Physical Mechanisms and Theoretical Computation of Efficiency of Submicron Particles Agglomeration by Nonlinear Acoustic Influence

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ABSTRACT

This study models the agglomeration of submicron particles when they are exposed to different types of ultrasonic waves, viz., sinusoidal waves and shock waves (pulses). The nonlinear effects of the shock waves (the transfer of heat, drop in pressure, change in the particles’ collisional cross-section due to Brownian motion, and difference in particle concentration), which influence the particle coagulation rate, are simulated for the first time and evaluated. The results reveal the optimum duration for compression and depression shock-wave pulses. Furthermore, given the same total amount of ultrasonic energy, the submicron particles coagulate 20 times more quickly with shock waves than sinusoidal waves.

Keywords: Ultrasound, Shock wave, Coagulation, Submicron particles, Smoluchowski equation

1 INTRODUCTION

Aerosols being formed in the atmosphere due to the human-induced impact, technogenic accidents, terrorist acts, and natural processes, constitute a global challenge. Mankind has been forced to continuously combat fogs, smog, dust, aerosols of harmful, toxic and radioactive substances (Oren et al., 2001; Timoshenko and Chernov, 2004; Hansen et al., 2006; Khmelev et al., 2016; Khmelev et al., 2017; Li et al., 2017; Langner et al., 2020; Liu et al., 2020).

Aerosols of finely dispersed particles (smaller than 1–2.5 µm in size) presents the greatest danger. Those particles have a high total surface area (more than 55% of the total surface area of particles emitted into the atmosphere) and number concentration (more than 95% of the total number concentration) even at a small mass fraction (less than 1% of the total portion of aerosols contained in the atmosphere) (Oren et al., 2001; Timoshenko and Chernov, 2004; Hansen et al., 2006; Khmelev et al., 2016; Khmelev et al., 2017; Li et al., 2017; Langner et al., 2020; Liu et al., 2020).

Such aerosols, owing to their small size and mass, can stay suspended in the air for a long stretch of time and can easily penetrate into air vesicles, causing irreversible changes in human body. As of today, pre-agglomeration of particles (coagulation) under exposure to sinusoidal-wave ultrasonic (US) vibrations of high intensity is one of the most efficient methods of collecting aerosol particles, moving through exhaust ducts and emitting into the atmosphere, for further collection of agglomerated particles using conventional techniques (free or gravity sedimentation, filtration through porous material, etc.).

To date, the efficiency of the US coagulation has been proved many times for particles larger than 2.5 µm. Many authors describe coagulation modes (Rozenberg, 1970; Timoshenko and Chernov, 2004; Khmelev et al., 2016; Khmelev et al., 2017), at which the highest degree of agglomeration is achieved, and propose special-purpose equipment (Crook, 1995; Mizuno, 2000; Riera et al., 2006, 2015; Iajami, 2016; Khmelev et al., 2016; Khmelev et al., 2017) which is capable of coagulating particles in both an open space and enclosed volume. At that, US sources are to be installed in processing chambers of gas purifiers, which enhance their performance without modifying their construction.
The existing equipment, however, turns out not to be sufficiently efficient in coagulating particles smaller than 2.5 µm (Langner et al., 2020), and especially smaller than 1 µm. This is due to the peculiarity of sinusoidal-wave US exposure, making submicron particles be involved in the vibratory motion within the boundaries of areas between the zones of the minimum vibration speed (Rozenberg, 1970; Hansen et al., 2006; Riera et al., 2006, 2015; Jajarmi, 2016; Khmelev et al., 2017), and almost total absence of their collision within the boundaries of those areas. Given such action, the collisional cross-sectional area of 1 µm particles is 100 times smaller than one for 20 µm particles (Muller, 1936; Protodiakonov et al., 1985; Tsibarov, 1997).

The need to address the issue requires research of the interaction process of submicron particles under different conditions of ultrasonic exposure for the purpose of exploring new non-linear effects which are capable of increasing the coagulation probability of such particles.

### 2 RESEARCH OBJECTIVE

In view of the complexity of experimental research (Shuster et al., 2002), it is appropriate to carry out a theoretical study of the coagulation process of submicron particles through creation of different conditions of US action.

When analysing acting factors of the coagulation, consideration should be given to the exposure not only in the sinusoidal-wave mode; one should consider the exposure with a changed pattern of acoustic disturbance, where the disturbance energy is not uniformly distributed in time, but it is concentrated in a narrow time domain which is short in comparison with the disturbance phase. This exposure is an exposure in the shock-wave mode. The expediency of such exposure is determined by the availability of experimental data (Goldshtein et al., 1998; Shuster et al., 2002) evidencing possible acceleration of aerosol coagulation, although obtained through the exposure in the audio-frequency range (the acoustic disturbance range is more than 1/20,000 s).

Since it is impossible to generate shock waves of compression or depression in practice in the absence of co-occurring phases of depression or compression, we should consider cases of exposure at various relations between durations of the depression phase (τd) and the compression phase following it (τc), τd/τc, including limit relations—0 (the compression wave amplitude is zero) or ±∞ (the depression wave amplitude is zero).

