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12 page article in English entitled, "Precipitation of Zinc Oxide Aerosol in Sound Field, by D.B.Dianov, L.G.Merkulov, and V.I.Nikitenko of the Leningrad Electrotechnical Institute immini V.I.Lenin (UI'yanov), which appeared in the Akusticheskii Zhurnal, Vol. VIII, No. 1, 1962, pages: 60-66, a paper submitted to the Eighth ALL-Union Conference for the Uses of Ultrasound in Testing of Materials, February 1960.

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2. This important paper deals with the precipitation of an aerosol of zinc oxide particles in an adjustable resonant volume (1.15 X 0.55 X 0.75 X 0.40 m) under static conditions. With a view to cover a wide frequency range (0.2 to 21 keps) at constant irradiated power, the authors use three different types of sound sources: Hartmann Whistle, Acoustic Siren Type UZG-4-A and an Electrodynamic Generator. The sound sources irradiated inside the processing chamber through a membrane.

3. Antinode sound pressures up to 4.10^4 bar (166db) were reached under resonant conditions. The zinc oxide particles size distribution peaked around 3.6 microns. Initial aerosol concentrations averaged 5-7 grs/m^3. The coagulation rate was followed by means of a nephelometer and a technical discussion of this device is presented. Precipitation time without sound ranges from 40 to 50 minutes. With sound for all frequencies tested, the time is reduced to approximately 35 seconds.

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4. The important conclusion of this work was that there is little difference in precipitation time over the whole frequency range investigated. This is [redacted] the first significant work undertaken at constant irradiated power for aerosol coagulation studies. From the scientific view point, it means that hydrodynamic forces of the Oseen type predominate, in the low frequency range, over orthokinetic mechanisms. This had already been forcasted but not proven by Pshenai-Severin in 1959.

5. From the practical view point this work can be considered as a breakthrough since low frequency sound sources would be far more economical to operate than any high frequency industrial siren. It opens the way to large scale aerosol agglomeration processes in industry.

- End -

PRECIPITATION OF ZINC OXIDE AEROSOL IN SOUND FIELD*

by D. B. Dianov, L. G. Merkulov, and V. I. Nikitenko
Leningrad Electrotechnical Institute V. I. Lenin (Ul'yanov)

Akusticheskii zhurnal, Vol. VIII, No. 1, 1962, pages: 60-66

The results of the investigation of sound precipitation of polydispersed zinc oxide aerosol under static conditions within the 200-21,000 cycles frequency limit have been presented.

The dependence of the precipitation time from the magnitude of the sound pressure inside the sound chamber has been shown. The precipitation time has been found not to be significantly influenced by the frequency of the sound.

It is well known that strong sound and ultrasound oscillations accelerate the coagulation process of aerosol particles and may substantially increase the efficiency of precipitation. In spite of the considerable number of papers devoted to the investigation of the accelerating influence of sound oscillations on the coagulation and precipitation of aerosols, there are still not available at the present time reliable quantitative data about the dependence of these processes on various parameters (sound frequency, sound pressure amplitude, particle concentration, type of aerosol, etc.). A clarification of these relationships is of considerable practical interest since knowing all the relevant facts one could approach efficiently the problem of designing industrial devices for sound precipitation of aerosols.

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*Paper submitted to the Eighth All-Union Conference for the Uses of Ultrasound in Testing of Materials, February 1960.

We investigated the role of the frequency and sound pressure amplitude on the sound precipitation of zinc oxide aerosols (zinc white M-1). Tests using ordinary and electron microscope showed that zinc oxides are polydispersed aerosols consisting primarily of aggregates of primary particles. Linear dimensions (r) of particles immediately after the formation of aerosol in the chamber are logarithmically sufficiently approximated by the normal distribution [1]

$$f_0(r)\,dr = \frac{1}{\lg\beta\sqrt{2\pi}}\exp\left[-\frac{(\lg r-\lg r_m)^2}{2(\lg\beta)^2}\right]d\lg r \qquad (1)$$

with $\lg r_m = 0.48$ ($r_m = 3\mu$), $\lg\beta = 0.18$. Fig. 1 presents the curve $r f_0(r)$ as function of $\lg r$.

