Production of long-lived heterogeneous structures using plasma

V I Abakumov¹, V L Bychkov¹*, A R Bkmukhametova¹, V A Chernikov¹, G V Golubkov², D A Safronenkov³ and S V Zaitsev¹
¹Lomonosov MSU, Faculty of Physics, Leninskie Gory, Moscow 119991, Russia
²Semenov ICP RAS, Moscow, Kosygin str. 4, 119991, Russia

*bychvl@gmail.com

Abstract Experiments in the air were carried out using capillary and corona discharges in order to obtain long-lived heterogeneous structures up to 2 mm and greater. Electron microscopy photo of one of them obtained in the capillary discharge included a core of a vaporous substance and a quasi-solid shell. Carried out experiments with corona discharges over tap and distilled water, alcohol, glycerin and their mixtures have shown appearance of liquid jets and columns which can be caused by different hydrodynamic instabilities. In complex liquids there is a possibility of creation of structures with long lifetime.

1. Introduction
Investigation of large (up to 2 cm and greater in diameter) luminescent heterogeneous structures, the so-called long-lived heterogeneous structures (LLGS), is of significant interest for low-temperature plasma physics, both in terms of determining conditions and thermodynamics of such structures created in non-equilibrium conditions [1-4]. They represent interest from a point of view of their possible applications as combustion activators in plasma aerodynamics and plasma assisted combustion at injection of LLGS created in plasma into fuel mixtures [5], or creation of corona discharges in combustors for creation of small particles of a fuel in result of developed instabilities and applied as combustion activators.

Already during the last twenty years, studies have been carried out [4,6-8] showing that in a plasma created by erosive plasma generators, large enough long-lived luminous formations with a diameter of up to 8 millimeters and a lifetime of up to 6 seconds appear when plasma is exposed to samples of metals and polymeric materials. These formations have a complex composition and differ in structure from the drops of molten metal. A sequential study of the multiple production of such formations and their structure has not been carried out yet, therefore, studies of the possibility of formation of a flow of such objects are of considerable interest.

Because of possible applications in plasma aerodynamics present investigations were also devoted to formulation and undertaking of experiments with corona discharges realized over surfaces of different liquids. Investigations when a surface of one of the electrodes is covered by a liquid are of interest for hydrocarbon fuel activation, search of undesirable liquids elimination in engines, problems of ecology and disinfection [9]. They are of a fundamental interest, since the corona discharge allows to obtain new information on phenomena connected with development of electro-hydrodynamic instabilities in new, earlier unrealized conditions [10-12]. Such investigations are developing now, that is a reason for undertaking of these ones. In this work we were concentrated on electric-hydrodynamic features of corona discharges which are practically uninvestigated yet. However, there is limited...
information about any discharges over surfaces of liquids [9] and it is obtained mainly for electrolytes or water. From the application point of view it is interesting to know how a surface of a liquid fuel acts under an impact of gas discharges. Therefore our work is devoted to investigations of electric-hydrodynamic (EHD) effects caused by corona discharges over liquid. They are continuation of our experiments [10-12].

For investigations, we used two types of plasma generators - the capillary discharge [1-4], which usually is applied for interaction of high temperature plasma in air with a metallic or a dielectric surface [1-4].

Another type of generators is a corona discharge over liquids [10-12]. Interaction of plasma with a surface of a liquid leads to creation of charged structures with a long lifetime over their surface. The purpose of this paper is a continuation of previous investigation for creation of long-lived heterogeneous structures in plasma.

2. Experiments on generation of two-phase objects

Experiments on formation of heterogeneous luminescent formations - LLGS with a thin shell under action of an erosive plasma generator plasma jet on various materials, including pure metals, metallic alloys and metals covered with a film of alcohol or glycerin, were performed. Under heterogeneous or two phase objects we mean the objects consisting of vapor and a shell (cover) of quasi-liquid or quasi-solid material. In plasma experiments such objects can have additional plasma cover and in this case they represent three-phase objects.

A scheme of the experimental device is shown in Figure 1. It is the same as we used in [3,4,8]. It represents a capillary plasma generator with plasma forming dielectric plate between two electrodes, which works in pulse modes with the pulse time ~10 ms. The energy input during the pulse is about 200 J.

![Figure 1](image)

**Figure 1** Capillary plasma generator: 1,4 - electrodes, 2 - dielectric plate with a capillary (discharge chamber), 3 – a base of the plasma generator of Plexiglas.

In Figure 2 in the left photo one can see an appearance of the set up during generation of the erosion plasma. Experiments were carried out to obtain LLPF based on the alloy of tin and lead, pure tin, pure lead and pure copper using the erosion plasma generator with energy input of about 190 J during an action a time of ~ 10 ms. A lifetime of objects with a shell and a vapor nucleus reaches 4 seconds.

