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**ELECTRICAL PROCESSES  
IN ENGINEERING AND CHEMISTRY**

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## **Electrokinetic Effects under the Action of Ultrasound in Liquid Media**

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**Abstract**—The electrokinetic phenomena under the influence of ultrasound (US) on aqueous solutions and melts of fusible metals are experimentally investigated. It is established that the combined action of the ultrasonic and electric fields on the liquid media intensifies the processes of diffusion and dissolution of substances. This allows one to accelerate the processes of ultrasonic clearing in media of polar liquids and to raise the durability of the soldered connections.

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### INTRODUCTION

The action of powerful ultrasound (US) induces well-known effects in liquid media: the acoustic cavitation [1] associated with pulsations and the collapse of cavitation bubbles, the ultrasonic capillary effect [2], the sonoluminescence [3], the vortex effects as micro- and macrostreams [4], the enhancement of diffusion processes [5], etc. When the intensity of the ultrasound exceeds  $10^5$  W/m<sup>2</sup>, the kinetic energy of the collapsing bubbles concentrated in a negligibly small volume transforms partly into force pulses and partly into heat energy. In liquid media under the action of ultrasonic vibrations, electric phenomena are also induced in the form of short pulses due to the negative electric charges of cavitation bubbles [3] and due to the electrokinetic phenomena related to the directed motion of the charged particles.

The electrokinetic phenomena are observed in disperse systems and capillaries. They manifest themselves in the form of the relative motion of one phase with respect to another phase under the action of an external electric field (electroosmosis, electrophoresis) or as the appearance of a potential difference in the direction of the relative motion of the phases owing to the mechanic forces (the Dorn effect, the flow potential).

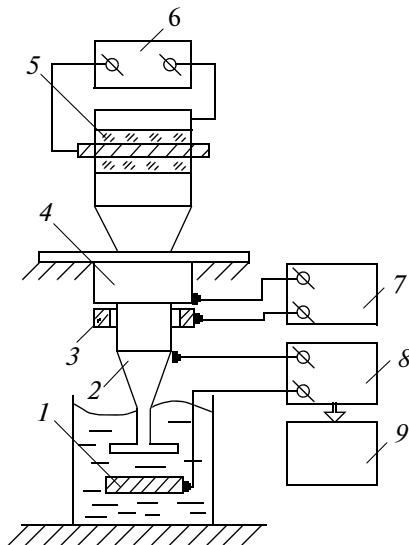
The electrokinetic phenomena in liquid media appear also owing to the increasing of the mobility of the charge carriers and the appearance of their directed mass transfer directed from the radiator under the action of the pressure of the ultrasonic wave. When cavitation bubbles collapse, the formation of additional carries—negatively charged cavitation bubbles—also taking part in the mass transfer is possible.

### EXPERIMENTAL

Longitudinal ultrasound oscillations were excited using an immersed piston radiator with the aim to detect and investigate the electrokinetic phenomena of the second type. Ultrasonic oscillations with a frequency of 22–44 kHz were supplied to a piezoelectric transducer consisting of two piezoceramic rings with their diameter being 30 mm and two frequency lowering cover plates connected with thread studs. The amplitude was registered using a sensor located near the oscillating waveguide surface and connected to a vibrometer. The value of the electric current induced in the liquid owing to the directed motion of ions under the action of the ultrasonic field was registered using an F116/1 microammeter connected between the radiator and the stainless steel electrode located at the bottom of the bath with the liquid investigated (Fig. 1).

We also inserted a third gridlike electrode near the radiator to which a constant voltage of 400–500 mV was applied from an external source with the aim to check the hypothesis related to the influence of the polarity of the ions involved in the current formation.

