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Acousto-modulation speckle correlometry of evolving foams: the influence of acoustic stimulation on foam aging dynamics

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Received December 25, 2025
Revised December 25, 2025
Accepted December 25, 2025

The experimental findings on the evolution of the structure of model samples of gas-liquid foams using the speckle correlometry method are presented. The experiments were carried out with low-frequency acoustic impact (at a frequency of 2 kHz) on the studied samples and in the absence of impact. The used mode of sounding the sample corresponds to a cyclic alternating change in excess external pressure within the area of interaction of the laser beam with the foam. Accounting for the relationship between the length of the sound wave and the characteristic dimensions of the interaction zone, one can conclude that the excess pressure is quasi-uniformly distributed across the interaction zone. In both cases it was found that there was a self-similar nature of evolution of the foam structure, in which the average size of gas cells in the foam increased over time according to a power law with an exponent equal to 0.5. At the same time, the acoustic impact leads to a significant increase of the velocity constant in the power law and a more rapid growth of correlation time of fluctuations in the intensity of laser radiation scattered by the foam compared to samples of unsounded foam as the studied samples age. The qualitative interpretation of the features observed in the experiment is discussed.

Keywords: gas-liquid foams, structure evolution, multiple light scattering, speckle correlometry, acoustic impact.

DOI: 10.61011/TP.2026.05.63529.346-25

Introduction

The structural and physicochemical properties of foamed liquid media have been the subject of ongoing research interest for about a century and a half, starting with the classic work of Kelvin [1]. Numerous experimental and theoretical studies on the evolution of gas-liquid foams, from their origin to the stabilization of the foam structure or its destruction, cover such fundamental sections of physics as thermodynamics, kinetics, statistical physics, the physics of capillary and surface phenomena, etc. On the other hand, such studies are largely characterized by a pronounced practical focus, due to the widespread use of foam-like materials in modern science-intensive technologies, biomedical applications and everyday life. In this regard, one can note, e.g., metal foams as structural materials with exceptional performance characteristics [2–4], highly porous scaffold matrices based on foamed bioresorbable polymers [5–7], highly effective sound- and heat-insulating layers of polymer foams [8–10], etc.

Under constant external conditions (pressure and temperature) and in the absence of gas exchange with the environment, gas-liquid foams are thermodynamically metastable randomly inhomogeneous two-phase systems, in which slow changes in the statistical characteristics of the cells ensemble (gas bubbles) occur: the average radius of the cells $\langle R_c\rangle(t)$ and the dispersion of the distribution of cells by radius $\sigma_R^2(t)$ [11–15]. At the same time, the volume fraction of the liquid phase f gradually decreases, flowing under the action of gravity to the bottom of the foam-containing container along the walls of the bubbles and the network of Plato-Gibbs channels [16–18]. As a result, in gas-liquid foams there is a gradual transition from the state of „wet" (wetfoam, $f > 0.1$) to the state of „dry" foam (dryfoam, $f < 0.05$). The change in $\langle R_c\rangle(t)$ and $\sigma_R^2(t)$ during the slow evolution of foam is caused by the influence of gas exchange between adjacent cells through the walls separating them and the coalescence of adjacent cells during destruction of the separating walls. Gas exchange between cells is caused by diffusion flows of gas through the walls due to differences in excess Laplace pressure (and, accordingly, local concentrations of gas molecules) in the neighboring cells [17]. As a result, smaller pores are gradually absorbed by larger ones, and the average radius increases monotonically. It has been established [19] that the cell size distribution during slow foam evolution has a self-similar character, with the distribution shape being approximated with acceptable accuracy by a lognormal law, and the average cell radius changing with time as $\langle R_c\rangle(t) \propto t^{1/2}$.

Research into the dynamics of local changes in foam structure due to cell coalescence processes and absorption of small cells by large ones is of considerable interest both from the standpoint of establishing the fundamental features of foam structure formation at the mesoscopic level and for practical applications related to the synthesis of foam-like materials or the use of gas-liquid foams as technological agents (monitoring of foam structure in the near real time condition).

