Disaggregation of ultrafine powders in conditions of ultrasonic cavitation

Дезагрегация ультрадисперсных порошков в условиях ультразвуковой кавитации

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Abstract. Use of high-dispersive additives as structure modifiers is one of the most effective instruments of cement composites required technical exploitation characteristics increase nowadays. However, use of high-dispersive powders is complicated by the fact that their particles are consolidated into aggregates. Thus, the major advantage of high-dispersive powder, which is its possibility to form more bonds when their content is very low, appears to be unrealized. One of the promising directions of dispersed phase fine grinding is use of ultrasonic cavitation. Ultrasonic exposure does not always provide the efficiency required due to insufficient information about the effect of conditions defining ultrasonic radiation intensity on amount of aggregates damage. In this regard, it is necessary to define factors and conditions of ultrasonic exposure, which provide effective high-dispersive powders disaggregation. In article acoustic streaming in a fluid occurring due to cavitation and providing mass transfer intensification has been considered. The data about efficiency of cavitation bubble size and large-scale acoustic streaming effect on the distribution of powder particle size were obtained by calculation. The efficiency of high-dispersive powders disaggregation can be improved by using lowered hydrostatic pressure exposure.

Аннотация. Использование высокодисперсных добавок как модификаторов структуры, в настоящее время является одним из эффективных инструментов повышения требуемых технико-эксплуатационных характеристик цементных композитов. Однако использование высокодисперсных порошков сложено тем, что их частицы консолидированы в агрегаты. Тем самым не реализованным оказывается основное преимущество высокодисперсного порошка – возможность образовывать большее количество контактов при очень небольшом его содержании. Одним из перспективных путей тонкого диспергирования дисперсной фазы является использование ультразвуковой кавитации. Обработка ультразвуком не всегда обеспечивает требуемую эффективность, что связано с недостаточной информацией о влиянии условий, определяющих интенсивность ультразвукового излучения, на степень разрушения агрегатов. В связи с этим необходимым является определение факторов и условий ультразвукового воздействия, обеспечивающих эффективную дезагрегацию высокодисперсных порошков. В статье рассмотрены акустические потоки в жидкости, возникающие в результате кавитации и обеспечивающие интенсификацию массопереноса. Расчетным путем получены данные об эффективности влияния размера кавитационных пузырьков и крупномасштабных акустических течений на дисперсный состав порошка. Установлено, что эффективность дезагрегации высокодисперсных порошков можно повысить при обработке в условиях пониженного гидростатического давления.
Introduction

One of the promising directions of building material engineering development is hydration and structure formation of binding substance control by means of synthesized high-dispersive modifying agent introduction. Use of such powders allows one to change directionally the matrix structure of cement composites while their producing. Thus, the properties of cement composites can be regulated [1–5].

High-dispersive materials can be used as modifiers in building material engineering. These materials can be both naturally occurring as minerals (schungite, montmorillonite, smectites, palygorskite, chrysotile) and specially made powders of wide nomenclature nowadays [2, 6–8]. That became possible due to the instrument base development and accumulation of material synthesis knowledge.

However, there are two factors complicating the use of high-dispersive powders in the technological practice. Firstly, use of powders both synthetic and natural origin as modifiers is difficult because most of them are consolidated in sufficiently dense aggregates. Secondly, regardless of bond type, traditional disaggregation methods using mechanical impact are not effective with respect to objects of this class. If one cannot break the aggregates, the activate effect of nanocomponents introduced sharply decreases as the major advantage of high-dispersive powder, which is its possibility to form more bonds when its content is very low, appears to be unrealized.

One of the promising directions of dispersed phase fine grinding and disaggregation is use of ultrasonic cavitation. In this case, the destruction of material occurs due to the action of shock waves and liquid microjets. Although cavitation technology is widely used in industry nowadays, nevertheless, the influence of the factors mentioned above on the efficiency of destruction fine particles and aggregates is still insufficiently studied.

Purpose of work is to define factors and conditions of cavitation exposure, which provide effective high-dispersive powders disaggregation.