A schematic view of the experiment on the combined action of ultrasonic and electric fields on liquid media at the treatment of metallic (*a*) and nonmetallic (*b*) materials is shown in Fig. 2. Sample (1) was immersed in the liquid at the distance *h* from the bottom of bath (2) and it was connected with a wire to one of the poles of external power source (3). The second pole of the source was connected with the bath. When electric oscillations were supplied from the US generator to transducer (4), mechanical oscillations were induced in it and transferred to the bath via diaphragm radiator (5). Under the action of the direct electric current, the hydrogen ions move in the direction from the radiator to the sample; the reduction of the ions



**Fig. 1.** Scheme of the experiment for the investigation of the electrokinetic effect in liquid media. (1) Electrode, (2) radiator, (3) amplitude sensor, (4) waveguide, (5) transducer, (6) US generator, (7) vibrometer, (8) current-sensing device, (9) automatic recorder.

and the formation of gas nuclei occur in the liquid, which facilitates the cavitation process.

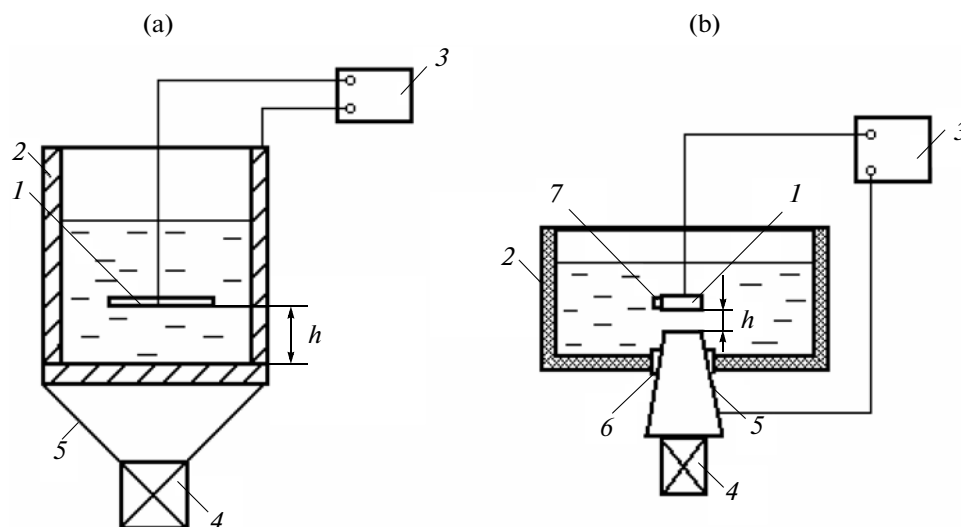
At the local introduction of the US oscillations in the bath, the sample (1) was located in the bath (2) at a distance  $h$  from the working end of the radiator. The nonmetallic bath (2) has a hole in its bottom where a concentrator type radiator is fixed using gasket (6). The contact ring (7) was connected to one of the poles of external source (3), and the other pole was connected to radiator (5). The mechanical oscillations induced in transducer (4) were amplified by the con-

centrator and transmitted to the liquid medium. This increased the intensity of the cavitation phenomena owing to the deposition of cavitation nuclei on the treated sample and the local introduction of US oscillations.

During the investigations of the electrokinetic phenomena of the first kind, owing to the combined action of the energy of the US and electric fields on the melts and the durability of the formed metal connections, the value of the direct current through the melt changed in the range of 0–15 A. It was registered using an M2020 device connected in an electric circuit with a shunt with an accuracy of 0.5 mA. The US oscillations were supplied to the melt over 20–30 s after the current was switched on with the aim to eliminate the measurement errors owing to the heating of the wires and ballast resistor.

## EXPERIMENTAL RESULTS AND DISCUSSION

The analysis of the data (see the table) shows that the most pronounced manifestation of the electrokinetic phenomenon of the second type (as the appearance of the potential difference and the increasing of the direct current value) was observed in the liquids with the least resistance and viscosity [6]. The time needed for the current to reach the equilibrium in a liquid is proportional to the liquid's viscosity. The maximal effect of the current increasing was registered for the interelectrode distance of 5 mm, since, in this case, the entire interelectrode zone is occupied by the cavitation bubbles. For less interelectrode distances, the current carries are partly pushed out from the zone owing to microstreams. When the distances exceed 5 mm, the electrokinetic effect in the liquids



**Fig. 2.** Scheme of the experiment for the investigation of the combined activation by the energy of US and electric fields.