An acceptable approach to the experimental implementation of contactless monitoring of local structural rearrangements in evolving gas-liquid foams is the speckle correlometry method, based on the effect of multiple dynamic scattering of laser radiation in the probed media. In this case, the correlation time of fluctuations in the intensity of speckle-modulated laser radiation is analyzed depending on the characteristics of the probed systems structure. It should be noted that previously the diagnostic methods using the effect of dynamic speckle modulation of multiply scattered laser radiation in non-stationary media (in particular, diffusion-wave spectroscopy [20,21]) were applied by various research groups to study the dynamics of gas-liquid and polymer foams progress. In this connection, special mention should be made of the work of D. Durian and co-workers on diffusion-wave spectroscopy of evolving foams [22–24].

The method of laser speckle correlation probing using low-intensity laser radiation is a non-invasive diagnostic approach that does not have any significant impact on the probed medium. This is one of its main advantages (especially in biomedical applications and in solving delicate diagnostic problems in physical materials science). At the same time, controlled physical effects on the probed medium during speckle correlation diagnostics, which do not lead to fundamental changes in its structural and physicochemical properties, will significantly expand the functional of diagnostics. A typical example is the study of the temperature effect on the correlation time of scattered laser radiation fluctuations intensity, which is essentially currently one of the routine procedures in the physical materials science of colloidal systems.

The efficiency of scattering of acoustic radiation and the nature of interaction with randomly inhomogeneous media depends on the ratio of the sizes and shapes of local heterogeneous inclusions and the length of the acoustic wave [25–28]. It is worth noting that high-intensity ultrasound exposure to metastable foam-like media can lead to nonlinear effects, such as self-adaptation of liquid films in cells, vortex flows of the liquid component, etc. [29,30]. In addition, sound impact in the low frequency range can, in certain conditions, influence the stability of gas-liquid foam [31,32].

In view of this, it is of interest to conduct speckle-correlometric diagnostics of evolving foams simultaneously with low-frequency acoustic impact on the probed volume of foam.

The propagation of acoustic radiation in a randomly inhomogeneous structure of gas-liquid foam may, in some way, impact the local structural rearrangements in clusters that unite several adjacent cells, accelerating or, conversely, suppressing them.

The study was aimed at implementing such method experimentally, which can be defined as „acoustic-modulation speckle-correlometry“, to study the effect of acoustic action on the dynamics of local structural rearrangements in evolving gas-liquid foams, and the interpretation of the obtained experimental data.

1. Experimental technique

Figure 1 shows a diagram of the laboratory speckle correlometric system used with the simultaneous effect of low-frequency acoustic radiation on the probed foam volume. The GN-5P helium-neon laser (wavelength 633 nm, output power 5 mW, linear polarization, manufacturer — NPO „Plasma“, Ryazan, Russia) was used as the radiation source *1*. The studied sample of gas-liquid foam was located in a cuvette *2*; the acoustic effect on the probed volume was carried out using a piezoelectric UL103 actuator (*3* Hunan Mac Sensor Co., Ltd), excited by a harmonic signal with an amplitude of 100 mV and a frequency of 2 kHz from a dedicated signal generator Agilent-33220A. Speckle-modulated laser radiation scattered in front of the foam sample was registered with a frame rate of $1000\,s^{-1}$ using CMOS camera OptronisCamRecord 3000 with a lens TamronforNikon 70-210 *4*. The camera-lens optical system focused on the sample surface, and the typical number of speckles within the frame ranged from 22 to 25.

Gillette FoamyRegular shaving cream was used as model samples of the evolving gas-liquid foam, which was pumped into glass cuvettes 75 mm high, 25 mm wide and 3 mm thick using a syringe immediately after discharge from a cream bottle. It should be stressed that during the experiment, the average diameter of the cells (gas bubbles) in the foam did not exceed $100\,\mu$m, which provided a significantly multiple scattering mode during the propagation of probing radiation in the foam layer. Indeed, according to [22], the typical value of the visible range radiation propagation transport length in foam with an average cell diameter $\langle D\rangle$ is approximately $\langle 3.5D\rangle$, which is significantly less than the thickness of the cuvette.