Experimental investigations of the precipitation under the influence of sound were performed using the laboratory apparatus indicated on Fig. 2 under static conditions. Zinc oxide was introduced into the upper part of the chamber 1 using a special atomizer. The working chamber constituted a resonant volume which could be varied by means of the movable rear wall ($l_{1\,max}$ = 1,150 mm, l_2 = 550 mm, l_3 = 750 mm, h = 400 mm). The walls were made of organic glass permitting a visual observation of processes inside the chamber.

We used for the sound source 3 a gas-flow generator (type Hartman) with a selection of resonators and jets designed for operation at frequencies between 5 and 21 kc, an acoustic siren UZG-4a, and for the low-frequency region a source of electrodynamic design. To eliminate the influence of inner air currents on the precipitation process of aerosols, the working volume of the chamber was separated from the sound source by a thin sound transmitting partition 4. The acoustical matching of the whole system was achieved by choosing the separation between the front wall of the chamber and the exit opening of the source. Because of such tuning we achieved effective magnitudes of antinode sound pressures up to 4.10^4 bar (166 db). The pressure ratio between the nodes and antinodes was about 10 on the average (at a 5 kc frequency). With an additional damping of the chamber wall it would decrease to some 3-4.

The compressor (ZIF-55) 5 provided compressed air for the gas-flow generator and siren. Pressure and air consumption were measured by means of a manometer 6 and a flowmeter 7 respectively.

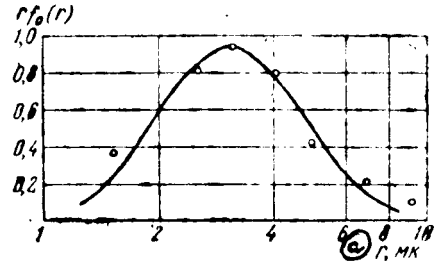

Fig. 1

a - r,

Preliminary acoustical measurements in the damped chamber supplied optimum operating conditions for the sound sources used and their acoustic-mechanical efficiency.

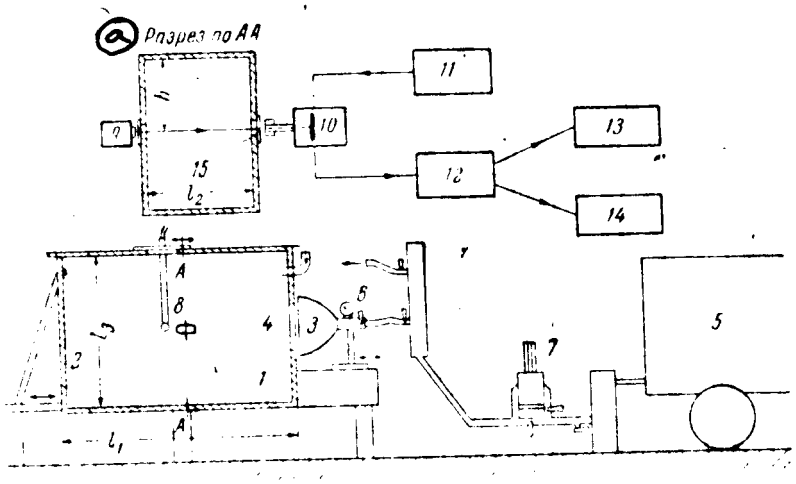

Fig. 2

a - Cross-section across AA.

During the actual tests the amplitude of the sound pressure and the frequency were checked by means of a spherical barium-titanate microphone* 8. The microphone output was amplified and then fed to a voltmeter, frequencymeter, and oscilloscope. Nonlinear distortions were estimated from the shape of the oscilloscope curve. The microphone could be moved along the vertical as well as along horizontal planes, and we were able in this manner to check the distribution of the sound field inside the chamber.

