LITERATURE CITED

1. P. D. Dan and D. A. Ray, thermal Tubes [Russian translation], Moscow (1979).
2. S. Chi, Thermal Tubes: Theory and Practice [Russian translation], Moscow (1981).
3. A. E. Missan, Trudy GIPKh, No. 44, 112-120 (1960).
4. I. F. Nefedova, Trudy GIAP, No. 1, 235-245 (1953).
5. A. D. Hunt, Analyst, 102, 846-851 (1977).
6. Short Chemical Encyclopedia [in Russian], Vol. 2, Moscow (1963), p. 726.
7. G. S. Beskova, M. G. Mamakina, and A. I. Butusova, Zavod. Lab., No. 3, 23-26 (1983).

SONOLUMINESCENCE AND SUBHARMONIC GENERATION IN A CAVITATION ZONE OF AQUEOUS SODIUM CHLORIDE SOLUTIONS

N. V. Dezhkunov, G. Iernetti,
P. P. Prokorenko, A. Francescutto,
and P. Ciuti

UDC 534.29:535.37

The variation of the sonoluminescence and the subharmonic component of the cavitation noise spectrum is investigated in a solution of sodium chloride in water.

The investigation of the acoustical and optical characteristics (sound absorption, generation of a characteristic subharmonic frequency of cavitation noise, and the generation of optical luminescence) of a cavitation zone in aqueous solutions of sodium chloride is of considerable interest from two points of view.

First, cavitation exerts a more intense destructive action on metals in aqueous salt solutions, sea water in particular, than in tap or distilled water. This fact has been attributed until now to chemical corrosion, which intensifies cavitation percussion effects [1-3]. Such an explanation has been deemed well justified, especially insofar as chemical reactions, in turn, are strongly intensified in a cavitation zone. However, with an increase in the salt concentration the physicochemical properties of the water change, and this can not only affect the chemical activity of the solutions, but also exert a direct influence on the properties of the cavitation zone has been disregarded to data, and the results of cavitation tests on pure water (e.g., in water tunnels) have been used without any kind of corrections for engineering calculations of systems operating in solutions, specifically in sea water [4].

Second, the physicochemical properties of aqueous sodium chloride solutions have been quite thoroughly investigated. This means that the results of studies of such solutions can be used to assess the influence of the properties of the liquid on the cavitation zone. Moreover, NaCl solutions from the basis of a number of industrial compositions used in ultrasonic industrial processes.

In light of the foregoing considerations, we now report an investigation of the influence of the solution of salt (NaCl) in water on the characteristics of a cavitation zone, viz.: the intensity I_Σ of the total acoustic signal sensed by a hydrophone placed in the cavitation zone, the intensity I_{sub} of the subharmonic component of the total signal, and the sonoluminescence intensity L. We note that the simultaneous investigation of subharmonic generation and sonoluminescence is also of major interest from the standpoint of ascertaining the mechanism of sonoluminescence.

EXPERIMENTAL APPARATUS

A block diagram of the apparatus is shown in Fig. 1. As in [5], we used a pulsed cavitation-generation technique. The pulse-modulated acoustic signal (with a carrier frequency of 0.7 MHz) was excited by means of a focusing piezoelectric ceramic radiator. The use of a

---

Institute of Applied Physics, Academy of Sciences of the Belorussian SSR, Minsk. Università di Trieste, Italy. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 51, No. 3, pp. 417-424, September, 1986. Original article submitted May 27, 1985.

0022-0841/86/5103-1052$12.50

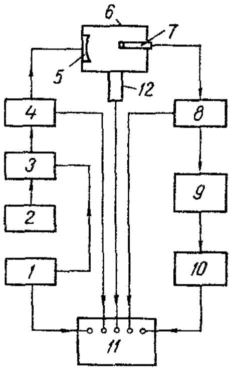

Fig. 1. Diagram of the experimental arrangement. 1) Pulse generator; 2) rf signal generator (0.7 MHz); 3) mixer; 4) amplifier; 5) focusing radiator; 6) working volume (acoustic cell); 7) hydrophone; 8) preamplifier; 9) filter (350 kHz); 10) selective amplifier; 11) oscilloscope; 12) photomultiplier.

focusing radiator made it possible to produce cavitation in the liquid volume and to eliminate the influence of the cell walls and cavitation nuclei situated at those walls on the inception and growth of the cavitation zone.

