FEATURES OF LIQUID DROPLET DYNAMICS AT THE INTERFACE OF TWO IMMISCIBLE MEDIA

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This work presents the results on visualization of dynamics of droplets prepared on the water or alcohol basis, moving inside liquid or crossing air - oil or water – oil interface in the three-layer system: air – oil – water.

For the first time investigations of various shapes formed by liquid droplets' impact on another liquid started a century ago [1], however, the interest to them is kept until now [2]. In particular, this is explained by the fact that structured deformations of droplets, observed experimentally, are caused by their linear or nonlinear hydrodynamic instability. The interest to the problems of media instability at the interface is increasing now because of many complex problems in the field of two-phase hydrodynamics and heat and mass transfer.

Recently the studies of droplet motion in liquid media are being developed intensively. Particular attention is paid to the droplets, which cross or lie on the interface of liquid layers, where intensive mass transfer with the ambient medium and chemical reactions occur [3]. These investigations are interesting for hydrology, biology and other science applications; they can be also used for development of modern technologies [4].

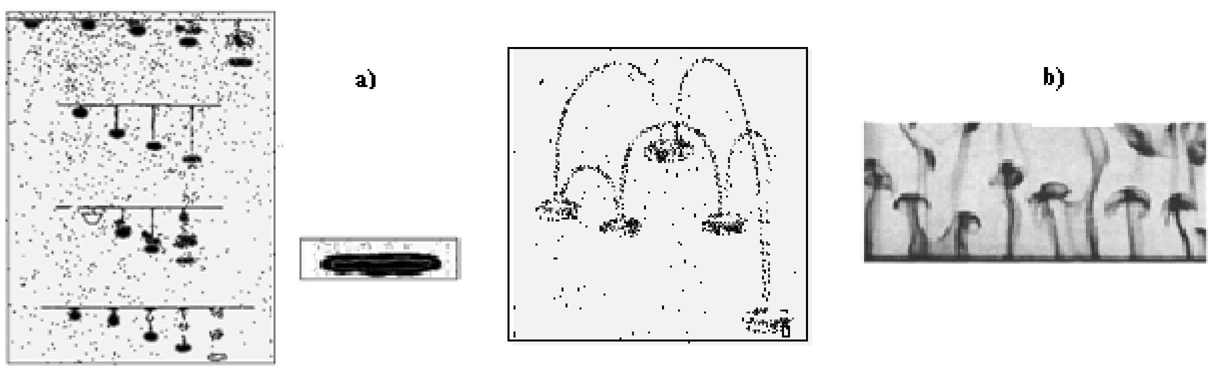

Fig.1

The development stages of droplet at its fall in another liquid (below it is called the experimental one) in accordance with experiments of [1] are shown in Fig. 1a. It is shown in [1] that if liquid is immiscible with the experimental liquid, the droplet keeps its shape along its way. Another pattern is observed, if liquids are mixable (Fig. 1a). In this case the droplet is transformed into a jet with a vortex ring in the frontal part. With droplet fall the vortex ring becomes unstable and disintegrates into a group of vortex rings of the smaller scale. It seems that the analogues of the observed jets with vortex rings at the front are the so-called thermals (Fig. 1b), formed above the heated surfaces.

Thus, it follows from [1] that the important characteristics of the considered problem are mutual properties of the used liquids.

In the current study we used the following liquids for droplet preparation: pharmacy alcohol solutions of iodine (1.7%, 2.6% and 5%,), brilliant green (1%), and water solution of dye rhodamine. The droplet diameter varied within 1 – 5 mm. The temperature of used liquids in every experiment was the same and it equaled 20 – 27°C**.** The transparent walls of the vessel allowed visual observations and video recording by a Web camera as well as photo shooting.

The photographs were made by the Pentax K10D camera with matrix of 10 Mpix with the size of 23.5×15.7 mm. Telescopic lens SMS Pentax of 50-200 mm was used. The images in jpg format were saved on the hard drive of PC. Recording was performed with the use of a camera tripod, and natural illumination or an ordinary lamp was used as the light source.

The Genius iLook 300 web camera with the matrix of 640×480 pix (0.3 Mpix) and shooting speed of 30 frames per second was used for recording. The video files were saved on PC hard drive, and then they were processed and analyzed.

