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Coagulation of cement paste due to ultrasonic vibrations is described. Various mechanisms (theories) of coagulation are compared with the results of the experiments.

Numerous experiments on the coagulation of the dispersed phase in aerosols have led to several tentative theories of acoustic coagulation [1-4], each of which can be used for examination of the coagulation process. However, there are hardly any experimental data on the acoustic coagulation of hydrosols.

In our investigations we used two methods of ultrasonic treatment of cement paste: the deep method, where a waveguide was inserted into the interior of the sample, and the surface method, where cement paste in a mold was mounted on a vibrator. We used a frequency of 19.6 kc in the deep method, and 11.0 and 25.6 kc in the surface method.

As a result of deep treatment the bulk density of the cement increases by 5.5-9.5% with increase in the W/C ratio (in the cohesion range) and the compressive strength of the solid cement increases correspondingly by a factor of 1.5-2.0 [5].

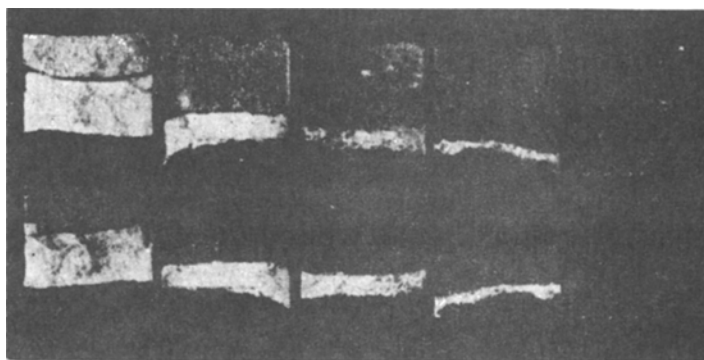
Surface treatment of the cement paste leads to the appearance of regions (centers) characterized by dark color and high density. These compaction centers are formed at the surface of contact between vibrator and cement paste, irrespective of the position of the vibrator. With increase in the duration of treatment the centers fuse together into a continuous dense layer, outside which lies a pale-colored friable mass. The thickness of the dense layer increases with increase in the water-cement ratio (see figure) and decrease in vibration frequency. An increase in the intensity of the ultrasonic vibrations accelerates the formation of compaction centers.

It should be noted that the resultant water-cement ratio in the dense layer corresponds to the value where all the water is in the bound state. The 28-day strength of the solid cement of the dense layer reached 200 mN/m<sup>2</sup> as against 50 mN/m<sup>2</sup> for control cement. The strength of solid cement from the loose pale mass was barely 3 mN/m<sup>2</sup>.

The increase in the strength of the solid cement as a result of deep ultrasonic treatment is due to homogenization of the cement paste and enhancement of self-coagulation of the particles due to liberation of additional surface energy in the dispersion of the solid phase.

Compaction of the cement paste by ultrasonic surface treatment can be compared with compression under high pressure. For instance, in L'Hermite's investigations solid cement with a compressive strength of 160 mN/m<sup>2</sup> was obtained. For this purpose cement with 8% water was compressed for 2 hr at a pressure of up to 200 mN/m<sup>2</sup> [6]. Other methods of treating cement paste do not lead to such high strengths.

According to the existing ponderomotor theory, coagulation of particles in an acoustic field is due to the forces of attraction (Bernoulli forces) developed between them under the influence of the acoustic flux. The attractive forces increase with increase in the size of the particles, the velocity of the flow (of liquid in this case) between them, and reduction in the distance between particles. The hydrodynamic attractive forces developed between the particles by the



Structure of solid cement after surface ultrasonic treatment of cement past with W/C (right to left) 0.23; 0.265; 0.35; 0.44; 0.53

flow of liquid act in a direction perpendicular to the direction of propagation of the ultrasonic waves. Since ultrasonic waves are propagated in straight lines, the particles in the cement paste approach one another only in a direction perpendicular to the propagation of the vibrations. In addition, when compaction centers are formed, the cement grains move in the direction of propagation of the elastic waves through a distance of up to one-quarter of the wavelength. This indicates that ponderomotor coagulation does not occur in the formation of compaction centers.

The orthokinetic theory of coagulation is based on the difference in the amplitudes of vibration of particles of different dispersity. Owing to their high mobility the small particles collide with the larger (less mobile) particles and coagulate on their surface.

This situation occurs in cement paste, since the dispersity of the cement varies from hundredths of a micron to approximately 200 microns. The collision of particles is probable owing to the following factors: a) the high concentration of the solid phase and the minimal distance between particles, which depends on the thickness of the solvate sheaths, approximately  $0.25-0.30 \mu$  for cement with a specific surface of about  $7000 \text{ cm}^2/\text{g}$  and normal consistency; b) the high vibration frequency, up to 20 000 vibrations per second.

However, as observations showed, compaction centers do not arise when the cement paste is treated with a deep ultrasonic vibrator, despite the fact that in this case the same conditions for the induction of coagulation apply. The compaction centers that follow surface treatment consist of compactly arranged large grains, whereas, according to the theory of orthokinetic coagulation, particles of greatly differing size should participate in the aggregation.

