Secondary ultrasonic atomisation mechanisms

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Abstract. Liquid atomisation is the basis for various practical applications such as pharmaceuticals, cosmetics, food industry, etc. In this case, the main challenge is the development of high-performance, highly dispersed atomisation systems. Generally, high-throughput atomisation systems form aerosol with large droplet sizes, and high-disperse atomisation systems have low performance. Secondary ultrasonic atomisation can solve this problem by allowing non-contact crushing of already formed aerosol droplets with large droplet sizes by means of a high-intensity ultrasonic field. For this purpose, a pre-generated stream of liquid droplets is directed into a cylindrical region formed by an emitter in which a high-intensity ultrasonic field is generated. Ultrasonic radiator, is a tube of stepped-variable cross-section, providing the formation of bending-diametral or diametral oscillations at a frequency above 20 kHz. At sufficiently high ultrasound intensity, conditions for further crushing of liquid droplets are realised, which leads to the formation of a highly dispersed aerosol. This paper describes the proposed mathematical model of the atomisation process and finds the regularities of the process depending on the determining parameters of the ultrasonic field and physical and chemical properties of the liquid. Two mechanisms of jet destruction are revealed: direct destruction of droplets when they hit the ultrasonic wave front and cavitation mechanism of droplet and jet destruction. The dominant crushing mechanism depends on the problem parameters and, in turn, determines the minimum size of the resulting droplets. The results of this work will help to optimise the secondary ultrasonic atomisation process and improve liquid atomisation technologies in various applications.

1 Introduction

Ultrasonic atomisation of liquids is widely used in industrial applications such as fuel combustion [1], emulsification of nanoemulsions [2], spray cooling [3], wet dust removal [4] and air humidification [5], spray pyrolysis [6], improving the efficiency of existing gas cleaning equipment [7], etc. The advantages of ultrasonic atomisation over other methods are the high dispersibility of the resulting droplets with a narrow size distribution, which is important especially in medical, chemical and pharmaceutical applications [8-10]. Another

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advantage of the ultrasonic liquid atomisation method is the high efficiency, up to 1 %, compared to other atomisation methods [11, 12].

However, for some practical applications, high liquid droplet generation performance is required, which is a challenge for the ultrasonic atomisation method. Moreover, the higher the required aerosol dispersity, the lower the throughput [11]. For example, not more than 0.1 ml/(s·cm^2) when generating droplets up to 10 μm in diameter.

In [13], a modified method of two-stage ultrasonic liquid atomisation is proposed. Its peculiarity lies in the second stage, when the aerosol stream generated by the traditional ultrasonic method is subjected again to powerful ultrasonic influence. According to the model proposed in this paper, the droplets will continue to be crushed as they pass through the ultrasonic field. The increase in productivity while maintaining the target droplet size (less than 10 μm in diameter) is achieved through the following idea. Primary ultrasonic atomisation of liquid is proposed to be carried out with maximum productivity but low droplet dispersion. And at the secondary ultrasonic crushing to achieve the necessary dispersity.

The proposed idea has obvious prospects for practical application and deserves further research and improvement. It is not necessary to use ultrasonic atomisation as a method of primary atomisation. Secondary ultrasonic crushing can be subjected to a stream of liquid droplets formed by any method, as well as a jet of solid liquid, fed into a region with a powerful ultrasonic field under pressure.

The mechanisms of ultrasonic dispersion of a liquid layer are well studied [14, 15]. Dispersion occurs due to the detachment of liquid droplets from the crests of capillary waves created on the liquid surface by ultrasonic vibrations. Ultrasonic dispersion is also affected by cavitation activity in the liquid layer. According to the "conjunction" theory, droplet formation is the result of the interaction between random hydraulic shock waves generated by cavitation perturbation and capillary waves [16].