The working volume comprised a water-cooled metal (stainless steel) cylinder with a useful volume of 160 cm^3. This cell was equipped with a conical cover having a ribbed surface to prevent the onset of standing waves. The experimental piezoceramic transducer ($f_0 = 0.7$ MHz; diameter 26 mm) had one flat surface and one concave surface with a radius of curvature equal to 50 mm. The radiator was attached under an opening in the bottom of the cell by a screw inserted through a rubber gasket. A thermocouple and a miniature hydrophone of sensitivity 1.8 V/bar were inserted into the cell. The total (integrated) signal from the hydrophone, which was placed behind the focal spot of the radiator, was sent to the preamplifier. Then the filter separated out the subharmonic $f_0/2$, which was sent through the final amplification stage to the multichannel oscilloscope.

The oscilloscope also recorded the total acoustic signal V_Σ, the photomultiplier output signal, and the excitation signal (voltage U).

The working volume was filled with two-day-settled liquid through a tube connecting it to the main reservoir in such a way as to minimize the variation of the gas content of the liquid due to the trapping of gas bubbles during its transfer from one vessel to the other. The liquid was changed after each measurement in order to diminish the variations of the gas content due to ultrasonic degassing.

RESULTS OF THE MEASUREMENTS

The voltage U applied to the transducer, the salt concentration, and the ratio of the pulse repetition period T to the pulse duration (off-duty factor) $N = T/\tau = (\tau\nu)^{-1}$ were varied in the experiment. Typical results are shown in Figs. 2-5. It is evident from the oscillograms and graphs that for sufficiently high ultrasonic intensities ($U \geq 120$ V) the sonoluminescence intensity increases smoothly with the salt concentration over the entire range of variation of C, attaining an eightfold increase in the saturated solution ($C = 270$ g/dm^3) in comparison with distilled water (Fig. 3). At relatively low ultrasonic intensities (<100 V) the L(C) curves exhibit an extremal behavior, the luminescence intensity dropping rapidly to zero after attained a maximum. With a decrease in U the maximum shifts to the left toward smaller values of C. The subharmonic intensity decreases with increasing concentration for any value of U.

With an increase in the ultrasonic intensity (which is proportional to the square of the voltage applied to the transducer, U^2) the intensity I_Σ of the total acoustic signal varies directly as U^2, i.e., is proporitonal to the ultrasonic intensity. In this range of U^2 the intensity I_Σ remains practically the same for solutions of different concentrations. Luminescence and subharmonic radiation are absent (Fig. 4). Then the spectrum of the total

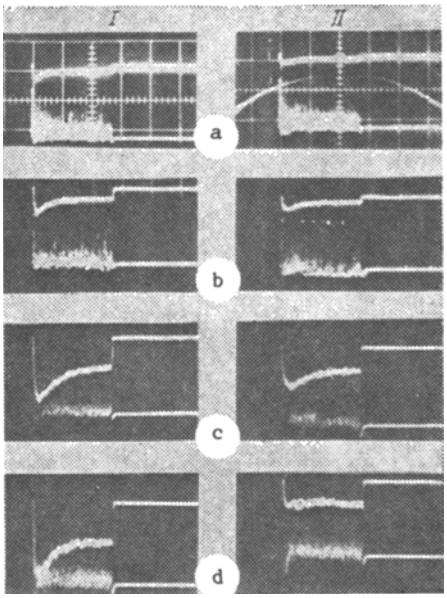

Fig. 2. Typical oscillograms showing the variation of the sonoluminescence and subharmonic intensities as the salt concentration of the water is increased. I) U = 110 V; II) U = 75 V; a) distilled water; b) C = 90 g/dm^3; c) 240; d) 270.