After pumping the foam samples, the cuvettes were sealed with glue with glass plates 1.3 mm thick to minimize evaporation of the liquid phase from the foam; foam samples were sounded through these cover plates. To control the amount of liquid phase losses during the experiment, sample cuvettes were weighed at the beginning and end of the experiment using a DL-123 laboratory scale (DEMCOM, China, with an error of at least 0.01g). It was found that the loss of the liquid phase due to evaporation did not exceed 1 % of the initial mass of the samples. Volumetric measurements showed that the volume fraction of the liquid phase f in the studied samples was ~ 0.07. During the experiments, microscopic images of the foam surface in cuvettes were also recorded at linear magnification of $2\,\mu$m/pixel. As a result of statistical processing of the

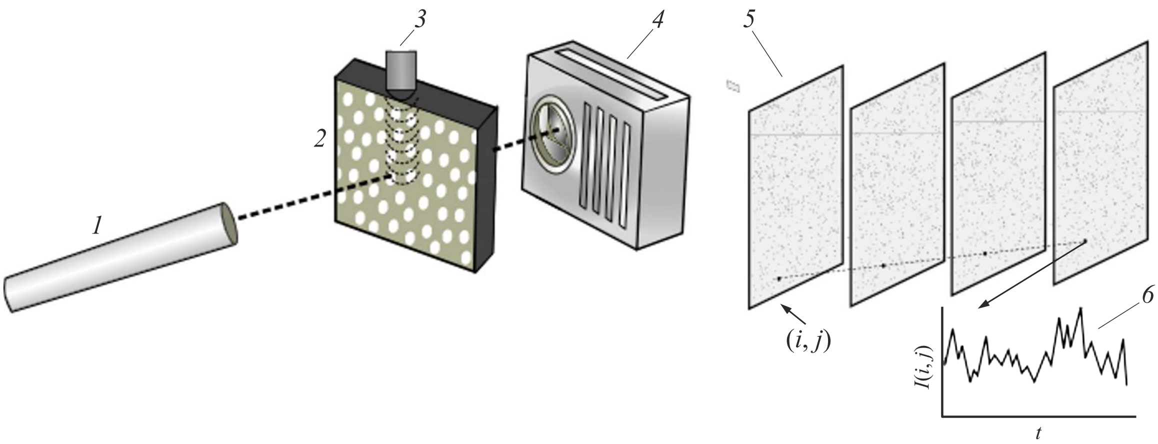

Figure 1. Experimental setup diagram: *1* — GN-5P helium-neon laser (633 nm, 5 mW); *2* — cuvette with foam sample; *3* — piezoelectric actuator; *4* — CMOS camera; *5* — schematic representation of the sequence of recorded frames; *6* — high-quality image of the temporal realization of intensity fluctuations in an arbitrarily selected pixel.

obtained images, the average radius $\langle R_c \rangle(t)$ of cells at various time points was then determined.

The analysis of speckle-modulated image sequences registered by CMOS camera OptronisCamRecord 3000 (position *4* in Fig. 1) was performed using the following algorithm:

— fragments with a duration of 30 s were isolated from the received video sequences and their shot-by-shot breakdown was performed;

— in each frame, a pixel with coordinates i, j was randomly selected in the needed fragment, for which the temporary realization of the recorded brightness $\{I_{i,j}^k\}$ was restored within the fragment, where k is the frame number;

— a normalized autocorrelation function was calculated for the selected pixel:

$$g_{i,j}^2(\Delta k) = \sum_{m=\Delta k}^{N-\Delta k} (I_{i,j}^m - \overline{I_{i,j}})(I_{i,j}^{m+\Delta k} - \overline{I_{i,j}}) \Big/ \sum_{m=\Delta k}^{N-\Delta k} (I_{i,j}^m - \overline{I_{i,j}})^2, \quad (1)$$

where $\overline{I_{i,j}}$ — the average value of the recorded pixel brightness over $\{I_{i,j}^k\}$ sample, N — the number of frames in the sample;

— this procedure was repeated for M randomly selected pixels, after which the normalized autocorrelation functions were averaged over the resulting ensemble;

— based on the obtained averaged autocorrelation function $\langle g_{i,j}^2(\Delta k) \rangle$, a non-integer value $\tilde{k}$ was generally determined, corresponding to a decline $\langle g_{i,j}^2(\Delta k) \rangle$ by e times; it was then used to calculate the fluctuations intensity correlation time $\tau_c \approx \tilde{k}/f$ at the time corresponding to the middle of the processed video sequence fragment.

Such a double averaging of the sample speckle correlometry data (over time and over the ensemble) significantly increases the robustness of the estimates of the intensity fluctuations correlation time.