However, the processes of secondary atomisation occurring in the flow of droplets or liquid jets passing through a powerful ultrasonic field have not been considered so far. Obviously, these processes are complex, and not one but different crushing mechanisms can be realised. Coagulation of droplets and evaporation of liquid may also occur. Therefore, it is to be expected that both ultrasonic parameters and conditions (temperature, humidity) as well as liquid properties influence the atomisation result (droplet size, process performance).

The aim of the work is to theoretically investigate the process of secondary ultrasonic atomisation and to identify its possible mechanisms.

2 Problem statement

A tubular transmitter was considered as a source of ultrasonic oscillations. Tubular transmitter is a tube of a stepped-variable cross-section, providing the formation of bending-diametral or diametral oscillations at a frequency above 20 kHz. The use of this type of transmitter allows the creation of a regular oscillation structure of large extent. Figure 1A shows a sketch of a tubular transmitter assembly with a longitudinally oscillating ultrasonic piezoelectric transducer. To increase the frequency of oscillation in real conditions, a multi-package piezoelectric transducer, considered in [17, 18], can be used. The shape of oscillations is presented in Figure 1B, and the formed ultrasonic field is presented in Figure 1C.

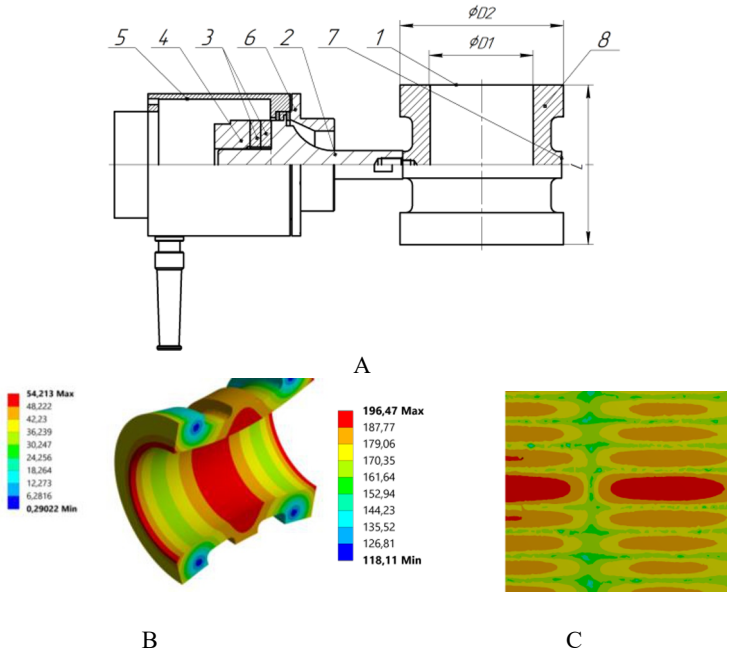

Fig. 1. Scheme of the tubular radiator: A - Sketch of the tubular transmitter; B - Oscillation shape; C - distribution of the formed sound pressure level; 1 - radiating element in the form of a bending and oscillating tube; 2 - concentrator of the piezoelectric transducer (radiating pad); 3 - piezoceramic elements; 4 - reflective pad; 5 - housing; 6 - flange; 7 - amplifying area for the threaded bores; 8 - amplitude correction area of the transmitter ends; L - transmitter length; D1 - inner diameter; D2 - outer diameter.

Calculation of the ultrasonic field formed by the tube transmitter was performed by means of harmonic acoustic type of analysis. Calculation of the oscillation shape of the tubular transmitter was performed using the modal type of analysis.

To atomise a liquid in a high-intensity ultrasonic field, a stream of droplets or a liquid jet is fed into a cylindrical cavity formed by the transmitter, and a cloud of finely dispersed aerosol is obtained at the outlet of the transmitter.

Two problems have been solved: The first one is droplet stream breaking. The second is atomisation of the jet fed into the ultrasonic radiator under pressure.

1. Let us consider the flow of droplets with initial diameter D_0, moving in air in a hollow cylinder with velocity v_l. A powerful ultrasonic field is created in the cylindrical cavity with a sound level of L_p, with frequency ω. To what size can the droplets collapse?