signal acquires a subharmonic. Now I_Σ deviates below the linear, indicating an increase in the absorption of ultrasound. At a certain value U* the absorption increases so rapidly that I_Σ no longer increases with U, but decreases as the latter is increased. As a result, the $I_\Sigma(U^2)$ curve acquires a dip. Then, after attaining a minimum at U = U**, the intensity I_Σ once again begins to grow, although not as rapidly as in the initial part of the curve. In distilled and tap water and in solutions with salt concentrations less than or equal to 100-150 g/dm^3, the threshold for the onset of luminescence lies in the range of ultrasonic intensities corresponding to the dip in the $I_\Sigma(U^2)$ curves, shifting to the right with increasing C relative to the voltage U** corresponding to the minimum I_Σ. For $C \gtrsim 200$ g/dm^3, e.g., in the saturated solution (Fig. 4), sonoluminescence sets in at voltages much greater than U**, when I_Σ one again increases until it reaches a plateau. The deviation of I_Σ from a linear dependence on U^2, like the magnitude of the dip in the $I_\Sigma(U^2)$ curve, decreases as the salt concentration is increased.

Thus, both the absolute value of the absorption of ultrasound by the cavitation zone and the relative increase in the absorption at the dip in the $I_\Sigma(U^2)$ curves decrease in the solutions in comparison with distilled water. We also note at this point that the sonoluminescence and subharmonic-generation thresholds increase with the salt concentration.

Luminescence sets in in pure water at a higher value of the off-duty factor, i.e., for a lower degree of excitation of cavitation nuclei. However, the maximum value of L in the solutions is higher; moreover, after the luminescence intensity attains a maximum, it decreases more rapidly in pure water than in the solutions. For example, for N = 1 (continuous insonification) L is almost an order of magnitude higher in a solution with C = 180 g/dm^3 than in distilled water.

DISCUSSION

One of the most general laws established in the present study in application to the cavitation zone (independently of the properties of the liquid) is the fact that the subharmonic-generation threshold practically coincides with the beginning of the departure of the $I_\Sigma(U^2)$ curve from linear, i.e., with the beginning of a significant increase in the absorption of acoustic energy in the cavitation zone, and the maximum rate of growth of the subharmonic signal is attained in the vicinity of the minimum I_Σ. Sonoluminescence, on the other hand, sets in at higher ultrasonic intensities, and the difference between the thresholds of these effects increases with the salt concentration of the solution.

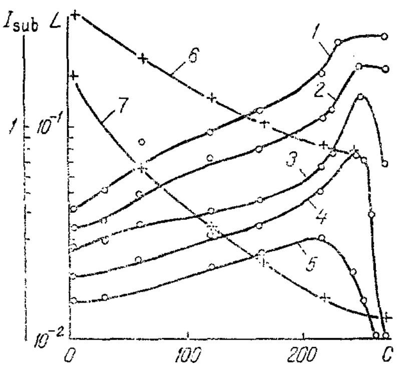

Fig. 3. Sonoluminescence intensity L (curves 1-5) and subharmonic intensity I_{sub} (curves 6 and 7) vs salt concentration C. 1) U = 150 V; 2) 123; 3) 105; 4) 75; 5) 35; 6) 130; 7) 140; T = 30 msec, τ = 3 msec; I_{sub} and L in relative units; C in g/dm³.

The increase in the ultrasonic absorption at intensities of the order of the subharmonic threshold is evidently attributable to the fact that this intensity is the threshold value for the rectified diffusion of dissolved gas from the liquid into bubbles. Upon attainment of the threshold intensity, the volume of the bubbles grows considerably, eliciting a corresponding increase in the absorption and, as a result, decreasing the signal intensity I_Σ, more than 90% of which forms the signal of frequency f_0 transmitted through the cavitation zone. The abrupt decrease of I_Σ [the dip in the $I_\Sigma(U^2)$ curves] begins when the bubbles become unstable and begin to implode (collapse), because, according to [6], fragments of the imploding cavitation bubbles form new cavitation nuclei, and their multiplication takes on the character of a chain reaction. As a result, the density of the bubbles rapidly attains saturation, whereupon I_Σ is observed once again to increase. The decrease in the depth of the dip in the $I_\Sigma(U^2)$ curves with increasing value of C obviously implies that the density of bubbles in the cavitation zone is lowered as the salt concentration is increased.