We found that when the plasma generator’s jet interacts with materials moistened by various liquids, LLPF number and size change. In particular, glycerol leads to increase the amount and size of LLPF, while alcohol, on the contrary, reduces these characteristics.

In Figure 2 (in the right photo) typical trajectories of LLPF obtained in experiments are presented. Figure 3 shows traces of the molten droplets and tracks of explosive LLPF. One can see in Figure 3 traces in the form of stars. Stars reflect explosion and destruction of LLPF. According to [4] it is the cover (shell) presence that allows to explain LLPF explosion when it falls on a substrate by an existence of a condensed vapor under the cover. Because of the objects fragility we studied various conditions and ways of LLPF shell conserving.

In Figure 2 one of the experimental setup arrangement variants is shown. The plasma generator capillary was directed upwards. We tried to conserve the undamaged shells by catching LLPF in various liquids (alcohol, glycerin, water).
Figure 2 Left photo: plasma generator: capillary directed upwards. Right photo: typical paths of LLPF.

Figure 3 LLPF tracks on the paper.

In Figure 4 in the left photo the LLPF (resulting cover with a ball core inside it) is presented. In Figure 5 in right photo one can see the shell and the "core" of LLPF (of the solder POC-61) dropped out of it.

Figure 4 Left photo: the envelope of the LLPF. Right photo: the shell and "core" of LLPF (of~600 μm diameter).

It was possible to keep the whole shells for a week, after which they disintegrated. Oil and glycerin were used to save the undamaged shells.
Investigations of spheres obtained in plasma

In these experiments, in order to obtain individual spheres for research, we used a syringe without a needle so that the LLPF should be preserved without damaging the shell. After a shot, plasma formations fell on a paper leaving traces in a form of tracks or stars, as shown in Figure 3. We did not succeed in retaining of LLPF in such experiments. Therefore, we conducted experiments, replacing paper for various liquids, namely: water, glycerin and alcohol.

Because of a high density of the liquid most of the LLPF was destroyed when it came into contact with the surface of glycerin, while contacting the alcohol surface, the LLPF was retained and drowned, but the most fragile formations were destroyed by interacting with the bottom of the cuvette. In the case of water, some samples were drowned, and some were broken when struck against the surface of the liquid. In all three cases, strong formations could be analyzed.

Predominantly water was the liquid into which the formations fell after the shot, because on the surface of it the greatest amount of LLPF with the shell was retained. From the surface of the water with a syringe without a needle, samples were carefully collected and placed in individual containers with water and oil. To continue research without contaminating the cuvette, and also to learn how the storage conditions affect the lifetime of the casing. As it turned out, in water the formations are stored on average 3 days, and in oil 5 days, individual samples can have shell lifetime up to 7 days in both cases.

After the destruction of the shell of the LLPF, the "core" fell out. The composition of the obtained spheres includes Al₂O₃, Pb, Sn. The presence of Sn and Pb in the analysis of the nuclei is explained by the fact that the sample on which the jet of the plasma generator acts, and this wire from the solder POC-61 consists of 59-61% of tin and 39-41% of lead.

![Figure 5](image)

Figure 5 Cores of the colored LLPF: red—to the left, blue—to the right.

Al₂O₃ is present in the analysis of samples, because of the sputtering from the electrode. The presence of carbon on one of the samples is explained by that during the shot a part of the nozzle, which is made of plexiglass (C₅O₂H₈)ₙ, is burnt. A photograph of two nuclei were obtained using an atomic force microscope, are shown in Figure 5. They differ in colors and sizes (because of different additions of small components). A photograph of LLPF shells were obtained using an atomic force microscope, see Figure 6. The thickness of the shell can be estimated as 5-10 microns.

Also, experiments were carried out in which we tried to preserve the whole shells by catching LLPF in various liquids (alcohol, glycerin, water). A spectral analysis of the resulting formations was carried out, and their linear dimensions, density, mass, and heat of evaporation were estimated.

The linear size of each "core" was about several hundred microns. For each of the formations, the volume was estimated. On average, the result was tenths of a cubic millimeter. With a help of spectral analysis, the content of tin, lead, and Al₂O₃ was determined. To determine the density, we used the obtained volume values, tabulated values of the density for tin, lead, and Al₂O₃, as well as the percentage of these metals and the oxide of the relative whole sample. On average, the density of "nuclei" was 5.13 g/cm³, and the average value of the mass of the nuclei was 1.7 mg.