2. Let us consider the case of destruction of a liquid jet of diameter D_f, moving under pressure p_0 in a cylinder in a powerful ultrasonic field. What will be the droplet size in this case? Let us consider several probable mechanisms of droplet and jet crushing.

3 Mathematical model of secondary ultrasonic liquid atomisation

3.1 Direct droplet destruction at the ultrasonic wave front

We consider a drop of diameter D, falling into the front of the sound wave. Analysing the forces acting on it, we obtain following [15] the values of the threshold intensity of particle destruction:

$$I_t = 2W_l \left(\frac{\sigma_{stp}}{D\omega\rho_l} \right)^2 \tag{1}$$

Where σ_{stp} is particle strength, N/m ^{2}, ω is radiation frequency, ρ_l is liquid density, kg/m ^{3}, $W_l = \rho_l c$ is wave impedance, c is sound velocity, m/s ^{2}. It should be remembered that the strength of water under pulse impact is several orders of magnitude lower than the reference (theoretical) value [19].

From equation (1), we obtain an expression for the minimum drop diameter to which a drop can be destroyed at a given level of impact intensity I:

$$D_{\min} = \frac{\sigma_{stp}}{\omega\rho_l} \sqrt{\frac{2W_l}{I}} \tag{2}$$

The intensity of exposure is related to the sound pressure level p by a known relationship:

$$I = \frac{p^2}{2W_l} \tag{3}$$

Given the expression linking the sound level L_p with sonic pressure $p = 20 \cdot 10^{-6} \cdot 10^{\frac{L_p}{20}}$, we obtain the dependence of minimum diameter $D_{\min}$, to which a drop can be crushed, from the sound level (Figure 2).

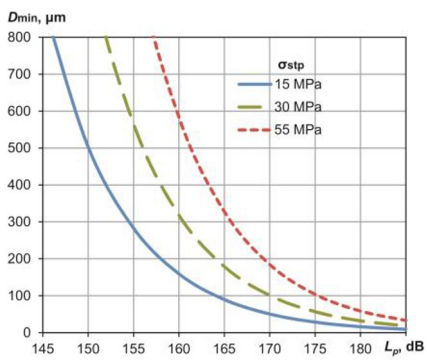

Fig. 2. Dependence of the minimum droplet size on the sound level for different values of the particle tensile strength σ_{stp}

If the initial size of the droplet is larger than 150 μm, a sound level less than 160 dB will not lead to its destruction according to the proposed direct destruction mechanism. It should be noted that the strength of the drop σ_{stp} is a free parameter of the model, requiring experimental determination. The minimum droplet size also depends on the exposure frequency (Figure 3).

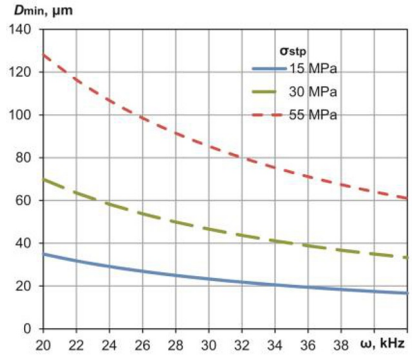

Fig. 3. Dependence of the minimum droplet size on the ultrasonic frequency, calculation for L_p = 174 dB

Next, let us determine the time that a drop needs to be in the ultrasonic field to collapse to the minimum values. Let the rate of destruction $\frac{dD}{dt} = v_f$ is constant. It is necessary that during the time of movement in the ultrasonic field the drop has time to collapse from the initial to the minimum value. Time in the field t_{exp} will be determined by the velocity of the droplet v_l and cylinder length $L_t - t_{exp} = L_t / v_l$. Fracture time is $t_f = \frac{D_0 - D_{\min}}{v_f}$. Then the necessary condition for destruction will be $t_{\exp} \geq t_f$, or

$$\frac{v_f}{v_l} > \frac{D_0 - D_{\min}}{L_t} \tag{4}$$

Considering that the length of the hollow cylinder is much larger than the size of the drop, condition (4) is fulfilled in most cases.

