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Acoustic method of resonant length calculation of ultrasonic waveguides for nanodispersions

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Abstract

In this article, the parameters required for an acoustic waveguide (concentrator) to produce acoustic cavitation effects in an ultrasonicator are derived. The derivation is based on the solutions to complex valued wave equations. Based on the derived equations, the length of the concentrator required to produce acoustic cavitation effects for various concentrator shapes can be found. The theoretical results are confirmed through experiment.

Keywords: acoustic cavitation; ultrasonic dispersion; nanotechnology; nanopowder; resonance; waveguides; concentrator.

Introduction

Ultrasonic dispersers use ultrasonic vibrations to break up particle agglomerates in a liquid in order to create a homogeneous dispersion of the particles. Under the effects of ultrasonic dispersion, the resultant suspensions display a dispersity increase by several orders of magnitude compared to traditional mechanical dispersion techniques. Often, ultrasonic dispersion effects can be enhanced with acoustic cavitation. In acoustic cavitation, the sound wave energy is greatly increased upon the pulsation and collapse of the cavitation bubble and can lead to changes in chemical and physical properties as well as improve chemical processes [1].

In recent years, scientific interest in ultrasonic dispersion has increased due to the development of nanotechnology [2, 3]. The effective use of nanoparticles in many technologies requires a uniform distribution of the particles in a liquid and thus dispersion is an essential part of the production process. Investigations into the dispersion of nanoparticle have demonstrated significant advantages of ultrasound compared to other technologies [4, 5, 6]. Nanoparticles well dispersed through acoustic cavitation have been found to improve the properties of liquids such as paints (see for example [7, 8]). Acoustic cavitation can also be used to improve sonochemical processes [9].

In order to create the acoustic cavitation effect, special devices called waveguides are used. The waveguides used for ultrasonic cavitation are called "concentrators" [10]. The concentrator is a device used to magnify the ultrasound intensity. There are two types of concentrators: focusing and rod type; which work according to different principles of action [11]. The latter is the focus of our studies. The rod concentrator is a rigid solid in which either its cross-section or density varies along its length. The variation in the cross-section or density causes change in the displacement amplitude of particles through momentum conservation (the principle of action of rod-type concentrators). In this study, variations in the cross-section of rod-type concentrators are explored with the density kept constant.

Theoretical Methods

In order to efficiently use the energy of the ultrasonic vibrations for the dispersion of nanopowders by acoustic cavitation, it is necessary to correctly calculate the geometric parameters of the concentrator. Theoretical studies have been carried out to determine the resonant shape of rod concentrators based on their wave equations through the use of functions of complex variables.

We introduce three assumptions to derive the wave equation for the concentrators:

1. The front of the stress wave propagating along the axis of the concentrator is flat.

2. The stresses produced in the concentrator are uniformly distributed across the cross-section.

3. The longitudinal deformations in the concentrator are elastic.

Implementation of the first two conditions leads to diffusion of the longitudinal oscillation mode in the concentrator such that the diameter of the concentrator D is less or equal to half of the wavelength λ of the sound oscillation

$$D \leq \frac{\lambda}{2}. \qquad (1)$$

The third condition excludes heating of the concentrator and is met at $\sigma_m < \sigma_{02}$ where σ_m is the value of the stresses in the concentrator and σ_{02} is the elastic limit of the concentrator material.

Under these assumptions, the displacement u of a cross section S of the concentrator in the axial direction x is associated with the relative elongation $\frac{\partial u}{\partial x}$ (deformation) by

$$du = \frac{\partial u}{\partial x} dx \qquad (2)$$

The elastic force F causing this displacement is proportional to the product of the relative elongation and the cross-sectional area as described by

$$F = -ES \frac{\partial u}{\partial x} \qquad (3)$$

where E is the Young's modulus (elasticity) of the concentrator's material.

The differential equation of motion of a unit volume of the concentrator can be written as follows:

$$\frac{\partial V}{\partial t} = -\frac{1}{\rho S} \frac{\partial F}{\partial x} \qquad (4)$$

where V is oscillatory velocity of a given cross section of the concentrator, ρ is the density of the concentrator, and t is time.