2. Experimental results

Figure 2 shows the dependences of the average radius of cells (gas bubbles) $\langle R_c \rangle$ on time in logarithmic coordinates in the absence of *1* and in the presence of *2* constant acoustic effect on the model foam samples. The values $\langle R_c \rangle$ were obtained as a result of statistical processing of raster microscopic images of the evolving foam structure recorded during the experiment.

This result is in good agreement with the known ratio for evolving gas-liquid foams at relatively small values of the volume fraction of the liquid phase $f \leq 0.2$: $[\langle R_c \rangle(t)]^2 - [\langle R_c \rangle(t_0)]^2 \approx \alpha(t - t_0)$ (e.g., see [16]). The values of $\langle R_c \rangle(t_0)$ and t_0 correspond to the transition to a self-similar mode of foam evolution, and parameter α can be considered as a velocity constant for the square of the average radius of the cells. Obviously,

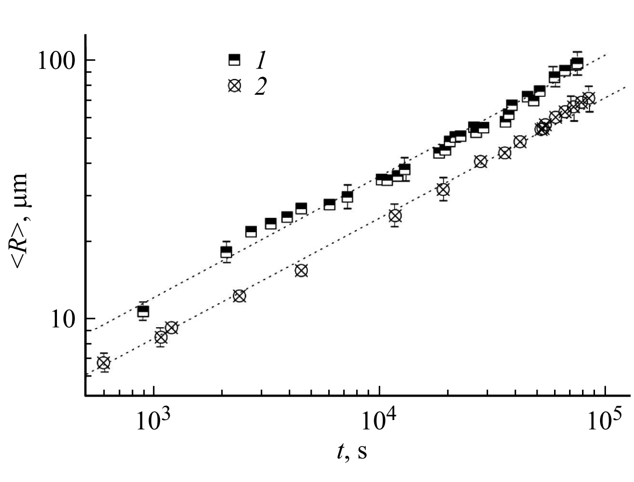

Figure 2. Dependences $\langle R_c \rangle$ on time during acoustic exposure to the test samples (*1*) and in the absence of acoustic exposure (*2*). The dotted lines correspond to approximation $\langle R_c \rangle \propto t^{0.5}$. The selectively shown confidence intervals correspond to a significance level of 0.9.

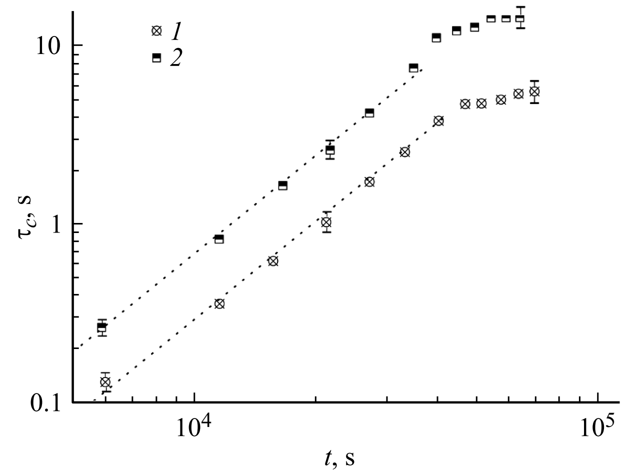

Figure 3. Dependence of the correlation time of the laser radiation intensity scattered by evolving foams on the time of experiment in the absence of acoustic exposure (*1*) and under exposure (*2*). The correspondence between the markers and the displayed data is the same as in Fig. 2. The dotted lines correspond to $\tau_c \approx \tilde{\alpha} t^{1.73}$. The selectively shown confidence intervals correspond to a significance level of 0.9.

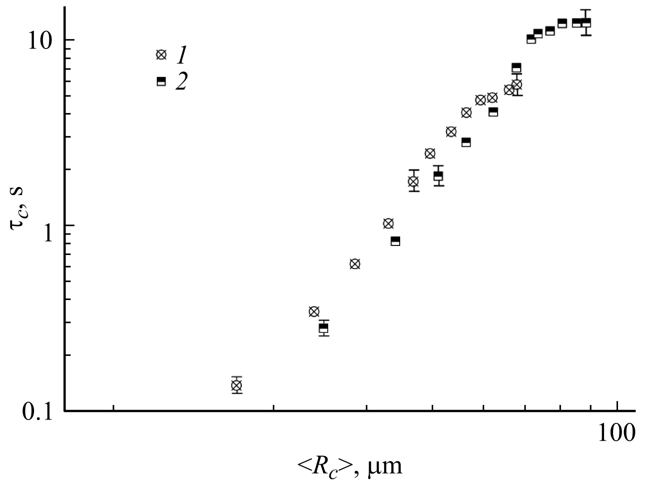

Figure 4. The relationship between the current values of the average cell radius and the correlation time of the scattered laser radiation intensity fluctuations for the studied model foam samples in the absence of acoustic exposure (*1*) and under exposure (*2*). The selectively shown confidence intervals correspond to a significance level of 0.9.