3.2 Cavitation destruction of a droplet

Further, a possible mechanism of drop destruction is presented, if the energy according to equation (1) will be insufficient (intensity of impact is less than threshold for a given drop size).

It is known that ultrasound of sub-threshold intensity can create cavitation inside a liquid droplet if there is enough time and energy for it. In this case, since the drop is moving, cavitation bubbles will not collapse, but only expand [20].

Suppose that the bubbles, expanding, merge inside the spherical drop into a single bubble. If the pressure in the bubble is greater than atmospheric pressure, then at some point the drop will collapse like a soap bubble, with the formation of small drops whose diameter corresponds to the thickness of the wall of the "soap bubble" at the moment of collapse. Such a model is proposed and confirmed experimentally in [21] for two methods of liquid atomisation. Let us consider it in relation to the problem posed.

In the work of Kedrinsky [20], the concept of cavitation coefficient equal to the ratio of the volume of cavitation bubbles to the liquid volume was introduced. In the case of developed cavitation, this ratio is equal to k = 0.2-0.3.

Let the volume of the drop before bubble expansion be $W_0 = \frac{\pi D_0^3}{6}$. The ultrasonic field creates conditions for the development of cavitation, and the total volume of bubbles W_b

almost instantly becomes equal to 20-30 % of the liquid volume in the drop (W_b / $W_0 = k$). Since the droplet is small in size (from tens of μm to mm), closely spaced bubbles in it merge into one and continue to expand, stretching the walls of the droplet. Bubble diameter D_b will be defined as:

$$D_b = \sqrt[3]{\frac{6 \cdot W_b}{\pi}} = \sqrt[3]{\frac{6 \cdot k \cdot W_0}{\pi}} = \sqrt[3]{k} D_0 \tag{5}$$

Volume of the expanded bubble before the bubble collapses will be equal to: $W_{\max} = \frac{\pi D_{\max}^3}{6}$.

The processes under consideration have a high speed, so the expansion of the cavitation bubble can be considered adiabatic:

$$\frac{D_{\max}}{D_b} = \left(\frac{p_0}{p_{\min}} \right)^{\frac{1}{3\gamma}}, \tag{6}$$

here γ is liquid vapour adiabatic index, $p_{\min}$ – pressure in the bubble at the moment of collapse, Pa, p_0 – initial pressure in the droplet, Pa.

The pressure in the bubble before collapse can be calculated from the relation of equality of the internal energy of vapour in the bubble and surface energy:

$$p_{\min} W_b = \sigma S_b, \tag{7}$$

Taking into account (6) and (7), we obtain the equation for determining $D_{\max}$:

$$D_{\max} = \sqrt[3]{k} D_0 \left(\frac{p_0 D_{\max}}{6\sigma} \right)^{\frac{1}{6\gamma}}. \tag{8}$$

The external diameter of the droplet at the moment of fracture is denoted by D_{end}. Taking into account the condition of equality of liquid mass of a drop,

$$D_{end}^3 = D_0^3 + D_{\max}^3 \tag{9}$$

It can be assumed that the minimum size of new drops will be equal to the thickness of the liquid layer at the destruction of the cavitation bubble. The thickness of the water layer at the moment of destruction, and hence the size of new droplets, will be as follows D_{drop} = ($D_{end} - D_{\max}$) / 2. Or, considering (9)

$$D_{drop} = \left(\sqrt[3]{D_0^3 + D_{\max}^3} - D_{\max} \right) / 2 \tag{10}$$

where $D_{\max}$ is determined from the equation (8).

Figure 4 shows the dependence of the resulting droplets on the initial droplet diameter (before fracture) at different values of the coefficient k. The calculation is made for water at p_0 = 0.1 MPa. The smaller the initial droplet, the smaller fragments it will collapse into.