Thus, by using equation (3) together with (4), it is a simple task to obtain the basic differential equation describing the change of the elastic force in time in relation to the change in the oscillatory velocity along the concentrator

$$\frac{\partial F}{\partial t} = -ES\frac{\partial V}{\partial x} \quad . \tag{5}$$

Taking into account the harmonic nature of oscillations, we can represent the expression for elastic force and oscillatory velocity as follows:

$$F = F_m \sin(\omega t + \varphi_F)$$

$$V = V_m \sin(\omega t + \varphi_V).$$

Transforming into a complex form using [12]:

$$F \leftrightarrow \dot{F} e^{j\omega t}$$

$$V \leftrightarrow \dot{V} e^{j\omega t},$$

where $\dot{V} = \frac{1}{\sqrt{2}} V_m e^{j\varphi_V}$ is the effective complex oscillatory velocity and $\dot{F} = \frac{1}{\sqrt{2}} F_m e^{j\varphi_F}$ is the effective complex oscillatory force.

In the complex form, the differential equations (4) and (5) become:

$$-\dot{V} = \frac{1}{j\omega\rho S}\frac{d\dot{F}}{dx} \tag{6}$$

$$-\dot{F} = \frac{ES}{j\omega}\frac{d\dot{V}}{dx} \quad . \tag{7}$$

In general, the cross-sectional area of the concentrator depends on the coordinate x. Thus by differentiating expression (6) with respect to x we obtain:

$$-\frac{d^2\dot{F}}{dx^2} = j\omega\rho\left(\dot{V}\frac{dS}{dx} + S\frac{d\dot{V}}{dx}\right) \tag{8}$$

When we substitute the values $\dot{V}$ and $\frac{d\dot{V}}{dx}$ from (6) and (7) in the last expression, we obtain the wave equation of the concentrator in the complex form:

$$\frac{d^2\dot{F}}{dx^2} - \frac{S'}{S}\frac{d\dot{F}}{dx} + k^2\dot{F} = 0, \qquad (9)$$

where $k = \frac{\omega}{c} = \frac{2\pi f}{c} = \frac{2\pi f}{\sqrt{E/\rho}}$ is the wave number, $S' = \frac{dS}{dx}, S = \pi R^2$ is the cross-sectional area of the concentrator.

Taking into account the above relations, equation (9) can be rewritten as follows:

$$\frac{d^2\dot{F}}{dx^2} - \frac{2R'}{R}\frac{d\dot{F}}{dx} + k^2\dot{F} = 0 \qquad (10)$$

where $R' = \frac{dR}{dx}$.

Analogously, we can obtain the wave equation of the concentrator in the complex form with respect to oscillatory velocity:

$$\frac{d^2\dot{V}}{dx^2} + \frac{2R'}{R}\frac{d\dot{V}}{dx} + k^2\dot{V} = 0 \qquad (11)$$

Thus, the calculation of the concentrator is reduced to determining its resonant length by solving the equation (10) or (11) under the following boundary conditions:

$$\left|\dot{V}'(0)\right| = \left|\dot{V}'(l)\right| = 0 \qquad (12)$$

$$\left|\dot{V}'(0)\right| = V_0 \qquad (13)$$

where l is the length of the concentrator, V_0 is the vibrational velocity applied by the transducer, and $\dot{V}' = \frac{d\dot{V}}{dx}$.

A concentrator calculated on the basis of (12) will be resonant and its joining to the electromechanical transducer will not change its operational mode (i.e. the condition (13) will

be satisfied). Thus, based on these considerations, the most efficient rod concentrator length to produce ultrasonic cavitation effects for the dispersion of nanoparticles can be calculated.

Experimental Methods

An experimental setup was created to test if the theoretically calculated concentrators would produce the desired ultrasonic cavitation effects. As a test substance, silica nanopowders (Tarkosil, type T05B06) with an average particle size of 53 nm was subjected to ultrasonic vibrations in water with a concentrator calculated according to our theoretical results. Copper nanoparticles were also dispersed in base motor oil. The silica nanoparticles were dispersed at a concentration of 0.1 wt% and the copper nanoparticles 0.5 wt%.

