplitude has exceeded the thresholds of SL and UCE by a sufficient amount, in general those of these liquids which exhibit higher SL intensity also show a greater ultrasonic capillary effect. For both small (Fig. 4a) and large (Fig. 4b) values of distance d, SL intensity first grows with A, achieves a maximum value, and then tends to decrease. This is in qualitative agreement with results related to studying the dependence of cavitation activity on ultrasound intensity [13]. While ΔP_0 is also exhibited when the larger value

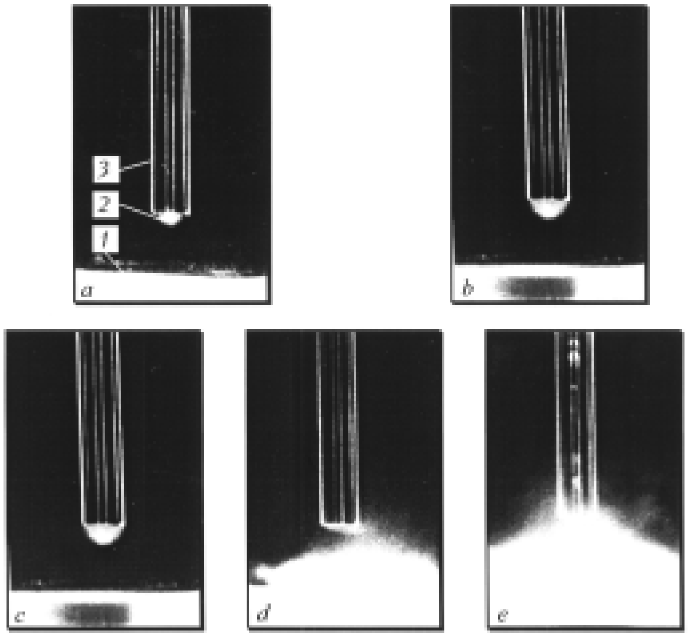

Fig. 5. Cavitation zone in water at the capillary inlet, for a distance d = 5 mm between transducer face and capillary butt-end, shown for different transducer amplitudes A: a) 1; b) 2.5; c) 6; d) 10 μm; 1) emitter; 2) cavitation cloud; 3) capillary.

of d is used (Fig. 5), no maximum is seen within the range of A used for water and glycerin–water mixture for the d = 0.05 mm case (Fig. 4b). While in these two cases ΔP_0 increases with A, it is entirely possible that extending the study to higher A might well reveal a maximum. This is supported by the fact that in Fig. 4a the liquid whose maximum occurs for the lowest value of A is acetone, the only liquid which does indeed show a maximum (a broad one at a high value of A, around 12 μm). The peaks in L occur at higher values of A than do the peaks in ΔP_0 for the same liquid at d = 5 mm (Fig. 5) and at lower values of A than for ΔP_0, if peaks are to occur at all, for d = 0.05 mm (Fig. 4).

For large d (Fig. 4b) the SL dependences are nearly the same as in the previous case (Fig. 4a). Hence while, as mentioned earlier, repositioning the capillary can affect the nucleation which occurs, this seems to be a second-order effect in measuring SL since the photomultiplier has a large field of view and takes sonoluminescence from the entire volume over the radiator. The ΔP_0 dependences differ significantly from those in Fig. 4. Here ΔP_0 at first increases, achieves a maximum, and then decreases, all the maxima occurring at $A < 9$ μm.

Figure 5 shows the visible cavitation clouds at the larger stand-off distance used (d = 5 mm). Here, the ultrasonically induced meniscus rise in the capillary begins when a cavitation cloud appears at the capillary butt-end (Fig. 5a). By increasing the vibration amplitude A the size of this cloud is increased as well as its optical density (Fig. 5a, b). The optical density is increased evidently because of increasing of the bubble concentration in the cloud. Under these conditions, the UCE also increases.

Similarly, the density of bubbles in the volume between the emitter and the capillary increases as well, and at high amplitudes (Fig. 5c–e) the density of bubbles in the volume of the liquid is nearly the same as at the capillary entrance. In this condition there is a decrease in the visible size of the cloud at the capillary (which is clearly separated from the bubble cloud in the cavitation zone adjacent to the transducer). The capillary cloud starts to dance chaotically on the surface of the butt-end of the capillary, and the average ΔP_0 decreases (see Fig. 4b, curve 2, at approximately 6 μm or curve 3 at 9 μm). It should be noted that under these conditions the level of the liquid in the capillary starts changing chaotically (sometimes in a jump-like manner) with amplitudes so great that the standard deviation of measurements of the UCE in these conditions is 25%. At A not much higher than the SL threshold, when the cavitation stays stably at the capillary (Fig. 5a, b), such large liquid level fluctuations do not occur, and the standard deviation is 10% (d = 5 mm).