The ultimate objective of research consists in determining the efficient conditions of exposure, which provide the coagulation of different particles, and revealing variations in the coagulation rate with respect to the influencing factors to be determined according to the following Formula (2), which follows from mass conservation law: $\sum_{i=1}^{n} n_i D_i^3 = \text{const}$:

$$D_{30} = \left( \frac{\sum_{i=1}^{n} \langle n_i D_i^3 \rangle}{\sum_{i=1}^{n} \langle n_i \rangle} \right)$$

$$\frac{1}{D_{30}} = \frac{\partial D_{30}}{\partial t} = \frac{\partial}{\partial t} \left( \ln D_{30} \right) = \frac{\partial}{\partial t} \left( \ln \left[ \sum_{i=1}^{n} \langle n_i D_i^3 \rangle \sum_{i=1}^{n} \langle n_i \rangle \right] \right) = \frac{\partial}{\partial t} \left( \ln \text{const} \right)$$

$$= \frac{1}{3} \frac{\partial}{\partial t} \left( \ln \sum_{i=1}^{n} \langle n_i \rangle \right) = \frac{\partial}{\partial t} \left( \sum_{i=1}^{n} \langle n_i \rangle \right)$$

where $D_{30}$ is mean volumetric diameter of particles, m; $n_i$ is number concentration of particles of size $D_i$, m$^{-3}$; symbol $\langle \rangle$ means averaging of the particle concentration based on the ultrasonic disturbance length.
To research the coagulation process of submicron particles, it is appropriate to apply the probabilistic approach proposed by Smoluchowski (1916), being the most developed for today by Sheng and Shen (2006, 2007), experimentally proved (Riera et al., 2006; Sheng and Shen, 2007) and enabling the investigation of the evolution of particles given the collisional probability described by Formula (3):

\[
\frac{\partial n_i}{\partial t} + (u_i, \nabla) n_i(r,t) = \sum_{j=1}^{n} \frac{1}{2} \sum_{k=1}^{n} \beta_{i,j} n_i(r,t)n_{k-i}(r,t) - n_i(r,t) \sum_{k=1}^{n} \beta_{i,k} n_k(r,t) \tag{3}
\]

where \( n_i \) is concentration of particles with the nominal diameter of \( d_i \sqrt{\rho} \), m\(^3\); \( u_i \) is average motion velocity of particles with a diameter of \( d_i \sqrt{\rho} \); \( \beta_{i,k} \) is collisional probability of particles with diameters of \( d_i \sqrt{\rho} \) and \( d_k \sqrt{\rho} \), m\(^3\) s\(^{-1}\); \( d_0 \) is diameter of smallest particle (nominal diameter), m.

To determine the probability of pair collision of submicron particles under ultrasonic exposure, the gas phase should be considered isotropic (Tsibarov, 1997; Shuster et al., 2002), and the characteristic heat transfer time should be considered longer than the time of pressure disturbance change, i.e., the process should be considered adiabatic.

The range of chosen intensity values of the US exposure is attributed to limits related to the shock-wave drag of gases and the features of ultrasonic sources, for which the mean level of pressure in the far field is not more than 150 dB, and the same in the focus is 160 dB (Khmelev et al., 2016; Khmelev et al., 2017). Moreover, pulses are formed from 2 µs (this is the minimum pulse duration implemented in the existing devices) at the averaged intensity up to 0.06 W cm\(^{-2}\) (the root-mean-square value of ultrasonic pressure does not exceed 500 Pa, the maximum momentary disturbance is not more than 5000 Pa).

When the compression shock-wave duration is more than 2 µs, the force that must be applied to accelerate the gas flow is \( f_{\text{max}} \sim \frac{P_A}{\rho c t} \sim \frac{\rho \text{RMS}}{\rho c t} \sqrt{\frac{T}{\tau}} \) (\( P_A \) amplitude of pressure of shock wave, Pa; \( \rho \text{RMS} \) is root mean square (RMS) of pressure of shock wave; \( T \) is period between shock waves, s; \( \tau \) is shock-wave duration, s; \( \rho \) is density of gas, kg m\(^{-3}\); \( c \) is sound speed in gas, m s\(^{-1}\)), that is less than 3 kN dm\(^{-3}\).

This research of ultrasonic coagulation considers monodisperse and polydisperse aerosols with a predefined initial number concentration, the size of particles within 0.1–0.5 µm, which is usually not captured by known devices.

It is obvious that during the ultrasonic coagulation of such particles, one should take into account changes in the collisional cross-sectional area due to the motion of gas-phase molecules, as well as changes in the particle concentration with great changes in gas-phase pressure due to inertia of particles and inter-diffusion of gas molecules along the free path.

The methodology and research findings on determination of the coagulation rate of particles subject to ultrasonic exposure conditions, properties and characteristics of those dispersed particles, are presented below.