An estimate was also made of the specific heat of vaporization of the obtained nuclear alloy by means of tabulated values of the specific heat of vaporization. The minimum values stored in each
"core" of the LLPF energy were 2 J, the values per unit mass of an average of 1139000 J/kg. It shows that such LLPF have high-energy content and can be considered as activators for combustion.

**Figure 6** Electron microscope photo of the shell.

### 3. Structures in corona discharge

Corona discharges are typically obtained by applying of a high potential between a small diameter needle, charged positively or negatively and plate or coaxial cylinder [9-13] charged oppositely. The principle scheme of the experimental device is represented in Figure 7. It consists of a ditch filled with a liquid (water, alcohol, kerosene, etc), and electrical circuit. A current in a discharge was measured by a milliamperemeter A1, a voltage was measured by a chain consisting of resistance R1 and a milliamperemeter A2. Experiments have been carried out with a help of different cavities: rectangular – dielectric, cylindrical – dielectric and steel. Sizes of the rectangular cavity were 70×20×15 mm (length, width and depth). Characteristics of cylindrical cavities were Ø125 mm - diameter, 10 mm - depth and 185 mm - diameter, 20 mm- depth, respectively. An investigating liquid was poured into a cavity, a negative or positive electrode was located directly over the liquid, a distance between the electrode and the liquid could be varied in a range (1-30) mm. The needle diameter was 3 mm, a radius of a tip was 0.4 mm. An electrode of another polarity was located directly in the liquid. (The metallic cavity was used as the electrode). It was connected with a feeding source through a hermetic unit. In this case, the liquid becomes the second electrode.

**Figure 7** Principle scheme of the experimental device. 1-Ditch, 2- Liquid (water, alcohol, kerosene), 3-Anode, 4- Post, 5- Cathode, 6-Power supply

A high voltage generator was used as the feeding source, it allowed to measure a voltage on the electrodes from 2 kV to 25 kV with a step of 1 kV. Typical range of the voltage was 5-25 kV. Appearance of the spark discharge was accompanied with a drop of the voltage at the capacity, which was detected by the voltmeter. In this device we used a ballast resistance, which value was 510 MΩ. Typical values of a current were 1-50 μA.
Experiments with discharges over tap water
Our experiments have shown that application of negative and positive corona discharges over water surfaces in plastic or metallic cavities lead to appearance of a rotating funnel, it was evidently caused by the ion wind.

Vortices over surfaces of liquids appeared in case of non-central position of the upper electrode with respect to the cavity or in rectangular cavities.

Experiments with alcohol
In case of alcohol in the plastic or metallic cavity, corona discharges lead to appearances of different electric hydrodynamic effects over the surface of the liquid.

Funnels appeared at both polarities of the upper electrode. In Figure 8 (left photo) we present a photo of such a phenomenon. In case of the positive upper electrode a rotating funnel usually appeared (it was evidently caused by the ion wind) initially. This process finishes by an appearance of liquid columns, see Figure 8 (right photo).

![Figure 8](image1)
Figure 8 Left photo: appearance of a cavity over the alcohol surface, metallic cavity, a distance between the upper electrode and a surface is 5 mm. Positive corona. Right photo: Appearance of a conical column over the alcohol surface, metallic cavity, a distance between the upper electrode and a surface of the liquid is 7 mm. Positive corona.

![Figure 9](image2)
Figure 9 Appearance of jets and drops over the alcohol surface, dielectric cavities, a distance between the upper electrode and a surface of the liquid is 7 mm (left photo) cylindric cavity, 17 kV and 6 mm (right photo) rectangular cavity, 20 kV. Negative corona.

The development of the process finally finishes by appearance of the electric arc. Usually this takes place at high applied voltage (15-25 kV). These columns are either standing in the funnel center or on a side of a funnel. Liquid columns have usually a widening at the tip. Sometimes a violet-bleu glow on the tip appears. It speaks about forming of the breakdown field near the tips. At voltage about 15-25 kV [10-13] these columns often transform into bursting jets. In Figure 9 one can see such jets
consisting of drops. Images were obtained with a help of a digital video camera Canon Digital IXUS 70 with frame duration 33 ms.

Namely creation of such charged drops can be used for applications for vaporization of liquid fuel in combustors [4].

Similar phenomena are known in EHD [14-17]. They are connected with a development of Rayleigh, Taylor, Tonks and Frenkel electrostatic instabilities on surfaces of charged drops and jets when Coulomb forces of accumulated charges become greater than the surface tension force.

*Experiments with Al$_2$O$_3$ powder*

Experiments with the Al$_2$O$_3$ powder on a liquid surface are of particular interest. The powder particles were of 30 μm in diameter. When the applied voltage exceeds a value of 7 kV vortices very intensively mixing of the liquid appears.