In a high-intensity acoustic field confined by walls acoustic streaming takes place. In the opposing flows at turns close to the vibration nodes of the standing wave the entrained particles drop out and, on colliding, coagulate. The collision of the particles is due to the fact that the acoustic streaming at the vibration nodes is directed away from the surface, whereas at the antinodes it is directed toward the surface, with the result that coagulation occurs close to the surface of the sample.

The theory of intensification of Brownian coagulation by acoustic vibration postulates the high probability of the collision of small particles with large particles, which are not involved in the Brownian motion, but vibrate along with the dispersion medium. If Brownian movement is assumed to be the cause of coagulation, then particles of different size would be uniformly distributed in the compaction centers and outside them, and the only difference would be the removal of free water from the compaction centers. As already mentioned, however, only large grains are concentrated in the compaction centers and outside these centers only small ones.

The radiation theory postulates the existence of radiation pressure in the acoustic field. The radiation pressure causes particles denser than the medium to concentrate at the vibration antinodes of the standing wave and coagulate owing to activation of the forces of attraction between the vibrating particles.

The progression of particles in the direction of the ultrasonic waves becomes more intense for larger particles and higher ultrasonic frequency.

When cement paste is treated with ultrasound, compaction centers are formed at the surface of the vibrator. Only the large fractions of the cement take part in their formation. This effect can be attributed to displacement of the large fractions of the cement by radiation pressure to the vibration node of the standing wave situated at the surface of the vibrator. At the same time, water is extruded from the cement paste and the very small particles suspended in the water (which are least affected by radiation pressure) are displaced to the node of the standing wave. The absence of compaction centers at other vibration antinodes of the standing wave set up in the cement paste can be attributed to the very pronounced attenuation of ultrasonic vibrations in this medium [7].

In a cement paste with  $W/C = 0.5$  and a vibration frequency of 25.6 kc the "vibration antinode - vibration node" zone occupies a depth of 14 mm (wavelength 56 mm), and the thickness of the compaction layer is 5.6 mm. For this case the displacements of the particles due to radiation pressure, calculated from Saint-Clair's equation, for a vibration amplitude of  $3 \mu$ , maximum duration of treatment 180 sec, and viscosity of the cement paste at  $W/C = 0.5$  (in the case of maximally destroyed structure) equal to  $0.057 \text{ N} \cdot \text{sec}/\text{m}^2$  [8, 9] are given in the table. These data show that particles of more than  $20 \mu$  can move within 180 sec from any point in the "antinode - node" zone into the compaction layer, whereas particles of less than  $20 \mu$  move through a very small distance and do not take part in the formation of the dense layer.

It follows from the foregoing that the creation of compaction centers is due to radiation pressure. The acoustic field gives rise to solid cement of two different structural types, differing in color, density, and strength. The results of experiments involving ultrasonic treatment of a cement-sand solution provided confirmation of this.

Treatment of a 1 : 1 solution composed of Vol'sk and ground quartz sand showed that the sand fractions did not take part in the separation of the mixture into two layers, since acoustic coagulation affects only the cement grains owing to their specific properties in cement paste. The cement grains in the paste are surrounded by sheaths of oriented

water molecules and form dipoles, which in turn are oriented by the ultrasonic vibrations and thus cause the formation of a cement paste with a compact spatial structure. This peculiarity of cement paste distinguishes it from other disperse systems.

With reduction in the vibration frequency the zone of thixotropic liquefaction and the thickness of the compacted layer of cement paste increase owing to the lower absorption of vibrational energy with increase in wavelength. If a suitable low vibration frequency is selected, the zone of compaction of the cement paste increases so much that it becomes possible to use acoustic coagulation in production conditions. According to preliminary data, the best frequency to ensure compaction of the cement paste to a sufficient depth is about 1000 cps. The lower frequencies produced by conventional electromechanical vibrators cannot create a radiation pressure of sufficient strength to move the cement particles through the distance required. In this case, owing to the extremely low radiation pressure, the cement paste must be treated for several days. Thus, manifestation of the radiation pressure effect in cement paste requires very high frequencies, which, however, owing to the small amplitude of the vibrations, do not cause the displacements of the grains of sand and coarse aggregate necessary for compaction of the concrete. In this case multifrequency vibration must be used. The aggregate structure must first be compacted at low frequency and then the cement mix treated at high frequency.

Displacement of Cement Particles  
in Cement Paste  
Due to Radiation Pressure

Initial distance from particle to vibration node (mm)	Displacement (mm) for particles of size				
	60 $\mu$	40 $\mu$	20 $\mu$	2 $\mu$	0.2 $\mu$
0	0	0	0	0	0
3.5	9.80	6.20	1.36	0.01	0
7.0	6.70	5.18	1.66	0.02	0
10.5	3.40	2.72	1.05	0.01	0
14.0	0	0	0	0	0

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