### 3 RESEARCH METHODOLOGY OF ULTRASONIC EXPOSURE EFFECTS ON THE PARTICLE COAGULATION RATE

As it follows from Formula (3), the particle coagulation rate is determined by their concentration and changes under ultrasonic exposure due to collisions, the probability of which is determined by the area of the equivalent collisional section and forces of particle interaction:

\[
\frac{1}{D_{10}} \frac{\partial D_{10}}{\partial t} = \frac{\partial}{\partial t} \left( \frac{\sum n_i}{n} \right) = \frac{\langle \beta_{i,j} n \rangle}{6} \quad ; \quad n = \frac{N}{V} \tag{4}
\]
where \( \langle \rangle \) is the symbol of averaging in a period of changes in pressure; \( V \) is selected volume of a cloud which size is small as contrasted to the exposure duration, but large as contrasted to the distance between particles, \( m^3; \) \( N \) is number of particles in the selected volume \( V; \) \( n \) is initial number concentration of particles, \( m^{-3}; \) \( \beta_{11} \) is probability of particle collision, \( m^3 \text{s}^{-1}. \)

The coagulation rate characterizes the increase in the mean volumetric diameter of particles per unit time.

The collisional probability of particles is determined using the following Formula (5) (Sheng and Shen, 2007):

\[
\beta[p](t) = \beta_{hyd}[p](t) + \beta_{ort}[p](t)
\]

where \( \beta_{hyd}[p](t) \) is constituent of hydrodynamic interaction, \( m^3 \text{s}^{-1}; \) \( \beta_{ort}[p](t) \) is constituent of orthokinetic interaction, \( m^3 \text{s}^{-1}. \)

The collisional probability constituents are determined using the formulas. The formulas are derived from models given in Sommerfeld and Blei (2002), Chernov (2004), and Sheng and Shen (2007):

\[
\beta_{hyd}[p] = \frac{1}{12\pi^2 \mu d} \int_{0}^{T} f_{11}[p](t, n) S[p](t) d\Omega(n) dt
\]

\[
S[p](t) = \pi \left( d + \sqrt{\sigma_r}[p](t) \right)^2
\]

\[
\beta_{ort}[p] = \frac{1}{T} \int_{0}^{T} \frac{\rho_l c}{\rho_p c} \left[ \frac{\rho'(t)}{\rho_l c} - \frac{\rho'(t)}{\rho_p c} \right] S[p](t) dt
\]

where \( f_{11}[p](t, n) \) is particle interaction force, \( H; \) \( S[p](t) \) is collisional cross-sectional area, \( m^2; \) \( n \) is unit vector, denoting the line direction of particle centres in relation to the wave vector of the ultrasonic field; \( \sigma_r \) is dispersion (uncertainty square) of the spatial position of particles, \( m; \) \( l \) is distance between particles, \( m; \) \( c \) is rate of propagation of ultrasonic vibrations in the carrying gas phase.

It follows from the comparison of two (3) constituents that \( \beta_{ort}[p](t) \ll \beta_{hyd}[p](t), \) i.e., the hydrodynamic interaction is determinative. The orthokinetic interaction is insignificant, since each particle, regardless of its size, is equally involved in the vibratory motion.

The probability of hydrodynamic interaction is proportional to the collisional cross-sectional area. According to the available data (Michaelides, 2015; Yang et al., 2015), the cross-sectional area of submicron particle collision changes due to the influence of the Brownian motion. Taking into consideration the Brownian motion, the collisional cross-sectional area is increased by dispersion \( \sigma_r \) of the spatial position of a particle according to Formula (7).

It is an obvious point that collisions between moving gas-phase molecules and particles (Fig. 1) cause changes in the collisional cross-sectional area according to Formula (7) and a diagram shown in Fig. 2.

Dispersion of the particle position is determined by the Monte Carlo method subject to the random generation of times of collision between a molecule and a particle, the velocities of molecules at the moment of collision and positions of a particle surface point at which the molecule collides with it.

Consideration should be given to the following assumptions when generating the above parameters:

- The particle velocity after collision of a molecule with it is determined by the law of conservation of momentum.
- Collisions are absolutely elastic.
- Collisions are equally probable in all feasible directions, since the most probable velocity of an arbitrarily chosen gas molecule is many times higher than the velocity of a particle, because the mass of an individual particle is much greater than the mass of an individual gas molecule.
Fig. 1. Diagrammatic sketch of the collisional process between gas-phase molecules and a submicron particle and the particle velocity change caused by an elementary event of collision ($v$ is random velocity of the gas-phase molecular motion; $u$ is random component of the particle motion velocity determined by heat-induced collision between gas-phase molecules and a particle; $U$ is deterministic component of the motion velocity caused by involvement of a particle in the vibratory motion of gas flow).

Fig. 2. The influence of Brownian motion on the collisional cross-sectional area: (a) collisional cross-sectional area without regard to Brownian motion; (b) collisional cross-sectional area with regard to Brownian motion.

The spatial position of a particle as a result of a sequence of collisional elementary events “particle-molecule of the gas phase” is determined according to the following formula:

$$ r_{i+1} = r_i + u_i (t_{i+1} - t_i) $$

(9)

where $t_i$ is occurrence time point of the $i^{th}$ elementary event of collision between a molecule and a particle, $s$; $u_i$ is particle velocity as a result of the $i^{th}$ elementary event of collision with a molecule, m s$^{-1}$; $r_i$ is coordinate vector of particle at the occurrence time point of the $i^{th}$ elementary event of collision, m.

