03.1;03.5 Dynamics of liquid mass transfer in a water bridge

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> The paper presents experimental results of the dynamics of mass transfer through a water bridge arising between two dielectric cups with distilled water under the action of a constant high voltage between cylindrical electrodes. It is established that, depending on the ratio of the electrode diameters, the total fluid flow in the water bridge can be directed both to the cathode and to the anode. It is shown that the inversion of the direction of mass transfer of liquid through the bridge is due to the redistribution of volume charges.

Keywords: water bridge, EHD flow, anode, cathode.

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The water bridge (WB) arises between two dielectric cups with distilled water under the action of a constant high voltage applied between the electrodes submerged into the liquid [1-7]. The studies performed revealed a number of WB peculiar features [1-3]; one of the main features is the liquid transport through the bridge. It was established that, after the WB formation, the total liquid flow is first directed mainly from the anode to cathode. After a time period of several minutes to several tens of minutes, the flow reverses its direction; further the systematic repetition of this reverse is observed during the entire WB lifetime. This repetitive flow reverse is a steady feature of the bridge; as assumed in [3,4], it is caused by the hydrostatic pressure induced by the difference in water levels in the cups. For the WB formation and stable existence, several models were suggested in [5]; however, besides [5], only one theoretical study concerning the flow inversion mechanism is known [6].

This paper presents the results of experimental study of the liquid flow direction, which show that, beginning from the moment of the WB formation, the water mass transfer through the bridge may proceed either in the anode-to-cathode or cathode-to-anode direction depending on the electrode diameter ratio. It is also shown that the liquid flow inversion may be caused by the volume charge redistribution in the cups with distilled water.

The experiments were conducted at the setup whose layout is presented in Fig. 1, *a*. Distilled water 60 g in weight was poured into Petrie dishes one of which was set on the electronic balance 0.01 g in accuracy, while the other was set of the variable-height Plexiglas support. Especial attention was paid to the purity and surface condition of electrodes for which copper wires of different diameters (0.1-6.0 mm) with insulated ends were used (Fig. 1, *b*). In 5 s after the water-filled dishes got into contact and voltage of 15 kV was applied to the electrodes, the dishes were drawn $10 \pm 0.5 \text{ mm}$ apart each other, which provided identical initial conditions for the formed WB about 2 mm in

diameter. The electric current was kept constant and equal to $0.2 \,\text{mA}$.

Fig. 2 presents time dependences of variations in the water mass in the cathode dish at different anode (d_a) and cathode (d_c) diameters. Values $\Delta m > 0$ correspond to the anode-to-cathode flow (forward direction), while $\Delta m < 0$ is for the cathode-to-anode flow (backward direction). One can see that, from the moment of the WB formation, the liquid flow through the bridge is first (during up to about 6 min) directed from the smaller-diameter electrode to that with larger diameter independently of their polarity; when the diameters are equal (a symmetric system), the mass is transferred in the forward direction (curve 1). Emphasize that all the works we have found about studying the dynamics of liquid flows through WB, including [1-5], demonstrate the predominance of the anode-to-cathode mass transfer. To our opinion, this is caused by predominantly using in those experiments plate electrodes whose sizes are comparable with the inter-electrode distance. Therefore, despite the presence of corners and formation of various defects on the electrode surfaces during long-term operation with applying forward and backward voltages, they may be regarded as a symmetric electrode system. This is why in those studies the liquid flow through WB is directed forward, which corresponds to curve 1 in Fig. 2. In our experiments, proper selection of different-diameter cylindrical electrodes with insulated ends allowed obtaining a variable-direction liquid flow through the bridge from the moment of its formation.

WB is a particular case of an electrohydrodynamic (EHD) system where the dielectric liquid mass transfer is caused by the action of Coulomb forces on volume charges [3] arising due to spatial redistribution of charges during the electric current passage [8–11]. As known [8–10], the main mechanisms of charge formation in liquid dielectrics under the influence of high—intensity electric field (of about 10^6 V/m) is ion injection from both the cathode and anode and



Figure 1. a — Layout of the experimental setup: I — Petrie dishes, 2 — electronic balance, 3 —variable—height Plexiglas support, 4 — electrodes, 5 —WB, 6 — glass support, 7 — lighting. b — examples of electrodes.



Figure 2. Time dependence of water mass variation in the cathode dish at different electrode diameters (in mm): $1 - d_a = d_c = 1.8$; $2 - d_a = 1.4$ and $d_c = 1.8$; $3 - d_a = 1.8$ and $d_c = 1.4$; $4 - d_a = 1.8$ and $d_c = 6$; $5 - d_a = 6$ and $d_c = 1.8$.

formation of non-equilibrium dissociation-recombination charge layers. Notice that the EHD pumps of the injection and dissociation type were developed based on these two effects, respectively [8].