It should be noted that an important law also lies in the fact that the L(C) and $I_{sub}(C)$ curves exhibit a distinctly different behavior (see Fig. 3). This evinces a different mechanism underlying the onset of the subharmonic-generation and luminescence phenomena. Considering the fact that two fundamental types of bubble motion are known to occur in a cavitation zone, i.e., pulsation and collapse [4, 7], and pulsations have the lower threhsold, the foregoing results lead to the conclusion that the onset of the subharmonic is associated with bubble pulsations, while luminescence is a result of their collapse.

On the whole, however, the variation of the investigated characteristics of the cavitation zone (Figs. 2-5) when salt is dissolved in water is clearly attributable to the variation of the physicochemical properties of the liquid, which are capable of affecting the cavitation-bubble dynamics in some degree or other. Principal among such properties are the saturated vapor pressure P_v, the surface tension σ, the density ρ of the liquid, the relative gas content α, and the viscosity η. The last two parameters undergo extreme variations [8]. As the salt concentration is varied from zero to saturation, the dissolved-air content decreases by more than an order of magnitude, and the viscosity increases 70%. The surface tension increases roughly 12%, and the density 20%. The saturated vapor pressure at room temperature decreases insignificantly (≈10%). At temperatures close to the boiling point, however, P_v decreases in the salt-saturated water more than two orders of magnitude. Considering the heating of the gas in the bubbles to high temperatures at the time of collapse, we can expect an increase of P_v to have a substantial influence on the collapse process, slowing it down. Moreover, with an increase in P_v the specific heat and the heat-transfer coefficient to the surrounding liquid increase as a result of the increase in the density of the vapor—gas mixture in the bubble interiors. All these factors diminish the degree of heating of the bubble contents and, according to the thermodynamic theory of sonoluminescence (see, e.g., [4]), lower the intensity of the latter.

A decrease in the gas content, on the one hand, causes the concentration of cavitation nuclei to decrease and, on the other, slows down the evolution of the nuclei into actual

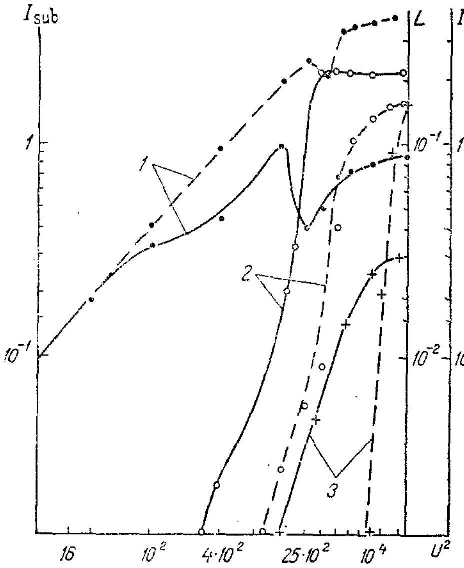

Fig. 4. Intensities vs transducer applied voltage squared U^2 in distilled water (solid curves) and in a salt solution at $C = 270$ g/dm^3, $T = 30$ msec, $\tau = 3$ msec. 1) total signal intensity I_Σ recorded by the hydrophone; 2) subharmonic intensity I_{sub}; 3) sonoluminescence intensity; I_Σ in relative units; U^2 in V^2.

cavitating voids. As a result, both the total density of bubbles in the cavitation zone and the density of pulsating but noncollapsing bubbles are decreased. This conclusion is corroborated by the fact that ultrasound is absorbed to a lesser degree in a cavitation zone in salt water than in distilled water, and also by the fact that the subharmonic intensity is lower in salt water. The decrease in the concentration and size of the cavitation nuclei as salt is dissolved in the water is also responsible for an increase in the necessary degree of excitation of the liquid in order for sonoluminescence to set in (Fig. 5) at a given excitation intensity.

With an increase in the surface tension, the force tending to compress the bubbles increases. Accordingly, the generation of cavitation at a given nucleus requires higher ultrasonic intensities. All things considered, the ultimate result is an increase in the cavitation threshold and, hence, an increase in the thresholds for the onset of subharmonic radiation and sonoluminescence in the salt solutions in comparison with distilled water.