The conditions of miscibility [1] in our experiments was satisfied for all droplets (alcohol and water ones), when they were kept in water. Results of visualization on droplet motion in water, shown in Fig. 2, prove the conclusions of [1]. The left pictures in Fig. 2 were obtained for rhodamine droplets, and the pictures on the right were obtained for iodine droplets. The upper layer is oil, the lower layer is water.

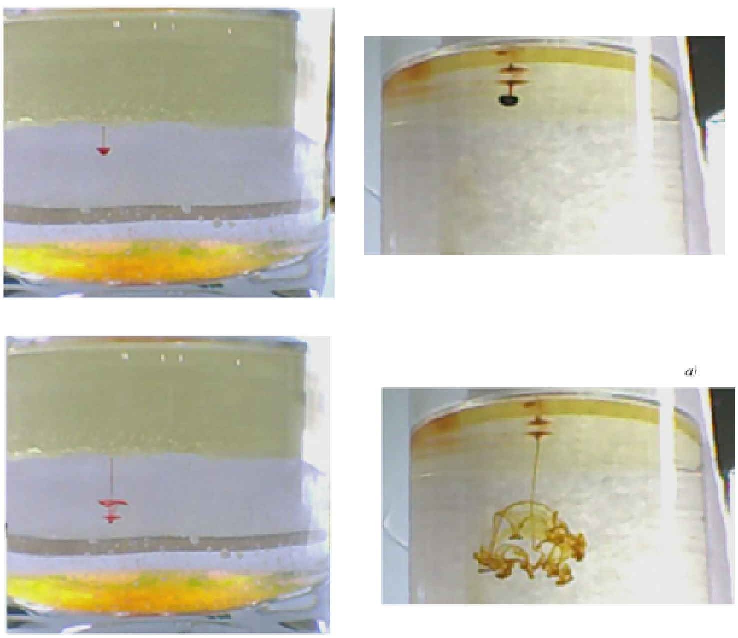

Fig. 2

It is necessary to note the difference between our experimental conditions and experimental conditions of [1]. We have used two liquids simultaneously: water and oil; they were separated by the obvious interface. In experiments of [1] the droplet was put into the experimental liquid from the very beginning, and in our experiments the droplet came into the experimental liquid after interface crossing. At this, water and alcohol droplets behaved differently on the interface.

The rhodamine water droplets stayed on the interface during some time, then they collapsed, and the pulsed emission of contained liquid into water occurred.

When alcohol droplets fell on the water – oil interface, sometimes they spread over this surface forming complex nonstationary structures, described below. If the droplet was kept, its following dynamics was similar to dynamics of the water droplets. If the pulse transferred to the alcohol jet at collapse was not high, after some descending motion it came back to the interface.