To reveal the basic mechanism of the volume charge formation under the condition of a steady-state WB existence, we have performed the following experiments. Dishes with water were set on aluminum foil plates 2×2 cm in size, to which voltage was applied. Thus, the electrodes did not directly contact the liquid. At voltages of 25-30 kV, we managed to "extend" the formed bridge only to 5 mm at the diameter not exceeding 1 mm, while the liquid mass flow rate through the bridge was by more than an order of magnitude lower than in the experiments with cylindrical electrodes. This confirms that in the case of WB with electrodes submerged in liquid the main role is played by the injection mechanism of charge formation. These ions of the same polarity as the electrodes are "frozen " in the liquid [10] and create via Coulomb forces the liquid counterflows directed from the respective electrodes [9–11]. Thus, an EHD flow through WB arises, whose outer layer rotates with the tangential velocity of about 0.3 m/s, while velocities of the near-axial regions range within ± 0.2 m/s depending on the total mass transfer direction (Fig. 3). This process may be regarded as complicated by the presence of the water bridge that is a "bottleneck" of the classical variant of the EHD flow in which counterflows occur even if the electrode sizes are equal [9,10].

Generally, the source of fluid-induced violation of electrical neutrality caused by the emergence of excess of charges of one type over the other is the asymmetry of the electric field distribution between the electrodes, charge carrier mobilities and electrochemical reaction rates [9,10]. Therefore, in the symmetric electrode system the total liquid flow through the bridge is directed forward from the moment of the WB formation due to a higher mobility of positive ions as compared with that of negative ions, which complies with experimental results. If the electrode system is asymmetric, the unipolar injection from the smaller-diameter electrode dominates because the electric field at its surface is stronger. This is why the liquid flow is directed towards the larger-diameter electrode just after the WB formation regardless of the inter-electrode voltage polarity.

It is necessary to notice the direct influence on the EHD flow dynamics from the condition of the considered electrochemical system whose disequilibrium degree changes during the electric current passage. For instance, in the electrically neutral anode volume (pH \approx 7), the liquid is gradually depleting of OH⁻ ions during oxidation at



Figure 3. Visualization of the liquid flow through WB by the tracer-particle technique with the frame frequency of 1 kHz [3].

the anode $4OH^- - 4e = 2H_2O + O_2$. In the course of time, the decrease in the number of these ions initiates another oxidation process $(2H_2O + 4e = 4H^+ + O_2)$ that results in the anolyte enrichment with H^+ ions. These two reactions lead to the establishment in the anode volume of a new ion equilibrium at pH < 7. In the cathode volume of liquid with the initial pH index, negative ions (pH > 7) begin dominating first due to the hydrogen ion reduction $(2H^+ + 2e = H_2)$ and then due to activation of another reducing reaction $(2H_2O + 2e = 2OH^- + H_2)$ caused by the decrease in proton concentration. However, the difference in the rates of electrochemical reactions in the anode and cathode directions makes the charge redistribution in the EHD system somewhat more complex due to the following factor.

Current carriers in the aqueous fluid are hydroxonium ions $\rm H_3O^+,$ protons $\rm H^+$ and hydroxyl ions OH- formed by water dissociation according to scheme $H_2O + H_2O = H_3O^+ + OH^-$ with the proton transfer from one molecule to another [12]. For instance, in work [13] the study of the WB conductivity by the method of quasielastic neutron scattering revealed two proton groups one of which is a component strongly coupled with oxygen atoms (H_3O^+) and the other is mobile (H⁺). Therefore, the positive charge transport in the aqueous fluid is performed mainly by protons because of their anomalously high mobility [12], while hydroxonium ions allow for the liquid mass transfer. In the case of the forward flow, the proton departure from the anode cup is to some extent compensated by their formation due to water molecules oxidation at the anode. In the catholyte, fast drift of protons towards the cathode with their subsequent reduction at the cathode surface promotes creation of a negative charge in addition to the injected ones. Therefore, during the current passage, the backward water flow begins dominating over the forward one when a certain negative volume charge is achieved in the system cathodic part: the inversion of the liquid flow through WB takes place (curve 4 in Fig. 2). The inversion at the backward flow direction caused from the very beginning of the bridge formation by the dominating unipolar injection of hydroxide ions from the cathode (curve 3 in Fig. 2) may be explained in a similar manner by analyzing the above-mentioned oxidation-reduction reactions with taking into account the anomalously high proton mobility.

Notice that the observed hysteresis and negative differential resistance in the WB I-V characteristic are also explained by the authors of [7] by redistribution of the volume charges. In this work, the flow retained its predominant anode-to-cathode direction with decreasing voltage, but, just when the I-V characteristic got to the section of negative differential resistance, the flow direction changed abruptly to the opposite one, which is equivalent to activation of an additional EHD source at decreasing external voltage. The effect of the "charge plug" formation in a classical EHD device, which is to some extent similar to the flow direction inversion observed in WB, is also caused by volume charges [10].

Thus, it has been experimentally confirmed that, just from the moment of the WB formation, the total liquid flow through the bridge is directed from the smaller-diameter electrode to the larger-diameter electrode independently of their polarity; in the symmetric electrode system, the mass transfer proceeds in the forward direction. The observed reverse of the liquid flow through WB is caused by periodic formation of excess volume homocharge at one of the electrodes depending on the initial flow direction with its subsequent transport through WB towards the opposite electrode.

Conflict of interests

The authors declare that they have no conflict of interests.

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