If, as mentioned, we assume that sonoluminescence is caused by bubble collapse, we can then attribute the increase of the luminescence intensity in the salt solutions, despite the reduction in the total number of cavitating voids, to an increase in the rate of collapse of the bubbles and in the efficiency of conversion of their potential energy stored during expansion into thermal energy of the vapor—gas mixture and luminescence energy of the latter. The increase in the rate of collapse in the solutions has a number of causes. First, the bubble concentration decreases. As a result, the bubbles exert less of a slowing action on one another during collapse. Second, owing to the decrease of the saturated vapor pressure in salt water, the pressure inside the bubbles, which counteracts the compression forces, is not as high during collapse as in distilled water. Furthermore, an increase in the rate of collapse is also promoted by an increase in the surface tension, which compresses the bubbles. Finally, an increase in the viscosity stabilizes the shape of the bubbles in accordance with [9, 10] and, in so doing, prevents their disintegration in the early stage of collapse, ensures a more profound collapse, and thus induces a higher degree of compression of the gas and a higher luminescence intensity. The rapid decrease of the sonoluminescence intensity after attainment of the maximum for low voltages (see curves 4 and 5 in Fig. 3) appears to be associated with the fact that the ultrasonic intensity corresponding to a given U becomes insufficient for the excitation of cavitation at the existing nuclei in the case of high salt concentrations, because the concentration and sizes of the nuclei decrease as the salt concentration is increased.

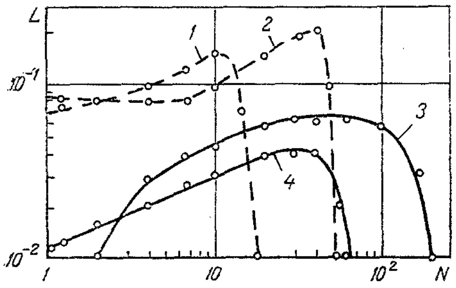

Fig. 5. Sonoluminescence intensity L vs ratio T/τ in distilled water (solid curves) and in NaCl solution (dashed curves) at C = 180 g/dm³. 1, 4) τ = 5 msec; 2, 3) 10.

TABLE 1. Cavitation Utilization of Acoustic Energy $\varkappa$, Efficiency γ of Conversion of Energy Spent in Cavitation into Subharmonic Energy, and Efficiency λ of Conversion of Energy Spent in Cavitation into Luminescence Energy Relative to Distilled Water (subscripts: 0 refers to C = 0, 1 refers to 180 g/dm³, and 2 refers to 270 g/dm³)

$\varkappa$, γ, λ	U, V									
	20	40	50	60	70	80	100	125	130	145
$\varkappa_0$	0,73	0,75	0,94	0,97	0,97	0,97	0,98	0,98	0,99	0,99
$\varkappa_1$	0,16	0,22	0,44	0,70	0,85	0,88	0,84	0,79	0,85	0,9
$\varkappa_2$	0,00	0,00	0,15	0,54	0,18	0,37	0,58	0,78	0,79	0,79
$\varkappa_1\mathrm{rel} = \varkappa_1/\varkappa_0$	0,22	0,29	0,47	0,72	0,88	0,94	0,86	0,81	0,86	0,91
$\varkappa_2\mathrm{rel} = \varkappa_2/\varkappa_0$	0,00	0,00	0,16	0,56	0,19	0,38	0,59	0,80	0,81	0,50
$\gamma_1\mathrm{rel} = (V^2_{1\mathrm{sub}}/V^2_{0\mathrm{sub}})\varkappa_0/\varkappa_1$	0,00	0,00	0,00	0,00	0,01	0,02	0,01	0,33	0,29	0,60
$\gamma_2\mathrm{rel} = (V^2_{2\mathrm{sub}}/V^2_{0\mathrm{sub}})\varkappa_0/\varkappa_2$	0,00	0,00	0,00	0,00	0,49	0,55	0,56	0,72	0,57	0,81
$\lambda_1\mathrm{rel} = (L_1/L_0)\varkappa_0/\varkappa_1$	0,00	0,00	0,00	1,93	2,3	3,1	7,7	8,1	7,8	6,3
$\lambda_2\mathrm{rel} = (L_2/L_0)\varkappa_0/\varkappa_2$	0,00	0,00	0,00	0,00	0,00	0,00	0,00	0,00	2,9	11