![Image of a column](image)

*Figure 10* Column has a length of 7mm and a diameter 1 mm a distance Between the upper electrode and a surface is 5 mm. Positive corona.

Experiments with dry and wet clay covered with aluminum powder were conducted also. Some of them demonstrated interesting results, such as arise of a clay column (see Figure 10) covered with metallic powder that provides a possibility to deeper understand the structure of columns. This composite column widens at its head as if some charge tried to separate it into drops. So, one can suppose a manifestation of two electro hydrodynamic instabilities in this case: the first one - the appearance of the column and the second one– Rayleigh instability of charged object electrostatic separation. Since we have realized instabilities then it was difficult to formulate an experiment where such columns were well observed.

4. Conclusions.

Experimental studies on creation of long-lived heterogeneous structures (LLGS) in air have been carried out.

It was possible to experimentally obtain LLGS of the alloy of lead and tin. The composition of the shell and the core of formations are significantly different; this means that boiling can occur inside the LLGS leading to the separation of the composition into a light part-the shell and the heavy part-the core. This result is new and it allows to consider LLGS as a new object for investigations in low temperature plasma. As a result of the work, it was possible to obtain photographs of the core and the second one– Rayleigh instability of charged object electrostatic separation. Since we have realized instabilities then it was difficult to formulate an experiment where such columns were well observed.

Creation of corona discharges has been realized over tap and distilled water, alcohol, glycerin and their mixtures. Conducted experiments and their analysis show that the corona discharge over liquids can cause different hydrodynamic phenomena including appearance of the ion wind and different
hydrodynamic instabilities. Funnels appear both in positive and negative coronas. Appearance of liquid jets and columns is a result of the Tonks - Frenkel and Rayleigh instabilities in conditions of positive and negative corona. This phenomenon depends on a value of electric field strength and is better realized in liquids with low density and surface tension. Separation of these jets and columns in drops are connected with instabilities of charged drops, which can also be considered as LLGS. These experiments help to clarify modes of discharges realization and their connection with appearance of funnels on the surfaces of liquids and columns in case of alcohol. Application of corona discharge for creation of charged particles and structures in plasma aerodynamics or plasma assisted combustion require additional investigations.

Acknowledgements
This work was carried out in the framework of State Assignment of Russian Federal Agency of Scientific Organizations (project 0082–2018–0001, registration code AAAA–A17–117112240026–5).

References
[1] Avramenko R F Bakhtin B I Nikolaeva V I, et al 1990 Zhur. Tekh. Fiz. 60 12 73.
[2] Avramenko R F Bakhtin B I Nikolaeva V I, Poskacheeva L P 1994 Ball lightning in the laboratory ed. Avramenko R.F., Bychkov V.L., Klimov A.I., Sinkevich O.A. (Moscow: Khimiya Publishers) 15
[3] Ershov A P Rozanov V V Sysoev N N et al 1994 Physical hydrodynamics 4 Preprint of phys. dep. of MSU Moscow MSU 8/1994
[4] Bychkov V L Chernikov VA Osokin A A et al 2015 IEEE Trans. Plasma Sci. 43 4043.
[5] Pokhil P F Belyaev A F Frolov Y V Logachev V S Korotkov A I 1972 Combustion of powdered metals in active media (Moscow: Nauka publishers)
[6] Emelin S E Semenov V S Bychkov V L et al 1997 Tech. Phys. 42 269
[7] Emelin S Bychkov V Astafiev A Kovshik A Pirozersky A 2012 IEEE Trans. Plasma Sci. 40 3162.
[8] Bychkov V L Bychkov A V Timofeev I B 2004 Tech. Phys. 49 128.
[9] Encyclopedia of low temperature plasma 2009 (vol.3. Applied Plasma Chemistry) ed. Lebedev Yu A, Plate N A, Fortov V E. (Moscow: Yahys-K publishers)
[10] Aleksandrov A F Bychkov V L Bychkov D V et al 2011 Moscow Univ. Phys. Bull. 66 390
[11] Bychkov V L Chernikov V A Volkov S A et al 2011 IEEE Trans. Plasma Sci. 39 2640
[12] Bychkov V L Chernikov V A Volkov S A et al 2011 IEEE Trans. Plasma Sci. 39 2643
[13] Landau L D Lifshitz E M 1982 Electrodynamics of continuous media (Moscow: Nauka Publishers)
[14] Sararin V A 2002 Equilibrium of liquids and its stability (Moscow: Institut Kompiuternykh Issledovanii Publishers)
[15] Grigor’ev A I Sinkevich O A 1984 Zhur. Tekh. Fiz. 54 7 1276.