Nephelometer apparatus measured the coagulation speed in the precipitation of the aerosol. A light beam from the light source 9 traveled through a system of condenser lenses and landed on the photomultiplier 10, connected to a highly stabilized power supply 11. The output signal of the photomultiplier proceeded to the compensating circuit 12 and then to the stretched wire oscillograph 13 and microe- Using the readings of these instruments one could follow the variations of the light beam intensity and, consequently, the variations in concentration of the aerosol inside the chamber. To eliminate the influences of wall dust deposits on readings of the nephelometer the chamber had opening along the light beam path covered with detachable optical glass covers 15. The glasses were changed after every test and their dust contamination, which was much lower than that of the walls, was taken into account during the final evaluation of results.

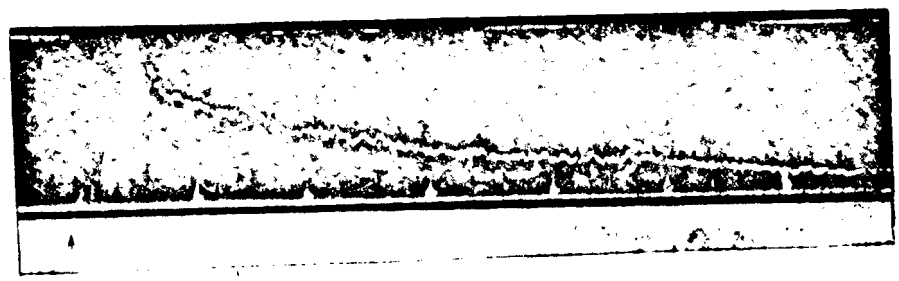

Fig. 3

*The microphone was calibrated at the All-Union Scientific Research Institute for Metrology D. I. Mendeleyev.

As long as the aerosol precipitation proceeded at low speed (low sound pressures) the nephelogram was obtained using the micrometer and stop watch. Fast processes were registered on the oscillograph plates. Fig. 3 shows a nephelometer recording plate (sound pressure $20 \cdot 10^3$ bar, $f = 5$ kc; time marks are every 15 sec; sound was turned on at the double mark).

Let us investigate basic relationships connecting the nephelometer readings with the characteristics of the aerosol being precipitated. The photomultiplier current I is connected at every instant to the intensity of the light beam by an approximately linear relationship $I_\Phi = kJ$. The attenuation of light inside the volume occupied by aerosol is due to scattering on particles and for $\bar{r} \gg \Lambda$ (Λ - wave length of light) is proportional to $\overline{nr^2}$, where n is the particle number density. The photomultiplier current is then expressed at the initial instant and at time t by the two expressions

$$I_{\Phi_0} = I_\infty \exp(-bl_2 n_0 \overline{r_0^2}),$$
$$I_{\Phi_t} = I_\infty \exp(-bl_2 n_t \overline{r_t^2}). \qquad (1a)$$

Eliminating $\exp(-bl_2)$ from the above equations one gets

$$I_{\Phi_t} = I_\infty \left(\frac{I_{\Phi_0}}{I_\infty}\right)^{\frac{n_t \overline{r_t^2}}{n_0 \overline{r_0^2}}} = I_\infty \left(\frac{I_{\Phi_0}}{I_\infty}\right)^{\alpha(t)}. \qquad (2)$$

The quantity $\alpha(t)$ determines the time variation of the photocurrent as function of the particle number density and the average radius squared. The latter is a function of time because of the coagulation and precipitation processes. Since one lacks the exact knowledge of the coagulation process, one can theoretically take account only of the (stockesean) precipitation and we calculated $\alpha(t)$ for this simplest case. Taking the element of volume at a distance h from the top part of the chamber one may write

$$f(rt) = \begin{cases} f_0(r) & \text{for } r \leqslant r_t \\ 0 & \text{for } r \geqslant r_t \end{cases}$$