An ultrasonic generator IL10-0.63 with a magnetostrictive transducer and interchangeable concentrator were used for the dispersion of the nanopowders as shown in Figure 1. The generator allows for adjustment of the frequency within a small range (± 5-10%) of the operating frequency. The operating frequency of the generator is 23000 Hz and power 630 watts.

The concentrator was constructed from the titanium alloy VTZ-1. The ultrasonication time was 3 minutes unless otherwise stated.

Results and discussion

Based on the derived complex form wave equation (11), we calculate several resonant rod concentrator shapes. First, we identify the family of generatrices ***R(x)***, in which the wave equation has an analytical solution. To do this, we represent the complex oscillatory velocity in the form $\dot{V} = \frac{\dot{u}(x)}{R(x)}$ and substitute it into equation (11). After reducing the equation into a canonical form, we obtain:

$$\frac{d^2\dot{u}}{dx^2} + \left(k^2 - \frac{R''}{R}\right)\dot{u} = 0 \qquad (14)$$

The substitution applied to the boundary conditions (12) and (13) give the following conditions:

$$\left|\dot{u}'(0)\right| = \left|\dot{V}'(0)\right| R_0', \qquad (15)$$

$$\left|\dot{u}(l)\right| = \left|\dot{V}'(l)\right| R_l', \tag{16}$$

$$\left|\dot{u}(0)\right| = V_0 \dot{R}_0 \ . \tag{17}$$

Then, equation (14) under the condition that

$$k^2 - \frac{R''}{R} = k'^2 = const > 0 \tag{18}$$

has solutions of the form

$$\dot{u} = \dot{A} \cdot sin(k'x + \varphi), \tag{19}$$

where $\dot{A}$ and φ are integration constants and k' is the wave number which takes into account the dispersion of the phase velocity in the concentrator.

The constants of integration are determined from the boundary conditions (15 - 17):

$$\left|\dot{A}\right| sin\,\varphi = \left|\dot{V}_0\right| R_0, \tag{20}$$

$$\left|\dot{A}\right| k' cos\,\varphi = \left|\dot{V}_0\right| R_0', \tag{21}$$

$$\varphi = arctan\left(k' \frac{R_0}{R_0'}\right) + n\pi, \tag{22}$$

where

$$\left|\dot{A}\right| = \frac{\left|\dot{V}_0\right|}{k'} \sqrt{R_0'^2 + \left(k' R_0\right)^2} = \frac{k}{k'} \left|\dot{V}_0\right| R_0 \ . \tag{23}$$

Therefore, equation (19) takes the form:

$$\dot{u}(x) = \frac{k}{k'} \left|\dot{V}_0\right| R_0 \, sin\left(k'x + arctan \frac{k' R_0}{R_0'}\right). \tag{24}$$

In order to determine the resonant length of the concentrator, we use the boundary condition on the outlet of the concentrator

$$\dot{A}\sin\left(k'l+\varphi\right)=\left|\dot{u}_1\right|R_1, \qquad (25)$$

$$k'\dot{A}\cos\left(k'l+\varphi\right)=\left|\dot{u}_1\right|R_1'. \qquad (26)$$

Here, $\tan\left(k'l+\varphi\right)=k'\frac{R_1}{R_1'}$ and taking into account that $\tan\varphi=k'\frac{R_0}{R_0'}$, we obtain

$$\tan k'l=\frac{k'\left(\frac{R_0'}{R_0}-\frac{R_1'}{R_1}\right)}{\frac{R_0'}{R_0}\frac{R_1'}{R_1}+k'^2}. \qquad (27)$$

Using equation (27), the resonant length of concentrators of various shapes can be determined. There are three types of functions that satisfy condition (18). They are characteristic in that $\frac{R''}{R}$ is constant for them. The functions are:

1) $R(x)=a\,x+b$; a straight line. If $a\neq 0$, the concentrator is conical; if $a=0$, the concentrator is cylindrical.

