4aPA5. The influence of a focused acoustic field on mass-transfer processes at a heterogeneous boundary

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The velocity of a Schlichting boundary flow generated by a focused acoustic field incident on a rigid boundary in a liquid is evaluated experimentally. The temperature increase in the acoustic boundary layer is also estimated using optical techniques. The velocity of the small-scale Schlichting flow is estimated indirectly based on the velocity of the concurrent Rayleigh flow using the particle image velocimetry (PIV) method. The velocity of the Schlichting flow achieved in the experiment is high enough to enhance significantly mass-transfer processes at a heterogeneous boundary. This is confirmed by the results of laboratory experiments on acoustic intensification of high-rate growth of salt monocrystals, which were carried out under strictly controlled conditions. The increase in the boundary-layer temperature is shown to be insufficient for a pronounced crystal-growth inhibition.
INTRODUCTION

Some of secondary acoustic effects can be successfully used for various practical applications. For instance, the generation of acoustic flows at a heterogeneous boundary – so-called Schlichting flows, which play a determining role in acoustic field intensification of heat-mass exchange at this boundary [1].

A critical parameter determining the intensifying action of a Schlichting flow on the heat-mass exchange processes is its velocity. In order to develop intense boundary flows, the falling acoustic field should be substantially nonuniform; i.e., a certain transverse gradient of the oscillatory velocity should exist, directed along the heterogeneous boundary. As well, there should exist the possibility of controlling heat-mass exchange processes at this boundary, changing the spatial distribution of the velocity in near-boundary flows. This is achieved via a controlled change in intensity, frequency, and the spatial structure of the acoustic field falling on the boundary.

A theory related to describing near-boundary hydrodynamic flows was elaborated in detail and quite complex [2, 3]. As to flows generated by the acoustic field, their description is insufficient [1, 4]. Usually, the steady velocity of the Schlichting flow is estimated by the expression \( \nu = V M_k \delta \), justified under the condition \( M_k \delta \ll 1 \), where \( M_k = V/c \) is the acoustic Mach number, \( V \) is the oscillatory velocity in an acoustic wave, \( k \) is the wave vector modulus, and \( \delta \) is the boundary layer thickness, \( \delta \approx \nu (\omega/c)^2 \). For a Rayleigh flow, the velocity of Schlichting flows at a rigid boundary has been experimentally estimated. The interface was located in the zone of Fresnel diffraction of the field created by a piston source at frequencies on the order of 1 MHz. It was shown that the velocity of near-boundary flows for acoustic field intensities at the initial aperture of the radiator on the order several W/cm² were fractions of mm/s. Similar values of the tangential velocity close to the boundary are easily attained by hydrodynamic means, and at such Schlichting flow velocities, acoustic means of accelerating mass transfer at a heterogeneous boundary cannot compete with hydrodynamic ones. Obviously, for practical application of acoustic methods in processes of intensifying heat-mass exchange, it is necessary to increase the transverse gradients of the oscillatory velocity of the field falling normally on the solid phase interface. The use of an intense focused ultrasonic field yielding the maximum possible transverse gradients of the oscillatory velocity seems quite promising. The aim of this study is to demonstrate the possibilities of a focused acoustic field for creating at a heterogeneous boundary Schlichting flows with velocities that are sufficient for effective intensification of mass-exchange processes at this boundary, in particular, processes of controlled growth of salt monocrystals.

ESTIMATION OF SCHLICHTING FLOW VELOCITY

The idea of how to estimate the velocity of so small-scale a flow as a Schlichting flow is quite simple. As an acoustic field propagates in a viscous liquid, when there is a solid boundary in this liquid, three types of flows can form: Schlichting, the Rayleigh flow cocurrent with it, and a large-scale Eckart flow. Such flows are differentiated by the mechanisms of their appearance and, importantly for the experimental situation considered in this study, the time of stabilization. For a Schlichting flow, the time of stabilization is hundreds of microseconds – units of milliseconds, for a Rayleigh flow, it is tens of milliseconds – hundreds of milliseconds, and an Eckart flow enters a steady-state regime of tens of seconds. The time of stabilization depends on transverse inhomogeneity of acoustic field. If it is possible to fix the Rayleigh flow which is the cocurrent for Schlichting flow, then it is possible with confidence to state that the velocity of the Schlichting flow is at least no less than the velocity of the Rayleigh flow.

To record the field of flow velocities arising in the liquid under ultrasound, the Particle Image Velocimetry (PIV) method was applied [7]. The experimental setup [8] at which measurements were conducted is depicted schematically in FIGURE 1.