Methods

There are a number of proved enough conceptions of ultrasonic material dispersion mechanism, according to which the powder particle destruction occurs due to shock waves appearing in medium when cavitation bubble microexplosion can be the cause of particles and aggregates destruction [10–12]. Taking a value of pressure occurred due to cavitation bubble microexplosion equals $10^2–10^3$ MPa [12], and considering, theoretical oxides strength counted according to the Griffith’s formula is $10^4–10^5$ MPa [13] and the center of collapsing cavitation bubble located at some distance from solid particle surface, one can conclude that the stress applied to the powder particle is two or three orders lower than its theoretical strength. Thus, in spite of real material strength being considerably lower than theoretical one due to structure imperfection the value of cavitation microexplosion energy is insufficient for destruction of particles characterized by low defect rate. Thereby, the main process determining mesh size distribution change is primarily disaggregation of bound by autoadhesion forces powder particles having considerably lower strength in comparison to uniform particles.

Another factor influencing the process of particles and aggregates destruction can be acoustic streaming occurring during cavitation. Powder particles and aggregates movement and collision occurs near cavitation bubble under the influence of acoustic streaming generated in liquids at the stage of cavitation bubble micro explosion. Large-scale acoustic streaming are considered [11, 12, 14] to be the most influencing on this process as they occur in the whole volume of the medium, compared to small-scale acoustic streaming.

Eckart and Rayleigh streaming are considered to be large-scale acoustic streaming. Eckart originates in free volume of medium, where the sound wave energy absorption occurs. The velocity of such streaming is proportional to the amplitude of oscillating velocity squared and may be determined from the expression given in [14]:

$$V = \begin{cases} V_0 \left[1 - \frac{r^2}{r_1^2}\right] - \left[1 - \frac{1}{2} \frac{r_1^2}{r_0^2}\right] \left[1 - \frac{r^2}{r_0^2}\right] - \ln \frac{r_1}{r_0} & \text{when } 0 < r < r_1 \\ V_0 \left[-1 - \frac{1}{2} \frac{r_1^2}{r_0^2}\right] \left[1 - \frac{r^2}{r_0^2}\right] + \ln \frac{r}{r_0} & \text{when } r_1 < r < r_0 \end{cases}$$

(1)

where \( V_0 = \frac{Br_t^2 \rho}{2\mu}, \)

where \( \rho \) is medium density, \( r_0 \) is radius of the cylindrical tube, \( r_t \) is the radius of the acoustic beam, \( \mu \) is shear viscosity coefficient, \( B \) is constant.

A feature of Rayleigh streaming is their two-dimensionality and the independence of its velocity from medium viscosity [11, 14]. The latter can be defined by the formulas:

\[
V_x = \frac{3V_0^2}{16c} \sin 2kx \left[ 1 - \left( 1 - \frac{\eta}{\eta_1} \right)^2 \right],
\]

\[
V_y = \frac{3V_0^2}{16c} k_y \cos 2kx \left[ 1 - \left( 1 - \frac{\eta}{\eta_1} \right) \left( 1 - \frac{\eta}{\eta_1} \right)^3 \right],
\]

where \( k = \omega / c \), \( \eta = y / \delta \), \( \eta_1 = y_1 / \delta \), \( \delta \) is the thickness of the acoustic boundary layer: \( \delta = \sqrt{2V_0 / \omega} \), \( \nu = \mu / \rho \) is kinematic viscosity coefficient.

It should be noted that the greater the losses of acoustic energy in the medium, the greater the intensity and velocity of both Eckart and Rayleigh streaming. It does not matter whether the mechanism of these losses associated with the medium viscosity, a chemical reaction, relaxation or caused by the medium nonuniformity (cavitation region, gas bubbles). The irreversibility of acoustic wave energy and pulse losses is only important [10].

The velocity of the corresponding streaming determines the rate of the dispersed phase. Using the equations (1–3) and the methods proposed in [15, 16] the estimates were made for the powder particles of 0.1; 0.01; 0.001 mm sizes. The results showed that the velocity \( v \) of liquid microjets, with their radius equal in order of magnitude to the minimum radius of the cavitation bubbles, is approximate to the velocity of their collapse and reaches values of the sound velocity in the fluid (1.5–2.0)*10^3 m/s. Since the particle collision process has short duration, one can neglect the energy losses caused by plastic deformation and equate the work of destruction to the magnitude of powder particles kinetic energy, which may be determined using the formula:

\[
W_k = K \frac{mV^2}{2} C \pi R r,
\]

where \( m \) is mass of powder particles; \( v \) is velocity of the powder particles; \( C \) is the concentration; \( 4 \pi R r \) is cross section of stream.

Calculations made for the powder particles of 0.1; 0.01; 0.001 mm size showed that the magnitude of the kinetic energy was sufficient to destroy them in the process of collisions. Moreover, the stress appeared to be in the range of 10–10^9 MPa, which is comparable to the pressure occurring when the cavitation bubble collapses (10^5–10^3 MPa) [10, 15]. Therefore, one can assert that, in addition to the energy released during cavitation bubbles micro explosion, collisions factor of particles moving under the influence of acoustic streaming can also affect the disaggregation of high-dispersive powders.