2) $R(x)=a\,e^{-\beta x}+b\,e^{\beta x}$; a catenary line. If $b=0$, the concentrator is exponential; if $a=b$, the concentrator is a catenoid.

3) $R(x)=a\cos(\beta x+\varphi_0)$; trigonometric curves. The concentrator will have a dumbbell shape.

For all the other functions that do not satisfy condition (18), the wave equation does not have an analytical solution.

We now give an analysis of the solutions for the resonant length of conical concentrators as they are common concentrator shapes and easy to produce.

For the concentrators whose radius varies linearly, $R(x)=a\,x+b$, $R'(x)=a$, and $k'=k$. Thus,

$$R(x)=\frac{R_1-R_0}{l}x+R_0.$$

Using $N=\frac{R_0}{R_1}$, the correlation between the input and output radius of the concentrator, the resonant length for a conical concentrator is calculated by

$$\tan kl = \frac{kl}{\frac{N(kl)^2}{(1-N)^2} + 1} \cdot \qquad (28)$$

In case of $R_o = R_1$, a cylinder (where $N = 1$), the resonant length is equal to

$$l = \frac{n\pi}{k} . \qquad (29)$$

where n = 1, 2, 3, …

If $N = \infty$ (i.e. for a pointed cone), we obtain from equation (28)

$$\tan k\,l = k\,l \ . \qquad (30)$$

The wave equations for the vibrational force and speed given in equations (10) and (11), respectively, are approximate but reflect well the physics of the phenomenon. In deriving these equation, the radial deformation of the concentrators was not taken into consideration. However, it is known that the longitudinal and radial deformations are related by the Poisson ratio μ. For metals, the value of μ is in the range of 0.25 - 0.35. The first calculation for the change in the resonant length of a cylindrical rod (constant cross-section) in the presence of radial deformations was given by Rayleigh [13]. He calculated that by taking into account radial deformations, the new resonant length $l_r = l(1-\Delta)$ where l is the resonant length in the absence or radial deformations and

$$\Delta = n^2 \mu^2 \frac{\pi}{4}\left(\frac{R}{l}\right)^2 , \qquad (31)$$

where $n = 1, 2, 3, \ldots$ In general, if the radius of a cylindrical concentrator is increased given a constant length, the value of the Rayleigh correction will increase.

To verify the theoretical calculations, we tested a cylindrical rod concentrator fabricated with a length according to equation (29). Cylindrical concentrators possess the maximal surface area for a given length and maximal radius when compared to the exponential, conical and catenoid shapes. In [12], it was show that the cavitation region is formed not only in the longitudinal direction but also around the entire lateral surface. Thus, the greater the lateral surface area, the greater the region of cavitation. Although Merkulov [14,15] stated that the catenoidal ("horn") shape has the maximum gain of acoustic energy in the longitudinal direction, as the surface area is smaller than for cylindrical cylinders the effects of cavitation may be less.

For the VTZ-1 used for our concentrator, the elasticity modulus (Young's modulus) was $E = 1.15 \times 10^{11}$ Pa, density $\rho = 4500$ kg/m^3, and Poisson ratio $\mu = 0.3$. The frequency of the ultrasonic oscillation of the generator was $f = 23{,}000 \pm 1{,}000$ Hz.

Thus, the wave number for our case was

$$k = \frac{2\pi f}{\sqrt{E/\rho}} \approx 28.57$$

and so our cylindrical concentrator required a length of

$$l = \frac{n\pi}{k} \approx 0.11m .$$

For our calculated cylindrical concentrator with n = 1 and a ratio R/l ≈ 0.2, the correction to the resonant length given by Rayleigh is $\Delta \approx 3 \times 10^{-3}$ which insignificantly changes the resonant length. The fabricated cylindrical concentrator is shown in Figure 2.