To visualize flows into distilled water filling an acoustic bath (1), white-colored particles of submicron size were introduced as markers; they were prepared from polyvinyl acetate latex. Using laser system (2) consisting of a 200 mW green Nd:Yag laser with a cylindrical lens, the plane containing the acoustic axis of concentrator (3) was illuminated. As a solid boundary, glass plate (4) was used, which was set up parallel to the focal plane and shifted from it toward the concentrator at relatively small distances (~ 2-3 mm). A flow visualized by laser radiation scattered on the tracer particles was filmed from above by a computer-controlled Videoscan-415-USB camera (5). The size of the filming area was 30x20 mm². For the purposes of the experiment, piezoceramic concentrator (3) was prepared with a working frequency of 1.5 MHz, a focal distance of 50 mm, and a full aperture angle of 53°.
The process of measuring the flow velocity was as follows: after switch-on of optical illuminating system (2) and onset of camera recording (5), a continuous acoustic field was excited. The acoustic field amplitude in the focal area was kept at a level of $5 \times 10^5$ Pa, and the video-recording rate was 25 fps. The video image was processed on a computer using standard PIV algorithms. As well, we obtained two-dimensional fields of flow velocity, in lattice points, created by various moments in time. The relative measurement error of the velocity at a fixed point was 5%; the spatial resolution (grid spacing) was about 1 mm.

Analysis of the video recording has shown that after switch-on of sound immediately from the location of the irradiated plate where the focal area of the concentrator was positioned, a narrow jet arises moving along the acoustic axis toward the concentrator. The time of appearance and stabilization of the flow velocity in immediate proximity to the surface of the plate did not exceed the interframe interval: $4 \times 10^{-2}$ s. Obviously, this is an average-scale Rayleigh flow, the presence of which unambiguously testifies to the appearance of Schlichting boundary microflows. The velocity field corresponding to arising Rayleigh flow is shown in FIGURE 2; the time of switching on the ultrasound is 0.4 s.

Under the conditions of this experiment, after approximately 10 s, a slower but larger-scale counter-directional flow begins to develop; i.e., the picture of the formation and competition of Rayleigh and Eckart flows is observed. FIGURE 3 shows the dynamics of Eckart flow in the situation without solid boundary, all other experimental conditions being equal.
The video image of the films was studied numerically with algorithms based on frame-by-frame cross-correlation processing of video signals. The velocity of the Schlichting boundary flow can be estimated by direct measurement of the velocity of the fully established cocurrent Rayleigh flow. This proved to be no less than 4 mm/s. This exceeds the estimate of the Schlichting flow velocity by approximately an order, which can be done based on examining the situation in [1].

Analysis linear field of acoustic concentrator shows that the characteristic transverse scale of the changing of the velocity of intense Schlichting flows in fact corresponds to the diameter of the focal spot (~3 mm) and the velocity of these flows is already sufficient for significant intensification of mass-exchange processes at a heterogeneous boundary [9]. The results of this experiment have shown that using a focused field under conditions ensuring a sufficient transverse gradient of the field and its sufficient intensity, it is possible to solve problems of local action on the mass-exchange process at the boundary and of spatial control of this mass exchange, for instance, by scanning with the focal area over the target surface.

**ACOUSTIC INTENSIFICATION OF THE CRYSTAL GROWTH PROCESS**

The necessity of selective and rigidly controlled action by the acoustic field arises in attempting to create methods for acoustically intensifying the rapid growth of salt monocrystals [10].

**Experimental Setup**

Experiments on acoustic intensification of rapid growth of salt monocrystals have been conducted on a laboratory setup, represented schematically in FIGURE 4 [10].

The main experimental setup was a hermetic acoustic bath (1) from a transparent plastic, which was simultaneously a liquid ultra-thermostat with a temperature regulation accuracy of up to 0.5°C. A focused ultrasonic source (2) was built into one of the side walls of the bath—a piezoceramic spherical concentrator analogous to the one described in FIGURE 1.

![FIGURE 4. Experimental setup.](image)

At the center of the bath was a cylindrical quartz cavity (3) with the studied crystal and an attached block of technological equipment. The axis of symmetry of the cavity coincided with the acoustic axis of the spherical concentrator and the optical axis of the recording system of the growth rate of the crystal surface (described below). The ends of the cavity were transparent to sound and light and made of a nonbirefringent material. The studied crystal surface (4) was the (100) face of the monocrystal potassium dihydrogen phosphate (KDP). In the setup, a technique of exact (to 0.2 mm) positioning of the studied crystal over three coordinates was implemented. In the cavity there was a KDP solution; an NaCl solution of the same acoustical impedance as the KDP solution was the immersion medium filling the bath. Along with acoustic action, hydro-mechanical action on the crystal was provided for, for which the cavity was equipped with a propeller stirrer (8). The temperature of the solution in the
cavity was fixed by a temperature sensor (9). The optical transparency of the route necessary for the system recording the rate of crystal growth is ensured by optical inset (5) in radiator (2). This is necessary for measuring in real time the growth rate (dissolution) of the studied crystal surface by nonlinear-optical means. In the system for accurately (0.1 μm/min) determining the growth rate, the birefringence capability of the KDP crystal was used [11]. The beam of a He–Ne laser (6) passed through the system, illuminating the crystal, and its output intensity, recorded by photodetector (7), is a periodic (16 μm) function of the crystal thickness. In experiments in real time, signals of the optical system and thermal sensor were digitized. The recording gives the time and temperature dependences of the growth rate. Analysis of records makes it possible to evaluate the efficiency of hydromechanical and acoustic action on the growing surface of the crystal under other equal conditions.