The particle velocity as a result of the $i^{th}$ elementary event of collision is determined according to the law of conservation of momentum:

- $v - u \rightarrow v - u + w$: change in the velocity of a collided molecule in the centre-of-momentum frame of a particle;
\[ v \rightarrow v + wn: \text{change in the velocity of a collided molecule in the laboratory frame of reference;} \]

\[ m(v + wn) + M_{\text{new}} = mv + Mu: \text{law of conservation of momentum for the system “particle-collided molecule”;} \]

\[ u_{\text{new}} = u - \frac{m}{M} wn: \text{common expression for the changed velocity of a particle as a result of the} \]

\[ \text{ith} \text{elementary event of collision.} \]

An unknown variable \( w \) is determined on the basis of the energy conservation law:

\[ M\left(\frac{u - \frac{m}{M} wn}{2}\right)^2 + \frac{m(v + wn)^2}{2} = \frac{Mu^2}{2} + \frac{mv^2}{2}; \quad u_{i+1} = u + \frac{2m}{M} (v_i - u_i, n_i) n_i \]

(10)

where \( M \) is particle mass, kg; \( m \) is gas-phase molecule mass, kg.

When generating times, velocities of collision between a molecule and a particle, positions of a surface point at which a collision occurs, it is supposed that the velocity distribution function at an arbitrary choice of a molecule follows Maxwell’s law.

\[ f(u, n, v_n) dv_n = \frac{\rho A}{\sqrt{2\pi} p} \exp\left(-\frac{1}{2 \pi} \frac{u^2}{\rho A}ight) \]

(11)

Velocities, times, and positions of a surface point, at which a collision occurs, are generated until they are equal to the distribution function of positions of an individual particle as a result of the next and previous collision events:

\[ F_{i+1}(r) = F_i(r) = F(r) \]

(12)

Preliminary calculations showed that the time sufficient to meet the condition (12) is much less than the characteristic time of pressure changing. This gives us grounds for considering pressure to be constant in determining an instantaneous effective collisional cross-section of particles.

An approach set forth herein made it possible to establish relations between the effective collisional cross-section and the vibration amplitude for various ultrasonic effects:

1. Continuous sinusoidal wave:

\[ p'(t) = p_A \sin\left(2\pi \frac{t}{T}\right) \]

(13)

2. Shock wave (pulse):

\[ p'(t) = p_A e^{-\frac{|t|}{\tau}} \]

(14)

which can be conveniently classified into compression pulses at \( p_A > 0 \) and depression pulses at \( p_A < 0 \).

\[ \left\lfloor \frac{t}{T}\right\rfloor \text{ means whole part of below of } \frac{t}{T}. \]

If influence contains both compression and depression pulses, then

\[ p'(t) = p_{A,\text{comp}} e^{-\frac{\left|\frac{t}{T}\right|}{\tau_{\text{comp}}}} - p_{A,\text{depr}} e^{-\frac{\left|\frac{t}{T}\right|}{\tau_{\text{depr}}}} \]

(15)

where \( T \) is acoustic disturbance time period, s; \( p_A \) is amplitude of US disturbances, Pa; \( \tau \) is
characteristic duration of a pulse, \( s \); \( \tau_{\text{depr}} \) is characteristic duration of depression pulse, \( s \); \( \tau_{\text{compr}} \) is characteristic duration of compression pulse, \( s \); \( p_{A_{\text{compr}}} \) is amplitude of compression pulse, Pa; \( p_{A_{\text{depr}}} \) is amplitude of depression pulse, Pa.

The forms of each type of shock wave are shown in Fig. 3.

In particular, the effective collisional cross-section for continuous sinusoidal-wave exposure \( \rho'(t) = p_A \sin(\frac{2\pi t}{T}) \) does not change very much (less than 10% increase). For shock-wave pulse action \( \rho'(t) = p_A e^{-t/\tau_{\text{compr}}} \), while forming a predominant compression phase \( p_A > 0 \) (at \( \tau_{\text{compr}} = -p_{A_{\text{depr}}} e^{-t/\tau_{\text{compr}}} \), \( p_{A_{\text{compr}}} > 0 \); \( p_{A_{\text{depr}}} = 0 \)), any change in the collisional area even for the finest particles (100–200 µm) does not exceed 35–40%, and only when forming a predominant depression phase on ultrasonic exposure \( p_A < 0 \), at \( \rho'(t) = p_{A_{\text{compr}}} e^{t/\tau_{\text{compr}} - p_{A_{\text{depr}}} e^{-t/\tau_{\text{compr}}}} \), \( p_{A_{\text{compr}}} = 0 \); \( p_{A_{\text{depr}}} > 0 \) the collisional area demonstrates 2 times’ increase at the energies provided by the modern emitters (Riera et al., 2006, 2015; Khmelev et al., 2017).

The resulting data on the collisional area value allowed proceeding with further research of the hydrodynamic interaction process which, according to Rozenberg (1970), is determined by radiation pressure of the US effect reflected from a neighbouring particle:

\[
f_{21} = \int (-p + \rho u(u,n)) dS
\]

where \( f_{21} \) is interparticle force, N; \( p \) is pressure disturbance of gas near a particle created by a

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Fig. 3. Shape of shock wave: (a) compression; (b) depression; (c) compression–depression interleaving.
neighbouring particle, \( P_a \); \( u \) is gas velocity perturbation near a particle created by a neighbouring particle, \( m \); \( n \) is normal vector to the particle surface.