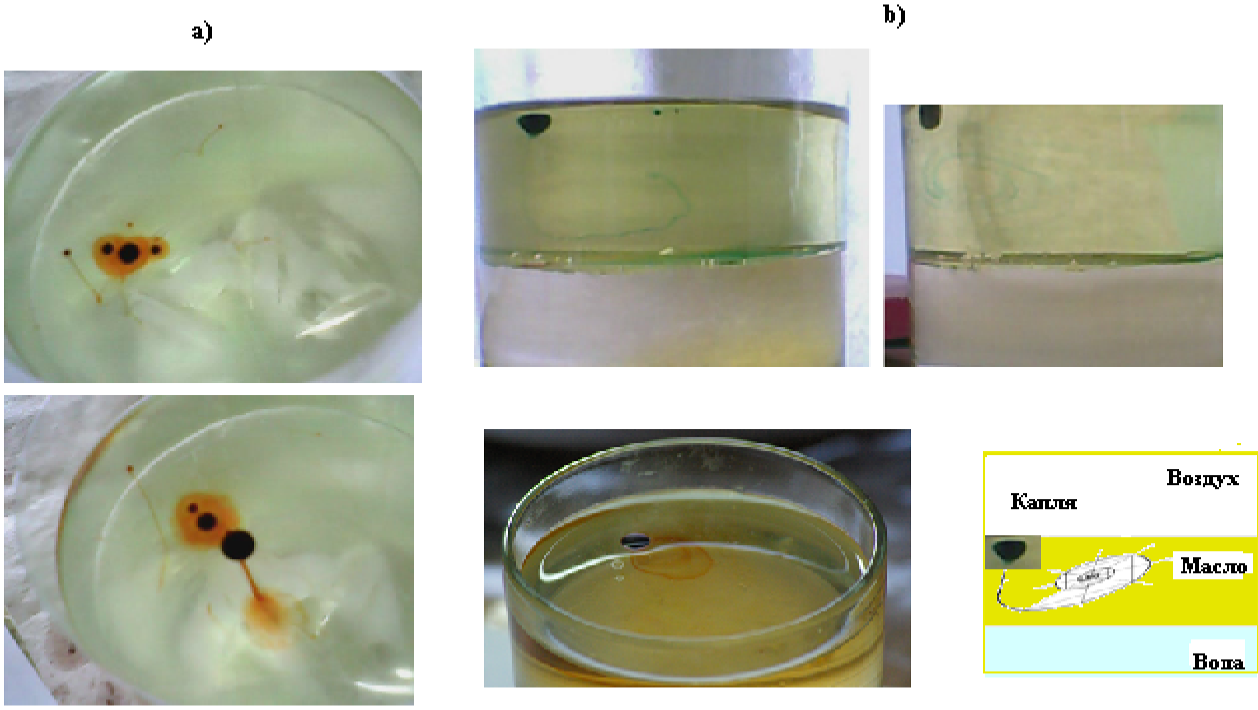

Fig. 3

At droplet motion in the oil layer the condition of miscibility was not satisfied for water droplets and, partially, for alcohol ones because the processes of alcohol and oil mixture are very slow. The latter influenced the patterns observed in experiments. Water droplets at motion in water kept their shape. Liquid from alcohol droplets started flowing out over time (Fig. 3), forming a thin dyed microjet – “tail”. Especially interesting results were obtained, when the alcohol droplet was placed on the upper oil – air interface. According to observations, several development scenarios are possible. The first scenario is often observed for the iodine droplets with iodine concentration less than 5%: the droplet spreads over the interface forming complex nonstationary structures, described below. Another scenario: a jet flows out of the droplet on the oil – air interface, with time it reaches the oil - water interface, and liquid flows from the upper surface through a formed channel to the lower one, where it forms a droplet (Fig. 3). We have also observed situations, when together with formation of a channel the droplet was separated from the upper interface surface and fell on the lower surface. At this, the channel formed before droplet separation stayed visible some time, what is illustrated in Fig. 3a.

One more scenario, which attracted special attention, is shown in Fig. 3b. In this case, the jet from the droplet moved downwards first to the lower interface, then, it turned by 90 degrees without a contact with the surface. In some cases the droplet even started motion in reverse direction towards the upper interface. In particular, in one experiment the jet from the droplet of brilliant green made several turns, forming a flat spiral: see the upper picture in Fig. 3b. The full turn of the jet from the iodine droplet is shown in the lower picture in Fig. 3b.

In general, the observations showed:

1. The "tails" were observed at any position or motion of alcohol droplets in the oil layer.
2. The area of droplet, where the jet was generated, and initial direction of this jet did not usually coincide with the axis of droplet symmetry or direction of gravity.
3. If the droplet stayed on the upper interface, additional jet branches could be formed there. At this, they stayed within the interface plane and did not grow in the direction of gravity action.

According to analysis of results obtained, an intermediate layer is formed on the droplet surface because of mutual mixing of oil and alcohol contained in the droplet. Then this intermediate layer flows as a microjet to oil. Following dynamics of a microjet is determined by its hydrodynamic stability and local density nonuniformity in oil, which effect the direction of jet propagation and its bend. On one hand this explanation can find the evidence in literature, on the other hand it is proved by our additional experiments, where the portion of alcohol in the droplet was increased. It was found out that an increase in the portion of alcohol leads to the faster appearance of jet from the droplet and its faster washing out in oil. The analogue of an alcohol jet observed in our experiments can be evaporating high-temperature jets studied in [5] as well as other jets, whose dynamics is mainly determined by the activity of molecules on the jet surface. In particular, the breakdown and bend of the high-temperature jet, propagating in vacuum, were detected in [5].