## Characteristics of the electrokinetic effect in liquid media

Liquid medium	Resistance of the medium, $\Omega$ m	Viscosity, Pa s	Current variation $\Delta I$ , mA	Time needed to reach equilibrium, s
Mineral oil	$10^{13}$	3.2–3.3	0	—
Glycerin	$10^{12}$	1.5–1.6	0.06	70
Distilled water	$10^4$	0.32–0.33	0.6	50
Tap water	$10^2$	0.32–0.33	1.0	10
Ethanol	$10^4$	0.24–0.25	1.4	10
Organic acid	10	0.25–0.26	2.0	5
Water with Cl ions	0.3	0.32–0.33	3.4	3

becomes weaker, since the cavitation region does not occupy all the interelectrode zone.

The greatest potential increasing and the increasing of the direct current owing to this were observed in the liquids with the least electric resistance and viscosity. The time needed for the current to reach the equilibrium in a liquid is proportional to the liquid's viscosity (Fig. 3).

The maximal current increasing was registered for the interelectrode distance of 5 mm, since, in this case, the entire interelectrode zone is occupied with cavitation bubbles. For less interelectrode distances, the current carries are partly pushed out from the zone owing to microstreams. When the distances exceed 7 mm, the electrokinetic effect in liquids becomes weaker owing to the absorption of the US energy and the mass transfer weakening.

The appearance of the potential difference and the increasing of the current through the liquid can be explained owing to the fact that the US vibrations increase the mobility of the charge carriers in the liquid; a mass transfer appears directed from the radiator. When the cavitation bubbles collapse, additional carries, negatively charged cavitation bubbles, are generated. The time needed to reach the maximal current value depends on the amplitude of the US oscillations and the polarity of the molecules of the liquid. When the amplitude of the US oscillations does not exceed  $5 \mu\text{m}$ , the formed microstreams are insufficient to induce the necessary mobility of the ions and the process of the current increasing is extended in time. At the amplitude of  $7\text{--}8 \mu\text{m}$ , the time needed to reach the current maximum reduces and its value increases, since the maximal quantity of ions is involved in the mass transfer. When the amplitudes exceed  $10 \mu\text{m}$ , the current extremum can be reached in the minimum time. The current amplitude decreases owing to the dissipative action of the cavitation cloud on the directionality of the ion stream.

The character of the temporal variation of the current in the liquid medium also depends on the potential of the grid electrode between the radiator and the main electrode. The current value increased on aver-

age by three times at the positive potential at the grid electrode. At the negative potential, the current changed its direction and its amplitude decreased. This can be explained by the fact that the positive ions of the liquid medium are mainly involved in the charge transfer.

Investigations of the US action on weakly ionized liquids (liquid glass) showed that, when a potential is applied to the radiator, the current between the electrodes increases by 20%. The current maximum was reached during the time of 130 s. The amplitude of the current through the liquid increased by 1.6 times when zink microparticles, which acquire a positive charge in the process of the US activation, were introduced. When formic alcohol containing negative OH ions was used, the initial current level was 3 times lower, and the current's amplitude exhibited a more pronounced extremum. We could not observe the electrokinetic effect in metallic melts owing to their high conductivity and the lack of free current carriers. However, a current variation was registered.

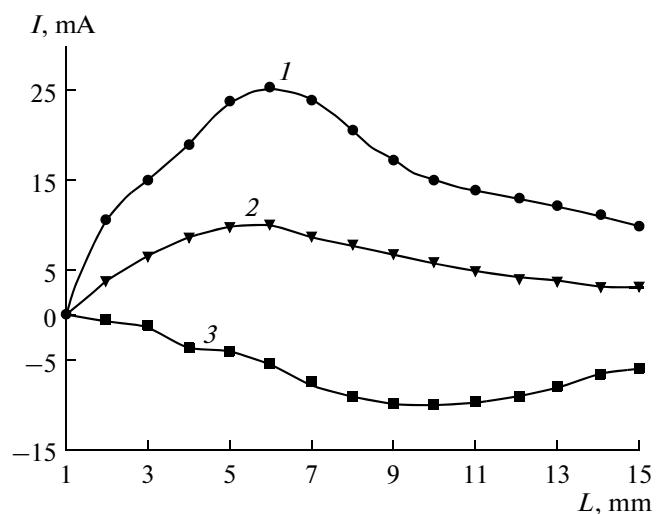


Fig. 3. Dependences of the current variation in the liquid in the US field versus the interelectrode distance. (1) Water with Cl ions, (2) organic acid, (3) distilled water.