Since the particles in question are submicronic, and the distance between them is consistent with the free length of gas-phase molecules, the following quasi-gas dynamic equations, considering the inter-diffusion of the gas phase and the non-equilibrium state of the operation of pressure change process in the gas phase, were used to calculate the field of reflected ultrasonic effects (Elizarova, 2005):

\[
\begin{align*}
\text{div}(j_m \otimes u) + \nabla p &= \text{div}\Pi \quad (17) \\
\text{div}(j_m) &= 0 \quad (18) \\
j_m &= \rho u - \tau (\text{div}(\rho u \otimes u) + \nabla p) \quad (19)
\end{align*}
\]

\[
\Pi = ru \otimes [\rho(u, V)u + \nabla p] + \tau f[(u, V)p + \gamma p \text{div } u] \quad (20)
\]

where \( j_m \) is gas-phase mass flux, \( \text{kg} \text{m}^{-2} \text{s}^{-1} \); \( u \) is gas-phase molecule motion velocity, \( \text{m s}^{-1} \); \( \rho \) is gas-phase density, \( \text{kg} \text{m}^{-3} \); \( \tau \) is relaxation time required for gas phase to move to the equilibrium state, \( s \); \( \Pi \) is stress tensor in gas phase, \( \text{Pa} \).

Solving the presented set of equations made it possible to determine the fields of pressure and gas velocity perturbations around an individual particle for a given ultrasonic exposure (sinusoidal one or one with predominant compression or depression phases) and calculate the radiation pressure force on a neighbouring particle from the part of the found disturbance fields.

Since the propagation process of ultrasonic exposure is adiabatic, the set of Formulas (17)–(20) is rearranged:

\[
\begin{align*}
\text{div}(j_m \otimes u) + \nabla p &= \text{div}\Pi \quad (21) \\
\text{div}(j_m) &= 0 \quad (22) \\
j_m &= \rho_{rel} \left( \frac{\rho}{\rho_{rel}} \right)^{\frac{1}{2}} \nabla \phi - \frac{\tau}{\tau_{rel}} \left( \text{div} \left( \frac{\rho}{\rho_{rel}} \right) \left( \frac{1}{2} \right) \nabla \phi \otimes \nabla \phi + \nabla p \right) \quad (23)
\end{align*}
\]

\[
\Pi = ru \otimes \left[ \rho_{rel} \left( \frac{\rho}{\rho_{rel}} \right)^{\frac{1}{2}} (u, V)u + \nabla p \right] + \tau f[(u, V)p + \gamma p \text{div } u] \quad (24)
\]

where \( \phi \) is velocity potential, \( \text{m}^2 \text{s}^{-1} \).

The system is further supplemented with the boundary condition on the particle surface (26), from which the reflection occurs, and with conditions (26, 27) at a distance exceeding the distance between neighbouring particles:

\[
\begin{align*}
u &= 0 \quad (25) \\
p &= \rho_{rel} + P_\Lambda(r, t) \quad (26) \\
u &= \frac{k P_\Lambda(r, t)}{k \rho c} \quad (27)
\end{align*}
\]

where \( \rho_{rel} \) is static pressure in gas with no ultrasonic disturbances, \( \text{Pa} \); \( P_\Lambda \) is pressure of ultrasonic
exposure, Pa; \( k \) is wave vector of field, m\(^{-1} \).

The distribution of pressure disturbances near a ball-shaped particle consists of three components:

\[
p = p_{\text{rel}} + P_{A}(r, t) + p_{\text{refl}}(r, t)
\]  

(28)

where \( p_{\text{refl}} \) is pressure in ultrasonic exposures reflected from a particle, Pa.

Since particle sizes are massively smaller than the ultrasonic-exposure-area length, radiation pressure on the surface of a neighbouring particle will be contributed by pressure \( p_{\text{refl}} \) because the other components are much less dependent on coordinates:

\[
\langle p_{\text{rel}} + P_{A}(r, t) - p_{\text{refl}}(r, t) \rangle
\]

(28)

where \( p_{\text{refl}} \) is pressure in ultrasonic exposures reflected from a particle, Pa.

Since particle sizes are massively smaller than the ultrasonic-exposure-area length, radiation pressure on the surface of a neighbouring particle will be contributed by pressure \( p_{\text{refl}} \) because the other components are much less dependent on coordinates:

\[
\langle p_{\text{rel}} + P_{A}(r, t) - p_{\text{refl}}(r, t) \rangle
\]

(28)

with pulse amplitude more than 5000 Pa and particle diameter less than 1 \( \mu \)m; where \( r_1 \) and \( r_2 \) are diametrically opposite points on particle; \( P_{A_{\text{max}}} \) is pressure amplitude of shock wave, Pa; \( d \) is particle diameter, m; \( k \) is wave number of shock wave; \( \tau \) is pulse duration of shock wave, s; \( R \) is particle radius; \( U_{\text{max}} \) is maximum gaseous speed in shock wave, m s\(^{-1} \); \( P_{\text{RMS}} \) is root-mean-square pressure of shock wave, Pa), and the integral of components \( P_{A} \) and \( p_{\text{rel}} \) is equal to zero.