**Temperature Influence on Intensification Process**

The crystal growth can be hindered (in contrast to dissolution) by heating the medium in the focal region of the concentrator. Therefore, experiments on measurements of the temperature of the boundary layer of the fluid phase, with the crystal surface exposed to ultrasound, were performed. For that growth and dissolution of crystals without and with acoustic field were observed upon programmable cooling of the solution. Figure 5 shows a characteristic record. Regions A and B correspond to the processes of crystal dissolution and growth, respectively. Point 3, at which the growth rate is zero, corresponds to the saturation temperature. The difference between the true (without ultrasound irradiation) and effective (with ultrasound) saturation temperatures determines the temperature jump in the boundary layer, caused by the action of the acoustic field.

**FIGURE 5.** Record for the experiment on temperature measurement in the boundary layer.

![Figure 5](image)

**FIGURE 6.** Dependence of the boundary layer heating on the acoustic pressure amplitude.

![Figure 6](image)
The obtained dependence of the change in the temperature of the boundary layer on the field amplitude is shown in Fig. 6. It has a parabolic shape, which is in agreement with the theoretical concepts [1]. These results are in good agreement with the known measurement data [12]. Even at the maximum attainable ultrasound intensity (4–5 W/cm²), the measured increase in temperature at the center of the focal spot does not exceed 2 °C. Hence, at the acoustic field intensities used in our experiments, the increase in the temperature of the boundary layer is insufficient for significant retardation of the crystal growth.

Experimental Results

In experiments, the size of the crystal face corresponded to the scale of intense Schlichting flows. In principle, the action of Schlichting flows can be extended to larger scales, applying mechanical scanning over the focal area.

The (100) face of the KDP monocrystal with an initial size no larger than 1.5x1.5 mm was regenerated in solution with a saturation temperature of approximately 45°C; it grew under overcooling of approximately 5°C until its size attained ~2x2 mm. Samples prepared in this way were normally insonified along the acoustic axis in the focal plane of the spherical concentrator.

Figure 7 shows a record of monocrystal growth. Time intervals of field action are highlighted, and the relative amplitude of action is shown (the field of flow velocities corresponding to the given acoustic-hydrodynamic situation is shown in Fig. 2). The maximum amplitude of acoustic action \( P_{\text{max}} \) was chosen from the conditions of noncritical heating of the growing surface of the crystal.

As is seen from Fig. 7, at a sound pressure amplitude of \( \sim 0.5P_{\text{max}} \) growth acceleration of the observed face begins and at \( P \sim 0.75P_{\text{max}} \) a growth rate is established that is approximately two times faster than the characteristic rate for conditions of free convection; i.e., a nearly kinetic growth regime is established. For the given crystal size in passing to the kinetic regime, an increase in crystal growth by a factor of 2–2.5 is characteristic in comparison to its value under conditions of free convection [13]. We were unable to measure the growth rate at the maximum amplitude of action, since operation of the optical measuring system was violated by intense ultrasound.

Under usual hydrodynamic action on the crystal phase to ensure the kinetic regime in which the rate of crystal growth is extreme, a flow velocity of \( \sim 10 \) cm/s is necessary [9]. Thus, the result of those experiments show that the intensifying action of Schlichting boundary microflows induced by focused ultrasound are equivalent to the action of an external hydrodynamic flow with a velocity on the order of the oscillatory velocity in an acoustic field. Under the conditions of our experiment, the oscillatory velocity at a focus of \( \sim 25 \) cm/s corresponded to pressure \( P_{\text{max}} \).
CONCLUSIONS

The results of the experimental investigation reported in this paper show a fundamental possibility of controlling crystal growth by varying the parameters of an inhomogeneous acoustic field. The interphase mass-exchange is enhanced via formation of boundary and external flows, whose scale corresponds to the size of the crystal face exposed to ultrasound. The importance of this experimental fact is that all estimations made within the existing theoretical models [1, 4, 14] indicate impossibility of such an effect with a positive result.

The results obtained determine the line of further promising investigations. It is planned to analyze in detail the scale, dynamic, mass-exchange, and thermal characteristics of the boundary microflows caused by an inhomogeneous acoustic field and cocurrent medium-scale flows in experiments on model systems with application of the developed visualization tools, measurement of flow velocities, and construction of the velocity field. The measured values of the temporal and spatial scales will be used to modify the existing theoretical models in order to predict optimal parameters of hydro-acoustic action on crystal growth in practical applications.

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