The experiments performed with the fabricated cylindrical concentrator were observed to produce cavitation bubbles when subjected to ultrasonic frequencies about 23000 Hz. If the frequency was changed by about ±500 Hz (≈3 %), cavitation bubbles ceased to be formed. The maximum production of cavitation bubbles were observed at 23277 Hz for the silica nanoparticles in water and at 22900 Hz for the copper nanoparticles in base motor oil. Thus, the experimental results support the validity of the derived equations.

UV spectra (Lumos SF-56) of the solutions produced were measured to determine how well the nanoparticles were dispersed in the media. Spectra were measured for silica nanopowders mixed into water by stirring, dispersed by ultrasonication at a frequency above the cavitation regime, and dispersed in the cavitation regime 7 days after dispersion. The results are shown in Figure 3. It was observed that nearly 100 % transmittance was obtained for the hand-mixed samples. The samples ultrasonicated without cavitation had less transmittance and the samples ultrasonicated with cavitation had the lowest transmittance. Thus, the reduced light transmittance through the sample prepared by acoustic cavitation compared to the other samples showed a higher degree of dispersion of the nanoparticles within the sample. Indeed, if observed visually, only the samples produced with acoustic cavitation had no nanoparticles sediment out of suspension. These samples were also found to be stable for several months [16]. Similar results were obtained for copper nanoparticles dispersed in base motor oil. Figure 4 shows copper nanoparticles dispersed in base motor oil by hand, with 1 min of acoustic

cavitation, and 3 min of acoustic cavitation. Increasing times of acoustic cavitation reduced the suspension light transmittance showing a higher degree of particle dispersion. Particle size analysis of suspensions showed that after being mixed by acoustic cavitation, the particles were deagglomerrated as the average size closely matched the average particle size as measured through TEM as shown in Figure 5 for silica nanoparticles dispersed in water. Thus, suspensions produced through acoustic cavitation had much higher stability and dispersion compared to the other samples and support the validity of the derived equations.

Conclusions

Equations for resonant length of rod-type ultrasonic concentrators were developed through theoretical analyses of complex-variable wave equations. The analysis was simplified due to the exclusion of the time variable ***t*** from the sinusoidal function. The introduction of the assumption that longitudinal deformations are elastic in the concentrator lead to a lack of active losses and so wave attenuation would not be observed. The theoretical conclusions were tested by fabricating a cylindrical concentrator based on the theoretical calculations and using it to disperse silica nanoparticles in water and copper nanoparticles in base motor oil. The constructed concentrator, when subjected to the resonant frequency it was designed for, successfully demonstrated acoustic cavitation while acoustic cavitation did not occur if the frequency was changed by 3%. Thus, the experimental results supported the validity of the developed equations. The derived resonant length equations should prove useful to the enhancement of various ultrasonic processes.

Acknowledgments

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Figures

Figure 1. Experimental setup for the ultrasonication of silica nanopowders in water.

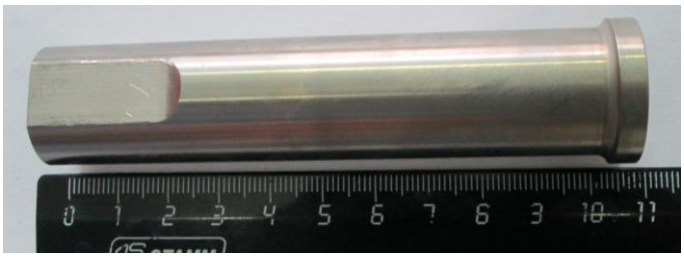

Figure 2. Experimental setup for the ultrasonication of silica nanopowders in water.