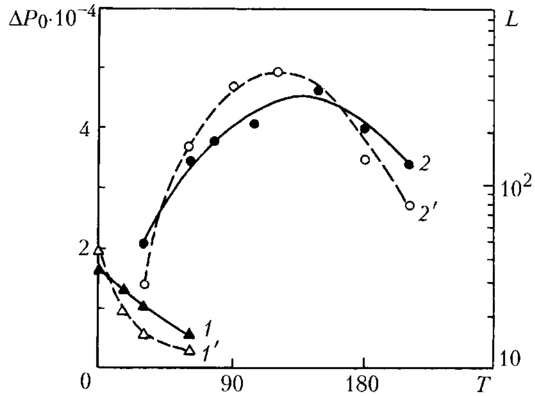

Fig. 6. SL intensity L (dashed lines) and pressure ΔP_0 (solid lines) versus temperature for different liquids: 1, 1′) water, d = 5 mm; 2, 2′) glycerin, d = 0.05 mm.

At small d (Fig. 4a), such fluctuations are not seen for any value of A used, and the standard deviation of measurements does not exceed 10%.

Figure 6 shows the temperature dependences of L and of ΔP_0 at d = 0.05 mm. For water, both L and ΔP_0 decrease with temperature. For glycerin they increase, achieve a maximum, and decrease by increasing temperature. With respect to their temperature dependences in both liquids, the variations in the sonoluminescent and the ultrasonic capillary effects correlate well.

From Figs. 4b and 6, it is seen that at d = 0.05 mm the luminescence peak occurs at lower amplitudes of A than does the peak in ΔP_0 (if it is seen to occur at all). At higher amplitudes the behavior of SL and UCE may differ significantly. If one is to look for a regime where UCE is an indicator of sonoluminescent activity, it needs to be restricted to below this peak. Indeed, the temperature-dependence study of Fig. 6 was conducted in this regime and showed a correlation between the measures of SL and UCE. At the larger value of d (5 mm, Fig. 4), the peak of ΔP_0 occurs at lower driving amplitudes than does the peak in L, and the UCE amplitude threshold is significantly higher than the SL threshold (Table 1).

Discussion of the Results. There is evidence of a correlation between SL emission and the ultrasonic capillary effect (Table 1, Figs. 4 and 6), the quality of which varies with certain parameters (e.g., stand-off distance d, driving amplitude A, and whether one is measuring threshold or activity). First, thresholds in driving amplitude A for both phenomena are closer for the smaller d (0.05 mm) than for the larger one (5 mm), for the reasons discussed earlier. Second, for much of the range of driving amplitude and temperature tested, an increase in SL is reflected by an increase in UCE, the only exception being for those conditions between maxima in the two measures (assuming that peaks in ΔP_0 would occur could sufficiently high values of A be achieved in Fig. 4b, lines 2 and 3).

This degree of correlation between the phenomena may be considered as confirmation of the association between cavitation and the UCE and evidence to support the hypothesis of a cavitational mechanism for UCE [2, 14]. In accordance with this hypothesis, the mechanism of UCE is as follows. Under the action of ultrasound a cavitation zone (or cavitation cluster) appears at the capillary entrance. Cavitation bubbles collapse asymmetrically with the formation of microjets of the liquid. On entering the capillary channel, every such jet adds to the height of the capillary rise by a magnitude ΔH_r. When summed up these increments result in the experimentally observed height and speed of the rise (or penetration) of the liquid in the capillary channels. The higher the concentration of bubbles at the capillary inlet and the more violently they collapse, the stronger the ultrasonic capillary effect one may expect. The same is true for sonoluminescence in the general sense, although while sonoluminescence may be associated with jetting [15–17] this is not an exclusive relationship.

Given that in this experiment the sensor for sonoluminescence covered the entire liquid volume between the capillary and the transducer faceplate, one would expect better agreement in, for example, measures of the thresholds for UCE and SL when the local sensor (the capillary) is placed close to the faceplate (where cavitation occurs first) than when it is placed further away (in a volume of liquid which requires higher values of A to cavitate). This is seen in Table 1 and Fig. 4b. Clearly, a direct comparison of the two measures would require restriction of the field of view of the photomultiplier to the local region around the capillary tip.

The fact that UCE and SL arise through different mechanisms is another reason for the differences cited above. Consider, for example, the detection threshold of the two sensors. For a photomultiplier this consists of detecting a statistically significant increase in the photon count above the background level, which depends, in turn, on thermal and electronic factors, the ambient light, etc. With the UCE the sensitivity depends on other factors. Within the interval between two sequential penetrations of the jet into the capillary channel, if inertial effects are allowed then liquid may flow out of the capillary tube under the gravitational forces or excess pressure ΔP_0 (depending on the method of measurement). The height of the rise, in this case, is reduced by a value of ΔH_d before the next collapse at the capillary entrance. When the statistically averaged ΔH_d becomes equal to the statistically averaged ΔH_r, the rise stops.