Here $f_0(r)$ is fixed by equation (1) while $r_t = \sqrt{9\eta h/2g\rho t}$

corresponds to the stokesean radius of those particles which in the instant t reached the level h and crossed the beam (η - air viscosity, ρ - particle density, g - acceleration of gravity). Note that the apparent density of particle clusters (entering the expression for r_t) may be significantly lower than the true 5.5 g/cm^3 density of zinc oxide and approach the "poured on" density of 0.5 g/cm^3. Quantities entering the exponent of equation (2) are determined from the following expressions:

$$n_t = \frac{\int_0^{r_t} n_0 f_0(r)\,dr}{\int_0^{\infty} f_0(r)\,dr}, \qquad (3)$$

$$\overline{r_0^2} = \frac{\int_0^{\infty} r^2 f_0(r)\,dr}{\int_0^{\infty} f_0(r)\,dr}, \quad \overline{r_t^2} = \frac{\int_0^{r_t} r^2 f_0(r)\,dr}{\int_0^{r_t} f_0(r)\,dr}.$$

Using these relations we get

$$x_t = \frac{n_t \overline{r_t^2}}{n_0 \overline{r_0^2}} = \frac{\int_0^{r_t} r^2 f_0(r)\,dr}{\int_0^{\infty} r^2 f_0(r)\,dr}. \qquad (4)$$

In case of the logarithmically normal low of particle dimension distribution we obtain after integration of (4)

$$\alpha(t) = \frac{1 + Erf(x)}{2}, \qquad (5)$$

where

Expression (5) fixes the photocurrent in case of a spontaneous (purely stockseen) precipitation as function of time, of the parameters of the aerosol, and the height of the nephelometer light beam.

We used a compensating circuit for the nephelometer to increase the precision of readings and for convenience. Here, at any moment the measured current is obtained from the difference of the compensating current I_k and the photomultiplier current $I_t = I_k - I_\phi$. If $I'_\infty = I_k - I_\infty$ denotes the current for a zero concentration of the aerosol and $I'_0 = I_k - I_\phi$ is the current at time t = 0 when the concentration is the largest, one obtains easily from (2)

$$Erf(x) = \frac{2}{\sqrt{\pi}} \int_0^x e^{-t^2} \cdot dt, \qquad x = \frac{\lg r_t - \lg r_m - 1{,}6 \lg^2 \beta}{\sqrt{2} \lg \beta}. \qquad (6)$$

In the circuit used $I_k = 300 \mu a$, $I'_\infty = -95 \mu a$. Values of I'_0 for various beginning concentration were determined from the calibration curve on Fig. 4. The latter was obtained by weighing a zinc vapor receiver after accepting samples of air from the chamber.

The stability of calibration of the entire nephelometric device was checked again prior to every particular test using standard optical filters.

For the quantitative examination of sound coagulation of aerosols it is desirable to have a best possible uniform distribution of the sound field in the working chamber. It is not possible to reduce the dimensions of the chamber since it reduces the precision of the nephelometer (one reduces the "base" of the light beam). One obtains a sufficiently uniform sound field when $\lambda \ll l_1, l_2, l_3$, i.e., if there are many wave lengths fitted into the length, width, and height of the chamber, or when $\lambda > l_2, l_3$, i.e., when the working conditions approach the plane wave situation. With the values of l_1, l_2, and l_3 given above the first case is realized for f > 5 kc

and the second for f $\leq$ 200 cycles ($\lambda \geq$ 1.7 m).