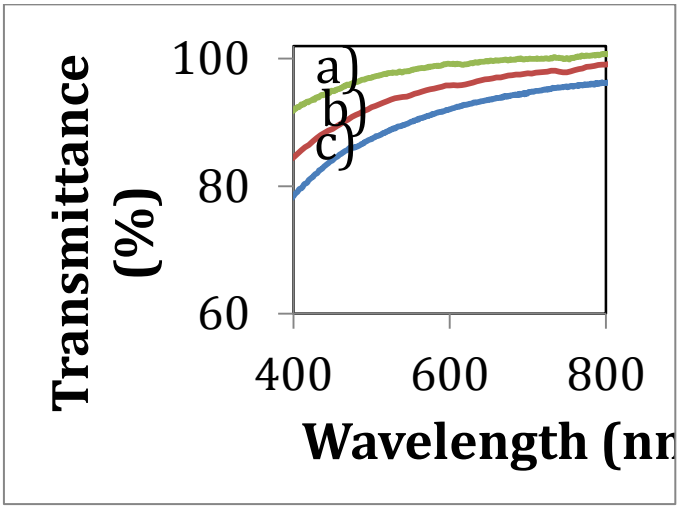

Figure 3. UV-vis spectra of silica nanoparticles dispersed in water by a) hand mixing, b) ultrasonication outside the acoustic cavitation regime, and c) ultrasonication within the acoustic cavitation regime.

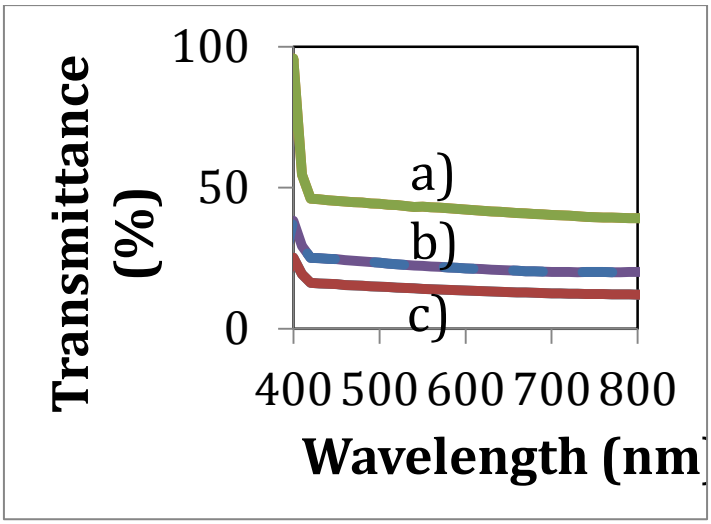

Figure 4. UV-vis spectra of copper nanoparticles dispersed in base motor oil by a) hand mixing, b) ultrasonication within the acoustic cavitation regime for 1 min, and c) ultrasonication within the acoustic cavitation regime for 3 min.

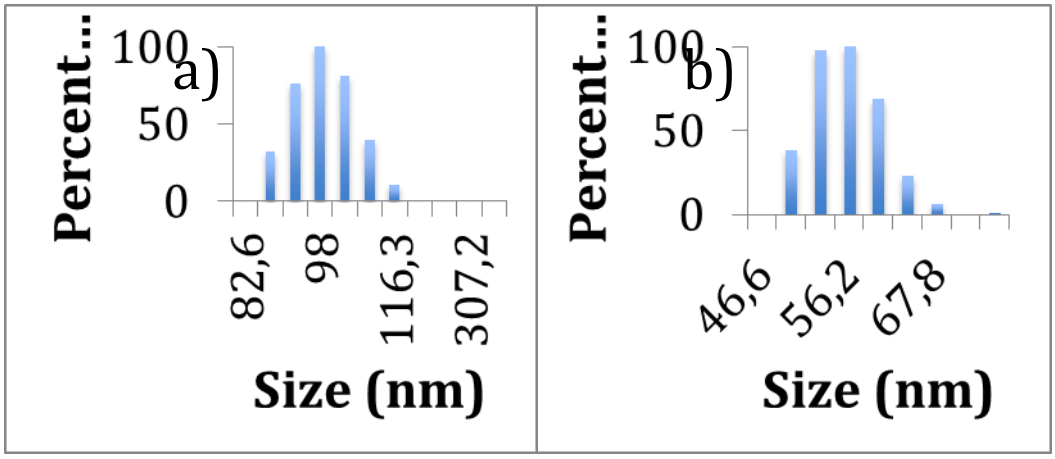

Figure 5. Particle size analysis of silica nanoparticles dispersed in water by a) hand mixing and b) ultrasonication within the acoustic cavitation regime.

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