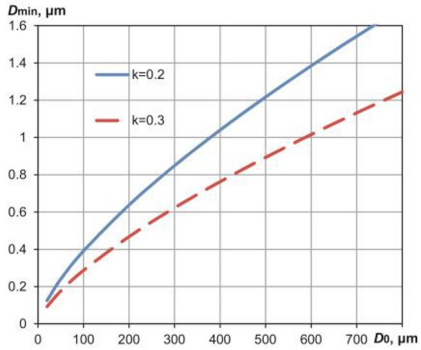

Fig. 4. Dependence of the minimum diameter of new water droplets formed as a result of cavitation destruction of droplets with diameter of D_0 of original diameter

3.3 Cavitation breakdown of jets

Let us consider the case of destruction of a liquid jet of diameter Df moving in a powerful ultrasonic field. Let the liquid be atomised under pressure p0. If the condition (1) is fulfilled, where $D = D_f$, the jet will collapse according to the direct mechanism described in (1) up to the droplet size D_{min}. But in case D_f is greater than some critical value, direct destruction will not occur. But the mechanism of cavitation destruction of the jet is possible. Let us estimate the droplet size in this case.

In the jet passing in the ultrasonic field, acoustic oscillations are generated, creating conditions for cavitation. In the unloading phase of the sound wave, a gap of width Z appears in the liquid, in which vapours accumulate. The following compression phase causes the formation of a bubble filled with liquid vapours. The value of Z is obtained from the known relations for wave processes:

$$Z = \frac{1}{\omega}\sqrt{\frac{I}{W_l}} \tag{11}$$

Since the cavitation bubble is formed on the basis of a rupture, in the volume of the jet it is possible to distinguish elements consisting of bubbles surrounded by liquid, and, the vapour volume in the bubble in relation to the liquid volume will be expressed by the cavitation coefficient k discussed above. Each bubble and fluid element will be a "soap bubble" that will collapse at the moment of maximum expansion of the cavitation bubble. Maximum diameter of the bubble before collapse:

$$D_{\max} = \frac{\sqrt[3]{k}}{\omega}\sqrt{\frac{I}{W_l}}\left(\frac{p_0 D_{\max}}{6\sigma}\right)^{\frac{1}{6\gamma}} \tag{12}$$

Instead of the equation (10) we obtain:

$$D_{drop} = \left(\sqrt[3]{Z^3 + D_{\max}^3} - D_{\max}\right)/2 \tag{13}$$

The joint solution of equations (11), (12) and (13) will give an estimate of the minimum size of new droplets depending on the characteristics of ultrasonic influence and liquid properties.

Figure 5 shows the dependence of droplets on hydrostatic pressure. The calculation is made for water, $k = 0.25$.

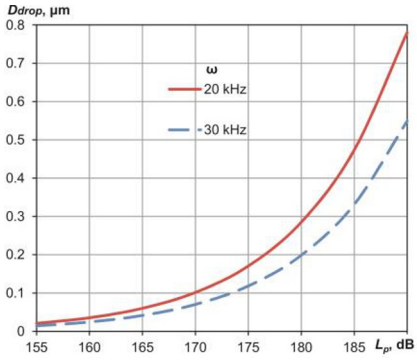

Fig. 5. Dependence of the diameter of new water droplets formed at cavitation breakup of the jet on the sound level

Figure 6 shows the dependence of the minimum droplet size on hydrostatic pressure.