In real gas-dispersion systems the interparticle distance is more than the free length of gas molecules (70 nm), and determination of fields of velocity and pressure disturbances is based on the asymptotic decomposition by gas relaxation time degrees \( \tau \) (time required to move to the equilibrium state and take microscopic parameters of a molecule assembly corresponding to gas parameters) and the amplitude of ultrasonic exposure:

\[
u = \sum_{n=0}^{\infty} u_{n,m} \tau_{\text{rel}}^{-n} = \sum_{n=0}^{\infty} u_{n,m} \tau_{\text{rel}}^{-n} U_{\text{c}}^{-m} \]

(29)

\[
p = \sum_{n=0}^{\infty} p_{n,m} \tau_{\text{rel}}^{-n} = \sum_{n=0}^{\infty} p_{n,m} \tau_{\text{rel}}^{-n} U_{\text{c}}^{-m} \]

(30)

\[
\rho = \sum_{n=0}^{\infty} \rho_{n,m} \tau_{\text{rel}}^{-n} = \rho_{\text{rel}} \left( 1 + \left( \sum_{n=0, m=0}^{\infty} \frac{\rho_{n,m} \tau_{\text{rel}}^{-n} U_{\text{c}}^{-m}}{\rho_{\text{rel}}} \right)^{\frac{1}{2}} \right) \]

(31)

By reference to the presented asymptotic decompositions, the set of mass and pulse conservation equations are decomposed into several equations based on relaxation time degrees:

\[
div (\rho_{\text{rel}} u_{0} \otimes u_{0}) + \nabla p_{0} = div \Pi_{0} ;
\]

(32)

\[
div (\rho_{\text{rel}} u_{0}) = 0
\]

(33)

\[
\left\{ div (\rho_{\text{rel}} u_{0} \otimes u_{1}) + div \left( \rho_{\text{rel}} u_{1} + \rho_{\text{rel}} u_{0} + \tau_{\text{rel}} \frac{\partial (\rho_{\text{rel}} u_{0})}{\partial t} \right) \otimes u_{0} \right\} + \nabla p_{1} = div \Pi_{1}
\]

(34)
and time and velocity degrees:

\[
\nabla p_{0,0} = 0, \\
\mathbf{u}_{0,0} = 0 ;
\]

\[
\rho_{0.1}(\mathbf{u}_{0,1,\mathbf{V}})u_{0,1} + \nabla p_{0.1} = 0
\]

\[
\text{div} (\mathbf{u}_{0,1}) = -\text{div} \left( \frac{\rho_{0.2}}{\rho_{0.1}} \mathbf{u}_{0,1} \right) ;
\]

\[
\nabla p_{0,1} = 0, \\
\text{div}(\mathbf{u}_{0,1}) = 0 ;
\]

\[
\rho_{0.2}(\mathbf{u}_{0,1,\mathbf{V}})u_{0,1} + \nabla p_{0.2} = 0 ;
\]

\[
\text{div}(\mathbf{u}_{0,2}) = 0
\]

\[
\nabla p_{1,0} = 0, \\
\text{div} (\mathbf{u}_{1,0}) = 0 ;
\]

\[
\rho_{1,0}(\mathbf{u}_{1,0,\mathbf{V}})u_{1,0} + \nabla p_{1,0} = 0
\]

\[
\text{div}(\mathbf{u}_{1,0}) = 0
\]

\[
\nabla p_{1,1} = 0, \\
\text{div} (\mathbf{u}_{1,1}) = 0 ;
\]

\[
\text{div}(\mathbf{u}_{1,1}) + \nabla p_{2,1} = 0
\]

\[
\rho_{2,0}(\mathbf{u}_{0,1,\mathbf{V}})u_{0,1} + \nabla p_{2,0} = 0
\]

\[
\text{div} (\mathbf{u}_{0,2}) = 0 ;
\]

\[
\text{div}(\mathbf{u}_{1,2}) = 0
\]

(Formulas (35)–(41) were solved analytically. General scheme of analytical solution is following:

1. Each velocity component \(\mathbf{u}_{ij}\) is potential, i.e., \(\mathbf{u}_{ij} = \nabla \phi_{ij}\), where \(\phi_{ij}\) is velocity potential, \(m^2 s^{-2}\).
2. Velocity component \(\mathbf{u}_{0,0} = 0\), density component \(\rho_{0,0} = \text{const}\).
3. For each component of each physical magnitude \(\mathbf{u}_{ij}, \rho_{ij}, \phi_{ij}\) pair of equations reduces to view:

\[
\begin{align*}
\nabla \phi_{ij} &= A_{ij} \left[ u_{ij,\phi_{ij,\phi_{ij}}} \rho_{ij,\phi_{ij,\phi_{ij}}} \right], \\
\text{div}(\mathbf{u}_{ij}) &= \frac{1}{\rho_{ij}} B_{ij} \left[ u_{ij,\phi_{ij,\phi_{ij}}} \rho_{ij,\phi_{ij,\phi_{ij}}} \right],
\end{align*}
\]

where indexes \((0,0) \leq (i_j, i_j, \phi_{ij, \phi_{ij}}) < (i_j, \phi_{ij, \phi_{ij}})\). \(A_{ij}\) and \(B_{ij}\) are operators which represent polynomial combination of \(\rho_{ij,\phi_{ij,\phi_{ij}}}(\mathbf{u}_{ij})\) for \(\phi_{ij,\phi_{ij}}\).