Since charged particles with various concentrations and mobility are present in the liquid medium simultaneously, the total density of the flux of the particles in a of unit time amounts to

$$J_{el} = \sum_{i=1}^n B_i C_i F, \quad (1)$$

where  $B_i$  is the ion mobility,  $C_i$  is the concentration of the ions of the active reaction component in the melt, and  $F$  is the strength of the US field.

The current's value due to the electric field induced in the liquid medium is as follows:

$$I = \sum_{i=1}^n B_i C_i Q_i F S, \quad (2)$$

where  $Q_i$  is the ion charge, and  $S$  is the flux's cross section.

The appearance of an electric field in the liquid medium and the activating action of the US field influence the mass transfer processes. One should add to the diffusion flux owing to the gradient of the concentration of the diffusing substance  $J_d$  the flux of particles arising owing to the action of the pressure of the US field  $J_{US}$  and the flux of the ions under the action of the forces of the electric field  $J_{el}$ . Then, the total flux of the diffusing particles amounts to

$$\begin{aligned} J &= J_d + J_{US} + J_{el} \\ &= -D \frac{\partial C}{\partial x} + UC_1 \cos \beta + \left( \sum_{i=1}^n B_i C_i \right) F, \end{aligned} \quad (3)$$

where  $C_1$  is the concentration of the mobile particles,  $U$  is the velocity of the particles in the US field,  $\beta$  is the angle between the forces of the US field and the diffusion flux, and  $\alpha$  is the angle between the vectors of the electric field and the diffusion flux.

The force of the US field, which acts on the flux of the particles with the cross section  $S$ , is

$$F = \rho c \omega A S. \quad (4)$$

Substituting Eq. (4) in (3), we obtain

$$J = -D \frac{\partial C}{\partial x} + \nu \pi c \omega A S C_1 \cos \beta. \quad (5)$$

The combined action of the US and electric fields when the directions of their vectors coincide intensifies the diffusion processes of the active reaction components in the liquid medium. However, since the intensity of the electric field induced in the liquid medium is low, the electric transfer currents are insignificant and do not exert considerable activation action on the mass transfer processes.

With the aim to increase the intensifying factors and the intensity of the cavitation processes in the liq-

uid media, it was proposed to pass using an external power source an electric current with a density of 10–100 A/m<sup>2</sup> from the US radiator to the workpiece or to a contact element on the workpiece [7]. In a polar liquid under the action of a direct electric current, the mass transfer of ions increases. The reduction occurs of the hydrogen ions in the form of gas bubbles with dimensions of 50–100  $\mu\text{m}$ , which are the cavitation nuclei. The continuous accumulation of the cavitation nuclei in the working zone and their collapse under the action of the US oscillations increase the intensity of the cavitation processes. This facilitates the process of the destruction of grease films on the workpiece's surface and increases the cleaning quality owing to the more uniform distribution of the cavitation nuclei over the entire surface under the action of the current force lines.

With the aim to increase the locality and productivity, the electric field is oriented so that the hydrogen ions move to the radiator and reduce in the form of gas bubbles. When the amplitudes of the oscillations are considerable, near the radiator's surface, a rapid growth of the bubbles to the critical dimensions and their collapse occur; this is accompanied by intensive cavitation effects. The value of the current passing through the liquid depends on the liquid's polarity and the dimensions of the treated workpiece. The greatest increasing of the cavitation pressure by a factor of 2–2.5 measured using a cavitometer was registered at the optimal current density of 10–100 A/m<sup>2</sup>.