We now consider the variation of the cavitation utilization of acoustic energy [8] $\varkappa = (I_{\mathrm{rad}} - I_{\mathrm{tr}})/I_{\mathrm{rad}}$ (see Table 1), where I_{rad} is the intensity radiated at a given radiator voltage, and I_{tr} is the intensity after traversal of the cavitation zone. Making use of the fact that I_{rad} is proportional to U^2, we can determine $\varkappa$ from the relation $\varkappa = (V^2_{\mathrm{rad}} - V^2_{\Sigma})/V^2_{\mathrm{rad}}$, where V_{Σ} is the actual output of the receiving hydrophone, and V_{rad} is the signal that would be recorded from the hydrophone at the voltage U_i is sound were not absorbed in the cavitation zone.

The ratio $V^2_{\Sigma}/V^2_{\mathrm{rad}}$ for a given U_i is determined as the ratio of the ordinates of the points of intersection of the real function $I_{\Sigma}(U^2)$ or $V^2_{\Sigma}(U^2)$ with the straight line formed by the continuation of the initial linear part of this dependence from the abscissa U_i. Iernetti [5] has shown that it is preferable in the case of pulsed excitation of cavitation to determine $\varkappa$ through the volume acoustic energy $W_{\mathrm{cav}}/W_{\mathrm{rad}}$, where W_{rad} is the radiated acoustic energy density averaged over time and space, and W_{cav} is the acoustic energy density, averaged over time and space, that is spent in all cavitation processes. It is evident from the table that $\varkappa$ is higher in distilled water than in the aqueous salt solutions. This is caused by the large concentration of cavitation nuclei and bubbles in pure water. As a result, a large fraction of energy is spent in the formation and maintenance of the cavitation zone. However, the efficiencies γ and λ of conversion of this fraction of the energy into (respectively) luminescence energy and subharmonic-generation energy, defined in accordance with [5], differ in different liquids; γ turns out to be higher in water than in the solutions, whereas λ is higher in the solutions. It can be inferred from this fact that an increase in the concentration of cavitation nuclei and bubbles in the cavitation zone will not necessary increase the efficiency of utilization of acoustic energy for the excitation of secondary cavitation effects such as, e.g., subharmonic generation, light emission, or cavitation erosion. If the concentration of bubbles and dissolved gas is too high, the bubbles will cease collapsing efficiently, and they will clearly grow to large dimensions and pulsate, predominantly without collapsing.

NOTATION

I_Σ, intensity of the total (integrated) acoustic signal; L, sonoluminescence intensity; I_{sub}, subharmonic intensity; f_0, frequency of excited oscillations; T, period; τ, pulse duration; U, voltage on transducer; V_Σ, total acoustic signal received by hydrophone; V_{sub}, subharmonic signal; C, salt concentration; $\varkappa$, cavitation utilization of acoustic energy; γ, λ, efficiencies of conversion of energy spent in cavitation into subharmonic-generation and luminescence energy, respectively; N, pulse off-duty factor; ν, pulse repetition rate.

LITERATURE CITED

1. M. S. Plesset, Trans. ASME, Ser. D: J. Basic Eng., 85, No. 3, 360-364 (1963).
2. M. S. Plesset, Corrosion, 18, No. 5, 181-188 (1962).
3. W. H. Wheelen, in: Proc. 1955 NPL Symp. Cavitation in Hydrodynamics, Paper 21 (1956).
4. R. T. Knapp, J. W. Daily, and F. G. Hammitt, Cavitation, McGraw-Hill, New York (1970).
5. G. Iernetti, Acustica, 23, 198-207 (1970).
6. M. G. Sirotyuk, in: High-Intensity Ultrasonic Fields, L. D. Rozenberg (ed.), Plenum Press, New York—London (1971), pp. 263-343.
7. V. A. Akulichev, in: High-Intensity Ultrasonic Fields, L. D. Rozenberg (ed.), Plenum Press, New York—London (1971), pp. 203-259.
8. Handbook of Chemistry [in Russian], Vol. 3, Moscow (1978).
9. A Francescutto and R. Nabergoi, Acustic, 41, 215-220 (1978).
10. W. Lauterborn, in: Proc. 1973 Symp. Finite-Amplitude Wave Effects in fluids, Copenhagen (1974), p. 195.