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\(A_{ij}\) and \(B_{ij}\) are operators which represent polynomial combination of \(\rho_{ij,\phi_{ij,\phi_{ij}}}(\mathbf{u}_{ij})\) for \(\phi_{ij,\phi_{ij}}\).

4. Definition of \(l_{\text{max}}\), which is maximum value of \(l_i\), \(j_{\text{max}}\), which is maximum value of \(j\).

5. Analytical calculating of \(\mathbf{u}_{0,1}, \phi_{0,1}, \rho_{0,1}\). The calculation is based on decomposition of \(\phi_{ij}\) into Legendre’s polynomials.

6. By mathematical induction, the components \(\mathbf{u}_{ij}, \rho_{ij}, \phi_{ij}\) are calculated using decomposition into Legendre’s polynomials through \(\mathbf{u}_{ij}, \rho_{ij}, \phi_{ij}\) at all \((k, l) < (i, j)\) using (42).

7. Step (6) is performed for all \((i, j) \leq (l_{\text{max}}, l_{\text{max}})\) until \((i, j) = (l_{\text{max}}, l_{\text{max}})\).
of acoustic disturbances:

$$\frac{1}{D_{30}} \frac{\partial D_{30}}{\partial t} = \frac{1}{7} \int_0^\infty \left[ \frac{f_1 \rho(t, \mathbf{n})}{12\pi^2 n d} S \rho(t) d\Omega(n) \right] \frac{s}{6} \int_s \left( -p_{in} \mathbf{N} + \mathbf{u}(\mathbf{N}) \right) dS; \quad f_1 \rho(t, \mathbf{n}) = \int_s \left( -p_{in} \mathbf{N} + \mathbf{u}(\mathbf{N}) \right) dS; \quad (43)$$

where $\mu$ is viscosity of gaseous phase, $m$; $d$ is diameter of separate particle, $m$; $n$ is particle concentration, $m^{-3}$; $f_1$ is particles’ hydrodynamic interaction force, $N$; $S$ is square of equivalent collisional cross-section, $m^2$; $\Omega$ is solid angle of particles’ centre line, $sr$; $n$ is unit normal vector of centre line orientation; $\mathbf{N}$ is normal vector to particle surface.

The physical meaning of last expression is sum by neighbour particles interacting with observed particle from different angles between centre line and wave vector of acoustic field. The hydrodynamic force depends on both acoustic pressure and angle. However, collisional cross-section depends on acoustic pressure only.

The resulting dependences of the coagulation rate are set forth in the next section.

### 4 RESEARCH FINDINGS OF PARTICLE COAGULATION PROCESS UNDER

#### 4.1 Ultrasonic Exposure

Fig. 4 presents the determination results of the coagulation rate. As it follows from the presented dependences, any ultrasonic exposure in the form of pulses (both with a predominant compression phase and a predominant phase of depression) accelerated the coagulation rate as compared with sinusoidal-wave ultrasonic exposure. At that, the coagulation rate increases as the length of compression or depression phases reduces, even when the root-mean-square value of sound pressure is retained (total energy input).

This is attributable to non-linear effects (convection transfer, change of the collisional cross-sectional area under conditions of the Brownian motion), arising from a sharp change in the instantaneous value of sound pressure.

In those and subsequent relations, the value of $T$ (time of acoustic disturbances) is equal to 1/22,000 s. The model aerosol number concentration is $2 \cdot 10^{11}$ g m$^{-3}$. The model frequency of sinusoidal vibrations is 22,000 Hz. The model shock-waves’ cycle period is 1/220 s, and shock-waves’ duration is in range from 0.1T to $T$.

The obtained data shows the coagulation efficiency, using the pulsed ultrasonic exposure while maintaining the magnitude of the input energy. It has been established that a shock-wave exposure with the predominant compression phase increases 5 times the coagulation rate for submicron particles with a range of 100–500 nm in size only due to a change in the ultrasonic exposure conditions. Exposure with the predominant depression phase due to an additional increase in the collisional cross-sectional area (the uncertainty of the particle position changes) increases 8 times the coagulation rate of particles of 500 nm in size, and the particles of 100 nm in size—20 times compared with sinusoidal-wave exposure. It is explained by the following. The probability of collision of particles is proportional to hydrodynamic force and collisional cross-section. The predominant depression shock wave has negative sign of pressure and compression shock wave has positive sign of pressure. Less pressure, larger collisional cross-section (due to Brownian motion). Larger collisional cross-section follows more coagulation efficiency.

All of that confirms the highest efficiency of shock-wave action for small-size particles.

In view of the fact that it is impossible to carry out any pulse exposure with the compression (or depression) phase only, it is to be supposed that the maximum effect of coagulation acceleration (10 times and more) will be provided in practice on successive exposures to vibrations in which the duration of the depression phase is less than the compression phase duration.

Relationships between the agglomeration rate and the ratio of the depression phase duration ($t_d$) to the depression phase duration ($t_c$), $t_d/t_c$, are presented in Fig. 5.

As it follows from the presented dependences, the maximum coagulation rate is achieved
Fig. 4. Coagulation rate–RMS pressure value dependences at various particle sizes and exposure conditions: (a) 100 nm; (b) 300 nm; (c) 500 nm.