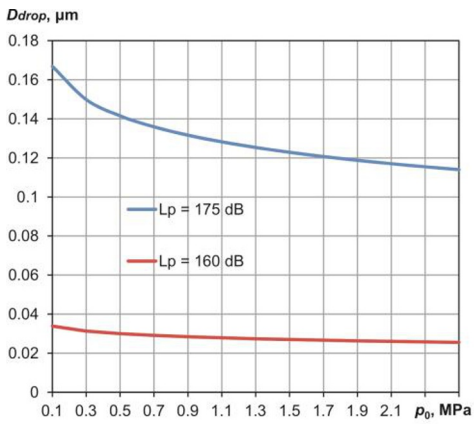

Fig. 6. Dependence of the diameter of new water droplets formed at cavitation jet destruction on hydrostatic pressure

4 Results and discussion

The paper proposes two mechanisms of liquid droplets and jets destruction when they hit a powerful ultrasonic field - direct and cavitation.

If the droplet size does not exceed some critical size (in our estimates, about 150 μm for a sound level of 160 dB), it can be destroyed by the direct impact of the sound wave front when it flies into this front. The minimum size of the resulting droplets in this case depends significantly and non-linearly on the sound level, falling with increasing sound level (Figure 2) and with frequency (Figure 3), and is independent of the size of the original droplet.

If the droplet size exceeds the critical size for a given sound level, it can be destroyed by the cavitation mechanism. Ultrasonic cavitation occurs inside a sufficiently large droplet, cavitation bubbles merge into one and expand. The droplet thus becomes similar to a soap bubble. When the wall strength limit, which depends on surface tension forces, is reached, the bubble bursts into fragments with a diameter on the order of the thickness of

the bubble wall. The size of the resulting droplets is smaller the smaller was the size of the initial droplet and the higher is the cavitation coefficient (Figure 4).

Destruction of the jet by cavitation mechanism is also possible. In this case, a slight decrease in droplet size will be observed with increasing atomisation pressure of the jet (Figure 6).

When describing the cavitation mechanism of jet destruction, calculations revealed a counterintuitive increase in droplet diameter with increasing sound level (Figure 5). This is explained by the fact that the magnitude of cavitation rupture in the continuous medium of the jet with the increase in sound level grows in accordance with the degree ½, the size of the cavitation element, i.e., the volume of water carried by the bubble, also grows. So, according to this model, the droplet size will also grow. But the considered mechanism will work only when some sound level is exceeded, when the cavitation regime in the jet is established. Thus, if a cavitation mechanism for secondary atomisation of jets is realised, the droplet size will only increase as the sound level increases. If it is required to achieve the minimum droplet size at cavitation destruction of the jet, it is necessary to influence it with the minimum sound level at which cavitation occurs in general.

5 Conclusion

Liquid atomisation is an important technological process for practice. It is particularly difficult to obtain fine aerosols with high production rates. For this purpose, it is proposed to use secondary ultrasonic atomisation, to which a stream of relatively large droplets or a jet of solid liquid is subjected. The mechanisms of such atomisation have not yet been studied.

We proposed and considered a model of secondary destruction of droplets and solid jets of liquid at their movement in the ultrasonic field, suggesting two possible mechanisms of destruction - direct destruction when the liquid hits the front of the acoustic wave and cavitation mechanism. Depending on the sound level and atomisation conditions, one or another mechanism of droplet and jet destruction can be realised (at relatively lower sound levels, cavitation, and at higher sound levels, direct). The model allows estimating the minimum droplet size at destruction, depending on the initial droplet size, liquid properties, and hydrostatic pressure. Thus, prerequisites for optimisation of the process of secondary ultrasonic atomisation of liquid have been created.

Free parameters of the model that require experimental determination - the liquid strength limit σ_{stp}, and cavitation coefficient k. We can consider the dependence of this coefficient on the parameters of the ultrasound field as a model development: the greater the ultrasound intensity, the larger the cavitation coefficient. A more complex dependence on frequency is possible.

We have not considered the slower processes of coagulation and evaporation that can take place under the influence of powerful ultrasound. These processes take place over longer periods of time. On the other hand, evaporation of small droplets, which are formed during secondary atomisation, occurs the faster and easier the droplet size. At high concentrations of such droplets, their coagulation cannot be neglected. Accounting for such processes is included in the development plans of the proposed model.

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