If the concentration of collapsing bubbles is low (ultrasound intensity is not much higher than the SL threshold), the probability of collapse of the bubble at the capillary inlet is small. Hence the statistically averaged time interval between sequential collapses is long and the portion of the liquid which entered into the capillary (as a result of the bubble collapse) has time to flow out from the capillary before the next collapse. As a result, the averaged increase of the capillary rise under ultrasound is zero although cavitation is there and SL intensity is not zero.

The photographic evidence suggests that the capillary is not a noninvasive sensor, in that a cavitation cloud forms around its tip. The appearance of the bubble cluster at the capillary butt-end by increasing the amplitude *A* (Fig. 5) is explained as follows. The butt-end of the capillary tube is produced mechanically, specifically by cutting and polishing. Microcracks appearing on the glass surface after this process are the sources of cavitational nuclei. The lateral surface of the capillary tube is formed during capillary production by meltback (sweating) of the glass. It is therefore much smoother and has significantly reduced potential to nucleate cavitation on the surface than that of the capillary butt-end. At moderate ultrasound intensities (Fig. 2) this will predispose the cavitation to appear first at the capillary inlet. However there is another reason for early cavitation nucleation here: the surface of the capillary may be considered as a rigid acoustic boundary, which will tend to increase the acoustic pressures at the interface.

Increasing the temperature from 10^{o}C to 80^{o}C (Fig. 6) causes the surface tension σ, density ρ, and viscosity η water to vary respectively by 10, 1.5, and 50%. These changes have only a second-order influence on the bubble collapse. Vapor pressure will increase by more than 20-fold over the same temperature range, and this will tend to make individual bubble collapses less violent and less efficient in transforming sound energy into the energy of shock waves, vapor-gas mixture heating, and the energy of microjets. As a result, both UCE and SL emissions decrease with increasing temperature in water, despite the fact that increase in vapor pressure can increase the number of nucleation events.

In the case of glycerin, two competing factors strongly influence the bubbles dynamic: the decrease in viscosity η and the increase in vapor pressure P_v. The rate of change of P_v with temperature in the range 20–120^{o}C is much smaller than that seen in water (approximately 75%). In this temperature range the prevailing factor is viscosity. It varies by more than 2 orders of magnitude (from 1480 to 5.2 mPa·sec). Decreases in viscosity promote more rapid and more efficient collapses. As a result, both SL and UCE increase in this range of temperatures.

In the 120–200^{o}C range, the variation of viscosity in glycerin is not so great (from 5.2 to 0.22 mPa·sec) and the vapor pressure increases by a factor of more than 50 (from 0.1 to 5.8 kPa). The increase in vapor pressure tends to decrease the velocity of bubble collapse, as discussed above, and to decrease the efficiency of energy transformation by collapsing bubbles. As a result, both UCE and SL intensity decrease in this range of temperatures in glycerin.

Conclusions. At ultrasound intensities lower than the SL threshold no increase of the capillary rise has been registered.

At small distances *d* between the capillary butt-end and the radiating surface ($d < 0.2$ mm), the ultrasonic capillary effect is a reliable instrument for detecting the threshold for multibubble inertial cavitation. As the driving amplitude is increased above the threshold, the increase in UCE appears to be a reliable indicator of the increase in SL, but only up to the amplitude at which the SL peaks.

At large values of *d* the rise of the liquid in a capillary under ultrasound is visually associated with the presence of cavitation at the capillary channel inlet. the UCE becomes a local measure of the cavitation at the butt-end, which is itself influenced by the presence of the capillary. As a result, the UCE would be a better monitor of cavitation in strong fields, as opposed to a field close to the threshold.

The results of this study support the hypothesis regarding the cavitational nature of the ultrasonic capillary effect.

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NOTATION

A, amplitude of emitter oscillations, μm; $A_{SL,th}$, minimum (threshold) amplitude of emitter oscillations at which sonoluminescence occurs, μm; μm; $A_{USE,th}$, minimum (threshold) amplitude of emitter oscillations at which the ultrasonic capillary effect occurs, μm; d, distance (gap) between the capillary and emitter, mm; d_{in}, inner diameter of the capillary, mm; f, frequency of ultrasound, kHz; g, free-fall acceleration, m/sec^2; H_0, height of liquid rise in the capillary without ultrasound, m; H_{us}, increment of the height of liquid rise in the capillary under the action of ultrasound, m; H^*, height of liquid rise in the capillary in the ultrasonic field, m; ΔH_r, increment of the height of liquid rise in the capillary when a liquid microjet enters the capillary channel, m; ΔH_d, decrease of the height of liquid rise in the capillary in its discharge from the capillary within the time interval between two subsequent collapses of the bubble at the entrance to the capillary channel, m; L, output signal of the photomultiplier, V; ΔP_0, excess pressure necessary to retain the meniscus at the height of an ordinary capillary rise, Pa; T, temperature, ^{o}C; σ, surface tension, mN/m; η, liquid viscosity, mPa/sec; ρ, liquid density, kg/m^3. Subscripts: us, ultrasonic; th, threshold; SL, sonoluminescence; UCE, ultrasonic capillary effect; m, maximum; d, discharge; r, rise.

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