Thus, the efficiency of particle and their aggregates destruction in the conditions of cavitation exposure depends on the shock wave power and velocity of acoustic streaming occurring in the medium when cavitation bubble collapsing. This efficiency is determined by the conditions of ultrasonic influence. It should be noted that the analysis of the data and the results of researches [10, 17, 18] show that, depending on the particle size of the dispersed phase and ultrasonic exposure parameters (amplitude, frequency), one of the factors mentioned above will prevail. Thus, larger particles are destroyed due to cavitation micro explosions acoustic currents. Smaller particles are destroyed due to powerful shock waves, since their pressure pulse has the greatest destructive effect only in the event when the dispersed phase particles float on the surface of the cavitation bubble. That is only possible when the dimensions of the bubble are much larger than particle size [10, 19]. In our opinion, an aggregate of particles, with the size smaller than the pores in such aggregate, can be considered as the version of the latter ultrasonic dispersing scheme. In this case, the gas bubble size is determined by a pore size. Thereby, the parameters of the ultrasonic field should provide a collapse of gas bubbles, which are located in the ~

pores of such aggregates. Thus, the problem reduces to the determination of the ultrasound exposure parameters that provide cavitation of gas bubbles having a size of $R_b < R_{agr}$.

**Results and Discussion**

Figure 1 presents data on the changes in length depending on the size distribution of the ultrasonic treatment of calcium carbonate powder, taken as a model. Ultrasound exposure contributes to the destruction of units: this is particularly intense in the initial period of exposure. Further treatment for 15 minutes leads to additional dispersion: number of fractions, consisting of large aggregates is reduced. Thus increasing the average size of less than 1 micron fraction reaches a certain size limit and begins to vary in the range of 600 to 1000 nm due to the wave nature of ultrasound. Moreover, its number tends to a limit not exceeding 15–20 %.

Having applied the well-known convection-diffusion equation of molecular physics theory, which allowed us to determine the gas concentration in any point in space at any time given we derived an equation:

$$\frac{\partial C}{\partial t} + v \frac{\partial C}{\partial r} = D \left( \frac{\partial^2 C}{\partial r^2} + \frac{2}{r} \frac{\partial C}{\partial r} \right),$$

where $C$ is gas concentration in the fluid, $D$ is diffusion coefficient, $r$ is the distance from the center to the origin of the spherical coordinate system, $v_r$ is the velocity of bubble boundaries movement.

According to Fick’s law, gas stream to a bubble per unit time is defined as:

$$\frac{dm}{dt} = 4\pi R^2 D \left( \frac{\partial C}{\partial t} \right)_{r=R},$$

In addition to relations written above, we have the equation of state

$$P_g V_g = \frac{m_g}{M} RT,$$

where $P_g$, $V_g$ are gas pressure and volume inside the bubble; $M$ is molecular mass of the gas; $B$ is the universal gas constant; $T$ is gas temperature; $m_g$ is gas mass in the bubble which can be determined from the equation:

$$m_g = \frac{4}{3} \pi R_b^3 \rho g,\quad (8)$$

The gas density inside the bubble can be determined in accordance with the following formula:

$$\rho_g = \frac{M}{RT} P_g.\quad (9)$$

Making use of differential Nolting-Nepairas equation describing the evolution of the gas-filled bubble:

$$R_b \ddot{R}_b + \frac{3}{2} \frac{R_b^2}{R_b} \ddot{R}_b + \frac{2\sigma}{\rho R_b} - \frac{1}{\rho} \left( P_0 + \frac{2\sigma}{R_0} \right) \frac{R_b^3}{R_0^3} = - \frac{1}{\rho} (P_0 - P_m \sin(w t)),\quad (10)$$

the relationship between gas pressure inside the bubble and the parameters of the surrounding fluid movement can be determined

$$P_g = \rho \left( R_b \dot{R}_b^2 + \frac{3}{2} \frac{R_b^2}{R_b} \dot{R}_b \right) + \frac{2\sigma}{R_b} - P_g + P (t),\quad (11)$$

where $\rho$ is the fluid density.