for $t \gg t_0$ and respectively $\langle R_c \rangle(t) \gg \langle R_c \rangle(t_0)$, there is a ratio $[\langle R_c \rangle(t)]^2 \approx \alpha t$ that is acceptably consistent with the experimental data presented in Fig. 2. At the same time, the parameter α in case of acoustic exposure is significantly (approximately 2.15 times) higher than the corresponding value in the absence of acoustic exposure. A possible interpretation of this feature is discussed in sec. 3.

Figure 3 shows the results of speckle correlation analysis of structural changes in evolving samples of model foams as smoothed correlation time dependences of the scattered laser radiation intensity fluctuations on the evolution time in the absence and in the presence of acoustic effects. The dependences are given in logarithmic coordinates and over short aging times (up to $\sim 4 \cdot 10^4$ s), power-law approximations $\tau_c \approx \tilde{\alpha} t^{\beta}$ are acceptable with appropriate accuracy, where the indicator $\beta \approx 1.73$ is almost the same for both data sets, and the normalization coefficient $\tilde{\alpha}$ for acoustic exposure is about 2.27 times greater (which is close to the ratio for constants speeds α).

For further discussion, the relationship between the current values of the average cells radius $\langle R_c \rangle$ and the correlation time τ_c of intensity fluctuations in experimental conditions is of interest. Figure 4 shows a similar relationship in the absence and presence of acoustic effects.

3. Discussion of results

As follows from the experimental findings, the influence of low-frequency acoustic effects on evolving model foams leads to an increase in the rate constant α, which determines the kinetics of increasing the square of the average radius of cells during foam aging (foam coarsening in English terminology) and the normalization coefficient $\tilde{\alpha}$ for the correlation time of intensity fluctuations. To interpret these features, it is necessary to consider, on the one hand, the main physical mechanisms that control local rearrangements of the foam structure during its evolution. On the other hand, it is necessary to account for the integral effect of these local structural rearrangements on the optical transport parameters of the evolving foam, which impacts the temporary decorrelation of intensity fluctuations. Based on the basic relation of diffusion-wave spectroscopy for scattered field fluctuations (see, for example, [20]):

$$|g_1(\tau)| = \int_0^\infty \exp\left(-\frac{k_0^2 \langle \Delta r^2(\tau) \rangle}{3} \cdot \frac{s}{l^*}\right) \rho(s) ds, \qquad (2)$$

It can be concluded that higher transport length l^* of the propagation of probing radiation in the medium obviously leads to a slower decrease in the time correlation function of scattered field fluctuations (and, accordingly, the shifted autocorrelation function of intensity fluctuations $g_2(\tau) = |g_1(\tau)|^2$), determined in this case from experimental data. In expression (2) $\rho(s)$ — probability density of the values of the propagation paths s of the partial components of the scattered light field in the probed medium, k_0 — wavenumber of the probing radiation, $\langle \Delta r^2(\tau) \rangle$ — average square of the displacement of the scattering centers during τ. Note that during the foam evolution, there is both an increase in the transport length l^*, proportional to $\langle R_c \rangle(t)$ [22], and a decrease in the mobility of scattering centers in the foam, the role of which is performed by interphase boundaries: $d\left(\sqrt{\langle \Delta r^2(\tau) \rangle}\right)/dt \propto d\langle R_c \rangle(t)/dt \propto t^{-\frac{1}{2}}$.