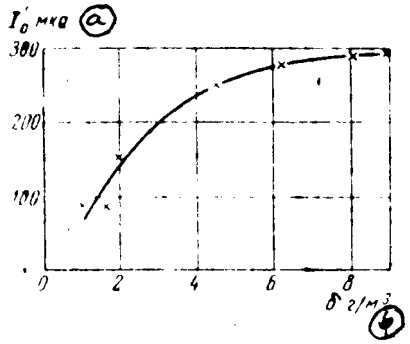

Fig. 4

а - μa; b - g/m^3

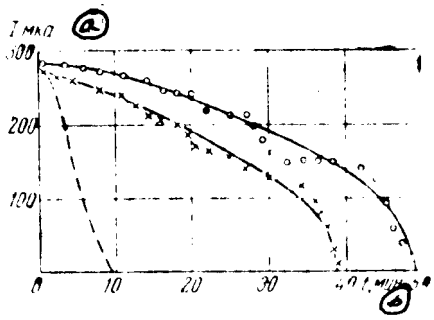

Fig. 5

а - μa; b - min

Let us look at the results. Fig. 5 presents the nephelometer current as function of the time for the spontaneous process. Precipitation time (up to a concentration of 0.6 g/m^3) is quite appreciable, of the order of 40-50 min. This is compared on the same graph with the dotted theoretical curve calculated using Equation (6). The initial aerosol concentration was taken as δ = 7 g/m^3 (I_0' = 280 μa), particle density as ρ = 1 g/cm^3, while the values for r_m and β were taken from the distribution function (Fig. 1). The comparison of the theoretical and experimental curves on Fig. 5 indicates that the convection air currents play an important role and significantly retard the precipitation of aerosols. The observed curve fluctuations should be likewise a consequence of the air currents.

The aerosol precipitation is accelerated under the influence of sound oscillations. Fig. 6 shows the precipitation curves taken at a 5 kc frequency and different sound pressures (1 - p = 0; 2 - 2·10^3 bar; 3 - 5.5·10^3 bar; 4 - 11.2·10^3 bar; 5 - 15·10^3 bar; 6 - 23·10^3 bar; 7 - 28.5·10^3 bar; 8 - 34·10^3 bar). At low pressures

the precipitation is qualitatively close to the spontaneous process. The decrease in time of nephelometer current is relatively slow, particularly during the starting interval, and one explains this behavior by a weak coagulation of the aerosol and slow precipitation of those particles which are located above the nephelometer beam. At larger sound pressures the transparency of aerosols increases rapidly on the account of fast growth in particle size and the subsequent precipitation.

A visual observation of the aerosol precipitation process uncovered the following peculiarities. With undamped chamber walls one sees a violent production of vortices and a noticeable production of sizeable particle aggregates. The aerosol precipitation occurs almost uniformly across the whole surface with the particles collecting at the velocity nodes at the floor of the chamber. A partial damping of the walls sharply reduces the vortices which, however, does not reduce significantly the precipitation speed as registered on the nephelometer (keeping the p-values the same). A displacement of the nephelometer beam along the horizontal does not practically influence the course of the curves.

The relationship between the aerosol precipitation time t_c as function of p for f = 5 kc is presented on Fig. 7. With p = $34 \cdot 10^3$ bar one finds $t_c \approx 35$ sec, i.e., almost two orders of magnitude smaller than the time for the spontaneous process. It is interesting to note that at a given frequency in the case of larger sound pressures ($p > 10^4$ bar) one finds a close agreement with the expression $t_c p^2 = \text{const}$. The latter indicates a very strong influence of sound pressure on the precipitation speed.

We investigated the frequency dependence of the precipitation speed in a wide range from 200 to 21,000 c/sec. Fig. 8 shows the frequency dependence at a constant p = 3,500 bar and an aerosol concentration of ~ 5 g/m^3. The ordinate indicates the variation of the nephelometer current per unit time. We observed a weak frequency effect and an insignificant rise of curves in the low frequency region even at higher sound frequencies. Although, generally, we did not find any practical frequency dependence on the polydispersed aerosol under consideration, the thinly dispersed fraction ($r < 1\,\mu$) did precipitate at low frequencies significantly slower than at higher frequencies. From this point of view it may seem probable that a wide distribution of particle sizes leads to a certain smoothing out of the frequency dependence of the precipitation speed.

Our results are of immediate practical interest. A sufficiently high precipitation speed at low frequencies indicates the way in which to increase the economic efficiency of the acoustic method of aerosol coagulation by designing low frequency devices with high mechanical

efficiency. The use of frequencies in the hundred cycle range may prove to be quite efficient.