Fig. 5. Relationship between the coagulation rate and the ratio of the depression-phase duration to the compression-phase duration \( \tau_d/\tau_c \): (a) root-mean-square value of pressure 200 Pa; (b) root-mean-square value of pressure 500 Pa.
when the compression phase duration in ultrasonic exposure dominates over the depression phase duration (the amplitude of the depression phase is higher than that of the compression phase).

Then the influence of acoustic disturbance on the particle size distribution in polydisperse aerosol was established in terms of the determined value of the coagulation rate. The Smoluchowski equation, upon substitution of the expressions obtained for the collisional probability allowed plotting histograms of the initial aerosol and aerosols after various ultrasonic exposures (Fig. 6).

The histograms were obtained by solution of Smoluchowski’s equation. For the solutions of spatial Smoluchowski equation (called population balance equation) today the fixed pivot method and CFD (computational fluid dynamics)-sectional algorithm are widely used (Shang et al., 2020a, b). The methods have following advantages:

• taking into account spatial inhomogeneity of particles’ concentrations;
• taking into account particles’ ensemble flow.

However, the methods have high computational complexity. Moreover, the methods do not take into account difference between velocities of same-size particles. Herewith, the main goal of the paper is research of shock-wave influence on coagulation in spatial homogeneous case (convective member is zero). Thus, the using of CFD-PBM (population balance model) algorithms is impractical.

Thus, the histograms were obtained by solution of Smoluchowski’s equation for spatial homogeneous case (particle concentrations do not depend on time) with using Runge-Kutta method for solution ODE (44):

\[
\frac{\partial n_k(r,t)}{\partial t} = \frac{1}{2} \sum_{i=1}^{k+1} \beta_{k,i} n_i(r,t)n_k(r,t) - n_k(r,t) \sum_{j=1}^{N} \beta_{j,k} n_j(r,t)
\]  

(44)

Fig. 6. Changed size distribution of particles with the initial modal size of 100 nm at different root-mean-square values of pressure and conditions of ultrasonic exposure: (a) 200 Pa; (b) 500 Pa.
\[
\beta_{i,j} = \frac{1}{T} \int_0^T \frac{f_2(t,n)}{12\pi^2 \mu} \left( \frac{1}{d_i} + \frac{1}{d_j} \right) S_j(t) \, d\Omega(n)
\]

(45)

where \( S_j \) is \( i^{\text{th}} \) and \( j^{\text{th}} \) particles’ collisional cross-sectional square, \( m^2 \); \( d_i \) is diameter of \( i^{\text{th}} \) particle. \( f_2 \) and \( S_j \) are calculated by proposed model described in previous section.

Formula (44) was solved by method of Runge-Kutta.

In both modelling cases the exposure time was 10 minutes; the initial concentration (total of all particle nominal sizes) was \( 2 \times 10^{11} \text{ g m}^{-3} \); the action frequency was 22 kHz; the ratio of the depression phase duration to the compression phase duration for the shock-wave (pulse) action is 0.2.

The initial particle size distribution of aerosol (the number concentration of particles of each size) followed the Rayleigh law.

It follows from the presented histograms that the coagulation efficiency is improved with increase in the particle diameter. Moreover, addition to the mass fraction caused by shock-wave exposure is essential for particles with the initial modal size of 100 nm.

The shown histograms support the improved efficiency of special conditions of exposure as compared to the sinusoidal-wave exposure even for polydisperse aerosols. Moreover, more than 5 times’ increase (for the modal size of initial particles—100 nm, RMS—500 Pa) in the quantitative portion of coarse fraction (particles whose diameter exceeds the modal diameter by more than 30%) is found while the coarse fraction portion shows no more than 3 times’ increase under the sinusoidal-wave ultrasonic exposure.

Time dependences of the mean volumetric surface diameter of aerosol particles at various root-mean-square values of sound pressure and various conditions of ultrasonic exposure were constructed based on the resulted histograms (Fig. 7).

The presented dependences attest to a reduction of time required to achieve the maximum

**Fig. 7.** Sauter mean diameter \((d_{32})\)–time dependences at various mean-square values of pressure; conditions of ultrasonic exposure and initial modal diameters of particles: (a) 100 nm; (b) 300 nm; (c) 500 nm.
size of particles when applying shock-wave exposure. For example, for particles of 100 nm, at sinusoidal-wave exposure, the maximum diameter is not achieved even in 150 minutes, while in shock-wave exposure a diameter being 95% of the maximum diameter is achieved in 60 minutes.

5 CONCLUSIONS

This study confirms the enhancing effect of sinusoidal waves and shock waves on the coagulation of submicron particles in gas media and establishes conditions for optimising agglomeration. Furthermore, compared to sinusoidal waves, shock waves accelerate coagulation by as much as 20 times.

The coagulation rate can be increased via shock-wave exposure by improving either the wave propagation (e.g., adjusting the reflection phase to radiate waves between sources and reflectors, forming sequences of pulses that vary in frequency and dispersion velocity in the gas media, or focusing on or interfering with oscillations generated by multi-frequency, spatially distributed emitters) or the gas flow (e.g., using interrupters, adjusting the density, or changing the direction).

These results provide a basis for developing a new class of high-tech submicron particle collectors that addresses environmental and health issues.

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