The pictures of jets from iodine droplets on the upper surface of oil are shown in Fig. 4. Some kind of umbrella in the frontal part of the jet attracts attention in the left picture; it is probably the vortex ring similar to that observed at water droplet fall in water. It can be seen in the right picture that with jet development under the action of random local nonuniformity of density the jet begins developing against the initial direction.

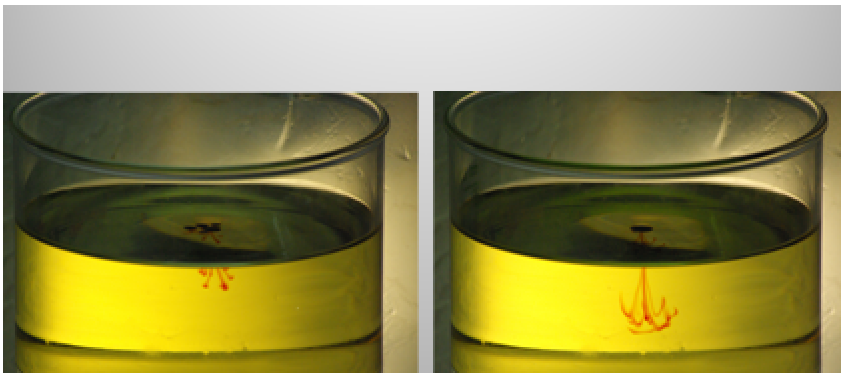

Fig. 4

The next group of the figures and pictures shows the structures, formed at droplet spreading over the interface. We should note that in our experiments the water droplets saved their shape crossing the interface, and the iodine droplets spread, if iodine concentration there was lower then 5%.

b)

a)