Based on the relationship between the current values of the average cell radius in the evolving foam and the correlation time of the scattered laser radiation intensity fluctuations, shown in Fig. 4, we can assume a fairly universal relationship between $\langle R_c \rangle$ and the parameter $d\langle R_c \rangle(t)/dt$, which characterizes the mobility of the interfacial boundaries averaged over the ensemble and, accordingly, the correlation time of intensity fluctuations. At least, such versatility, resulting from the power-law dependences $\langle R_c \rangle$ and τ_c on the foam aging time (Fig. 2, 3), manifests itself at $\langle R_c \rangle$ values not exceeding $80-90\,\mu$m. The saturation of τ_c values, which occurs at high values of the aging time and, accordingly, $\langle R_c \rangle$, is presumably due to the transition from a significantly multiple mode of dynamic scattering of laser radiation, described by the expression (2), to low forward scattering. A similar dynamic scattering mechanism „is activated“ when the transport length l^* becomes comparable to the thickness of the scattering layer. The change in the transport length during the evolution of the gas-liquid foam is also interrelated with a change in another radiation scattering parameter — scattering length l_s. Such interrelation is provided by anysotropy parameter g according to the ratio $l^* = l_s/(1-g)$. For the isotropic scattering mode, the anisotropy parameter is $g \approx 0$, for the anisotropic scattering with preferential forward scattering, the anisotropy parameter is $g \approx 1$. In the gas-liquid foam model, the development of the system over time leads to a change in the scattering mechanisms. And, as was shown in [33], during the time intervals covered in the experiment, an „optical“ inversion occurs as a transition from the scattering mode of gas bubbles in a liquid to the scattering by areas of intersections of foam cell boundaries in a gaseous matrix medium, which is accompanied by an increase in the scattering anisotropy parameter g from values of the order of 0.1 to values greater than 0.6.

The effect of acoustic modulation on the velocity constant found in this study can be qualitatively interpreted complying with the mechanism of local rearrangements of the foam structure, for the studied range of values of the liquid phase volume fraction, primarily associated with the diffusion transfer of gas from smaller neighboring cells to larger ones. As per [34] this process is delineated by the expression $dR/dt \propto 1/R - 1/R_{\text{cr}}$, where R_{cr} — critical radius of the cell. The critical radius of a gas cell of a gas-liquid foam is the conditional radius of instantaneous diffuse equilibrium with surrounding cells: gas cells with a radius greater than the critical one will increase due to the diffusion of gas from cells with radii less than the critical one. This process is controlled by the Laplace excess pressure difference inside a given cell and the critical pressure averaged over an ensemble of cells, determined by the average cell size. In the process of foam evolution, the critical radius increases over time [19].

The combination of local gas transfer processes between neighboring cells determines the rate constant α. According to [35], this parameter, called in English literature „the coarsening rate“, depends on a number of factors, including external pressure. It should be borne in mind that in the case of low-frequency acoustic modulation of an evolving foam, the length of the acoustic wave in foam is many times larger than the characteristic size of the area of interaction of laser radiation with the medium. Indeed, in the low-frequency region, the typical velocity of sound waves in gas-liquid foams with small volume fractions of the liquid phase is ~ 50 m/s (see, for example, [30]). At an acoustic impact frequency of 2 kHz, the sound wave length is approximately 2.5cm. Thus, considering that the transverse dimensions of the interaction zone are several millimeters, it is possible to assume a quasi-uniform distribution of alternating overpressure over the interaction zone. Such an effect, even with relatively small values of the amplitude of the sound wave, can lead to an intensification of local structural rearrangements in the probed volume and gas exchange between neighboring cells, which presumably is the reason for higher velocity constant α under acoustic influence.

Conclusion

Thus, as a result of conducted research, it was found that both in case of low-frequency acoustic effects on the evolving gas-liquid foam, and in the absence of exposure, there are universal relationships between the aging time and the ensemble-averaged characteristics of local structural rearrangements, leading to a self-similar mode of structure evolution corresponding to the power dependence of the average cell radius on the aging time with the indicator, equal to 0.5. At the same time, the acoustic effect has a great impact on the velocity constant corresponding to this self-similar mode, leading to its significant rise. This feature is presumably due to the intensification of local gas exchange between neighboring cells under acoustic influence.

Further research will be aimed at determining the influence of various factors (including temperature, frequency and amplitude of the sound wave) on the kinetics of structural rearrangements in evolving foams.

Funding

The study was supported by a grant from the Russian Science Foundation № 25-29-00679 https://rscf.ru/project/25-29-00679/ (Development of a complex acoustic and coherent-optical analyzer of morphofunctional characteristics of non-stationary dispersed systems and porous media for monitoring the synthesis and functionalization of materials).

Conflict of interest

The authors declare that they have no conflict of interest.

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Translated by T.Zorina