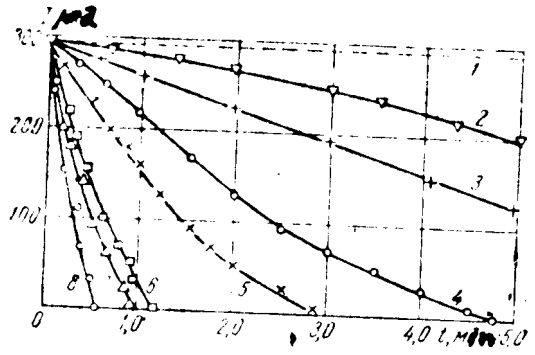

Fig. 6

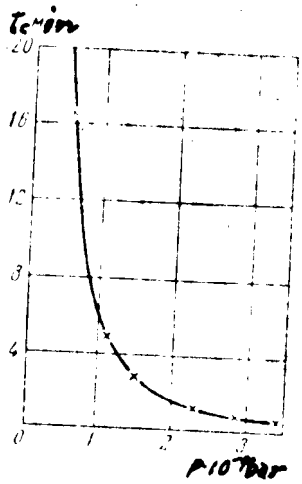

Fig. 7

At the present time we do not possess a definitive theory explaining the acoustical coagulation of aerosols. In view of this, our experimental results are of particular interest. The high precipitation speed at low frequencies compel us to conclude that the coagulation mechanism cannot be explained by King forces /2/. As is well known, these latter forces vary inversly with the wave length (of the standing wave). Elementary estimates indicate that at frequencies of the order of a few hundreds cycles per second the King forces are too weak to cause a significant approach of zinc oxide aerosol particles. The relatively uniform precipitation of aerosol inside the entire volume of the chamber indicates also that there is only a small effect due to the radiation pressure.

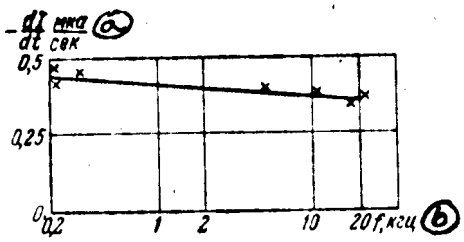

Fig. 8

a - μa/sec; b - kc/sec

Particle motions under the influence of strong sound oscillations correspond to the Reynolds numbers of Re $\geq$ 1 and one must treat them considering the Oseen attractive forces. In this case the effective coagulation mechanism may be due to the hydrodynamic interaction, as was shown by Pshenai-Severin /3/. Under the Oseen conditions the hydrodynamic interaction is much stronger than during the potential flow. In particular, the R dependence of the forces becomes R^{-1} instead of R^{-4}. Pshenai-Severin estimated the time of approach of two particles of equal radius r = 1-15 μ and ρ = 1 g/cm^3, p = $16 \cdot 10^3$ to $40 \cdot 10^3$ bar with f = 100-15,000 c/s, which numbers are close to the conditions during our experiments. At highest values of p the time of approach may reach fraction of a second. This is a fully acceptable value for the qualitative explanation of the fast precipitation of zinc oxide aerosols which we observed at high sound pressures. One should emphasize, however, that in the case of small interparticle distance these calculations cannot be accepted. Hydrodynamic forces in the Oseen approximation as well as the orthokinetic mechanism /4/ should lead to the observed maxima in the frequency dependence of the coagulation speed. The optimum values of f in both cases, as fixed by the size of the coagulating particles and their order of magnitude, are not very different. Hydrodynamic forces for r ~ 2 μ particles give for our case an $f_{opt} \sim 5 \cdot 10^3$ c/s and for r ~ 6 μ an $f_{opt} \sim 5 \cdot 10^2$ c/s. It is quite natural that the polydispersed character of aerosols leads to a smoothing out of the frequency dependence. One must regret that both the orthokinetic and

and hydrodynamic coagulation mechanisms are not presently developed for the case of polydispersed aerosols. This fact does not permit a qualitative comparison with experimental results.

Entered
November 15, 1960

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