Under the combined action of the US and electric fields on melts, the appearance of the electrodiffusion process depends on the correlation of the forces, which act on the thermally excited metal ion, both in the direction opposite to the electric flux (the action of the external field) or in the direction of the electron flux (electron wind). The influence of the electric field is small owing to the shielding influence of the electrons; therefore, the force of the electron wind prevails; it increases with the current density increasing. Consequently, there is a greater probability for the excited ion to transform into a vacancy when it moves in the direction of the electrons than when it moves in the opposite direction. Therefore, the vacancies move to the negative pole, and the metal ions move to the positive pole; this increases the width of the diffusion zone.

When the current densities exceed 1–1.5 A/mm<sup>2</sup>, the directional diffusion of the solder components or the diffusion of the connection material into the solder owing to the electromigration of the particles of easily diffusive metals increases the width of the diffusion zone and the durability of the connections [8].

When the currents are lower than 10 A, the short range order of the particles in the melt becomes distorted owing to the microstreams and cavitation effects. This increases the resistance of the melt and, respectively, decreases the current through it by 50–80 mA (Fig. 4). When the current exceeds 10 A, a consider-

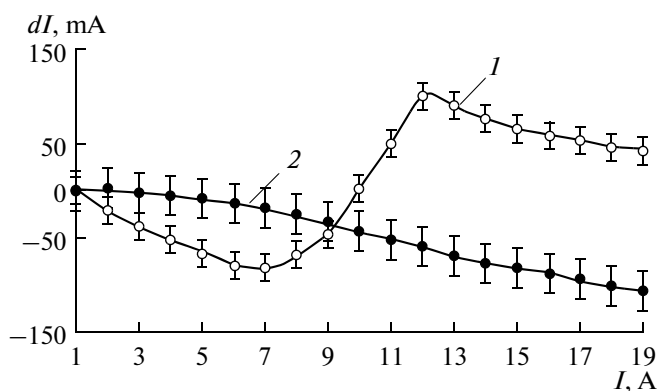


Fig. 4. Dependences of the current variation in the melt under the US activation versus the current value for various amplitudes of oscillations  $A$ ,  $\mu\text{m}$ : (1) 5 and (2) 15.

able mass transfer of charge carriers and their ordered motion between the electrodes arise. This increased the current through the melt to 100 mA at the amplitude of 15  $\mu\text{m}$ . When the current exceeded 12 A, the solder intensively heated; this increased its resistance and, consequently, the current variation through the melt decreased.

The durability of the connections of the POS 61 solder with the aluminum samples versus the direct current in the direction from the radiator to the work-piece (frequency of 22 kHz, amplitude 10  $\mu\text{m}$ , temperature 240°C, and time 10 s) increases for the currents exceeding 10 A and decreases when the current exceeds 15 A [9].

The electrical stimulation of the diffusion of the solder components into the material of the workpiece, which can be associated with the electrotransfer in the melted solder at high current densities [10], is a probable cause that the durability increases.

The passing of the current through the zone of interaction in the direction from the solder to the base increases the durability of the connection by 1.5–1.8 times, since the width of the diffusion zone increases owing to the electromigration of aluminum to the solder. The width of the diffusion zone registered using a scanning electron microscope in this case amounted to about 4  $\mu\text{m}$  for the POTs 10 and POS 61 solders.

## CONCLUSIONS

The action of an intensive US field on polar liquid media induces electrokinetic phenomena of the second

type in them owing to the increasing of the mobility of the charge carriers and the appearance of their directed mass transfer from the radiator under the action of the pressure of the US wave. The combined action of the US and electric fields on the liquid media owing to the action of the electrokinetic effects of the first type and the directed motion of the cavitation nuclei creates conditions for the intensification of the diffusion processes and dissolution and emulgation in the treatment zone. This allows one to facilitate the processes of the US cleaning of the surfaces of work-pieces in polar liquid media. The combined action of the US and electric fields provide the possibility to increase the durability of soldered connections owing to the enhancement of the diffusion processes at the melt–solid body interface.

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