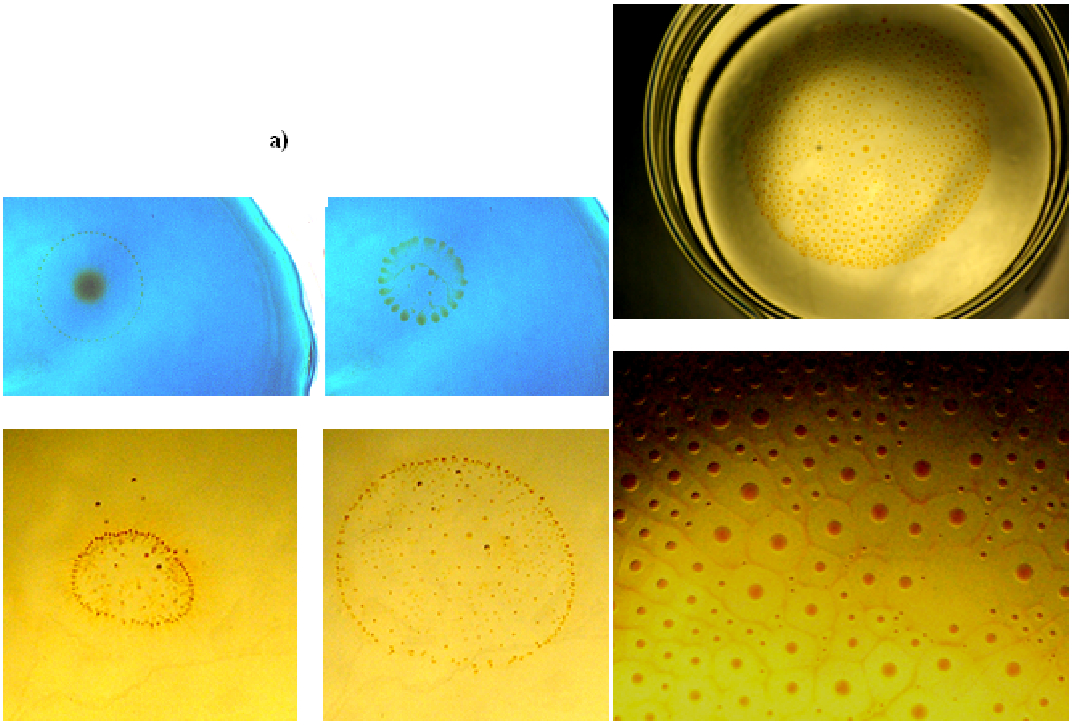

Fig. 5

The spreading stages of iodine droplet (1.7 and 2.6 %) put on the upper oil surface are shown in Fig. 5. The blue background of the upper pair of pictures was obtained via addition of brilliant green to the water layer under oil. After precipitation the droplets spread first, increasing their diameter, and then began constricting. It is obvious that the spread droplet is limited by a border of small droplets, formed at splitting of the initial drop. This border becomes obvious with droplet spreading.

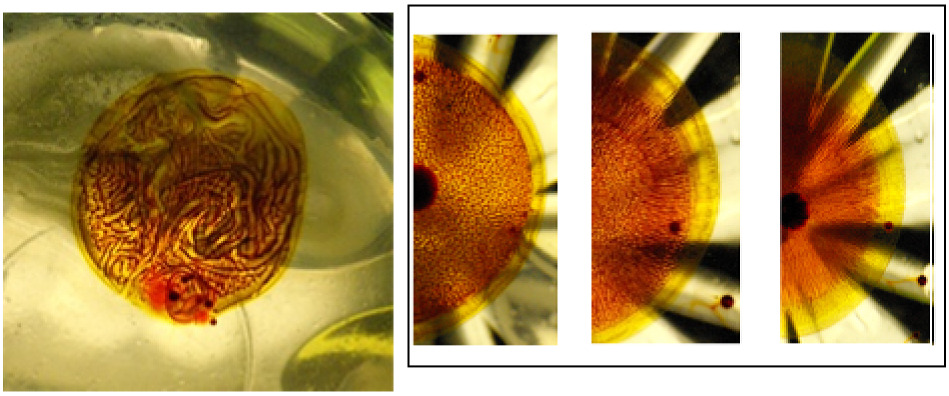

Fig.6

While constricting the border becomes thicker, the size of forming droplets increased because of adjacent droplets merging. The next stage of this process development is either a single droplet similar to the initial one or a group of droplets. Then, a jet starts flowing out a droplet to oil, and so on.

The pictures of spreading of an iodine droplet with iodine concentration of 1.7% over the oil surface are shown in Fig. 5b. An enlarged fragment of the upper picture is shown in the lower part of Fig. 5b. It is obvious that the spread drop is smaller droplets combined into the whole by the cellular structure. In other words, we can see the united cells with smaller droplets inside; the larger the droplet, the larger the size of a cell. Perhaps, the observed structures are the Marangoni cells, and they are explained by the difference in surface tension of oil and iodine.

The versions of structures registered at iodine droplet spreading over the water – oil interface and explained by the difference in surface tension of water and iodine are shown in Fig. 6. Three patterns on the right register the structure of one droplet, spread at different time moments. It is obvious that the Marangoni cells, similar to those in Fig. 5b, are formed at the initial stage. As in Fig. 5b, inside the cells there is a core, which is an iodine droplet, significantly less than the initial one. Then, the microjets are formed on the droplets of the cell core, and under the spread droplet we can see the system of microjets, which corresponds to the structure of the spread droplet. Then the pattern structure changes significantly from the periphery, it becomes ray-like.

Conclusions

1. The behavior of alcohol droplets on the oil surface (interface with air) depend significantly on iodine concentration.

At concentration of 5% the iodine droplets usually saved their closed shape, stayed on the oil surface, ejected a microjet, etc. Sometimes they spread slightly, but after microjet formation they began constricting.

At concentration of 2.6% the iodine droplets spread over the oil surface. At first, an iodine circle increased, reached its limit and began constricting sometimes to the size of initial droplet.

At even smaller concentration the droplets spread also and during spreading they disintegrate into the smaller droplets. Not regular, but obvious cells with droplets in their centers, resembling Marangoni structures were formed on the iodine film.

2. It does not matter, what was the interface, where the iodine droplet was spread. In any case the structure of regions with high or low iodine concentration is formed there. Almost immediately the microjets appear from the cells of this structure, and then the structure of microjets meets the structure of the spread droplet.

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