Having calculated time derivative (7) and using relations (6), (9), (11) the following differential equation can be derived:

$$D \left( \frac{\partial C}{\partial r} \right)_{r=R} = \frac{M_o}{3RT} \left( R_b^2 \dot{R}_b + 7 R_b \dot{R}_b \ddot{R}_b + \frac{9}{2} \frac{R_b}{\dot{R}_b} + \frac{3}{2} \frac{\sigma}{R_b^3} \right) + \frac{1}{\rho} \frac{d P (t)}{dt} R_b.\quad (12)$$

The last equation, equation (4) and continuity equation

$$\frac{\partial v_r}{\partial r} + \frac{2v_r}{r} = 0\quad (13)$$

form a system of differential equations. Integration of these equations allows one to define the movement of a single gas bubble boundaries, caused by the action of inertial forces and diffusion in the fluid. The calculations showed that bubbles with an initial radius of 1 to 10 microns collapse at a frequency of 18–22 kHz. The bubbles with an initial radius of more than 10 microns do not collapse but execute complex oscillations. These oscillations occur when the natural frequency of the bubble is close to the frequency of ultrasonic transducer forced oscillations. The oscillation process for bubbles with an initial radius greater than 150 microns is not cavitationally. Apparently, this is due to the fact that the initial radius of the bubbles becomes greater than resonance, which according to our calculations and according to the data

[19], for the frequency of 22 kHz is approximately 140 to 170 microns. Thus, bubbles with the initial radius of less than 10 microns are involved in cavitation. The bubbles with an initial radius greater than 10 microns execute complex oscillations and rise to the surface when reaching the resonant dimensions.

The calculations also show that the less is the hydrostatic pressure $P_0$, the less is the critical pressure $P_{cr}$. When local sound pressure exceeds the critical pressure, the bubble begins to rise sharply, the fluid collapses and cavitation occurs. Thus, the less is the pressure $P_{cr}$, the less is the critical radius $R_{cr}$, the so-called lower cavitation threshold. As a result, the range of cavitating bubbles expands radially, allowing a greater number of bubbles to collapse and thereby to intensify degassing, dispersion and destruction of the structure processes. Therefore, the hydrostatic pressure decrease (i.e. vacuum pumping) allows one to reduce the lower limit of bubble size (table 1). In this case, the occurrence of cavitation under vacuum requires ultrasound of less power [20].

<table>
<thead>
<tr>
<th>$P_0$, Pa</th>
<th>$f$, frequency, kHz</th>
</tr>
</thead>
<tbody>
<tr>
<td>1000</td>
<td>37</td>
</tr>
<tr>
<td>10000</td>
<td>68</td>
</tr>
<tr>
<td>100000</td>
<td>205</td>
</tr>
</tbody>
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Table 1. The radii of the bubbles corresponding to the resonance frequency for different values of the hydrostatic pressure and ultrasonic frequency ($R_b \times 10^6$ m)

Conclusions

The effect of ultrasonic influence conditions on the cavitation bubble size and acoustic streaming caused by cavitation were considered. The obtained results allow us to arrive at the following conclusions:

1. The efficiency of particles and their aggregates destruction during the cavitation exposure is determined by the pressure and velocity of acoustic streaming occurring in medium by the explosion of a cavitation bubble.

2. Depending on the size of aggregates, both shock wave and factor of powder particles and aggregates movement and collision influenced by acoustic streaming can be the cause of aggregates destruction. The efficiency of high-dispersive powders disaggregation can be improved by using lowered hydrostatic pressure exposure.

3. The parameters of the ultrasonic field should provide a collapse of gas bubbles, which are located in the pores of such aggregates. The bubbles with an initial radius of 1 to 10 microns collapse at a frequency of 18–22 kHz.

The bubbles with an initial radius of more than 10 microns do not collapse but execute complex oscillations. The oscillation process for bubbles with an initial radius greater than 170 microns is not cavitational.

4. The value of cavitation microexplosion energy is usually insufficient for destruction of particles characterized by low defect rate. The efficiency of high-dispersive powders disaggregation can be improved by using lowered hydrostatic pressure exposure.

5. To realize the full potential inherent in the fine powders one should make the search for new combined methods of dispersion, for example, based on the use of ultrasound in combination with surfactants.

References


Литература

2. Лотов В.А. Нанодисперсные системы в технологии строительных материалов и изделий // Известия ТПУ. 2007. Т. 311. № 3. С. 84–88.


6. Баженов Ю.М., Фаликман В.Р., Бугаков В.И. Наноматериалы и нанотехнологии в современной технологии бетонов // Вестник МГСУ. 2012. № 12. С. 125–133.


