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SEQUENTIAL ABSORPTION OF MICRODROPS BY DOUBLE-LAYER POROUS MEDIA

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Abstract

The subject of this paper is the absorption of microdrops by multilayer porous media. The consideration is based on numerical simulation of sequential absorption of two droplets at its arbitrary location on the surface of single- and double-layer porous media. For description of fluid flow from a droplet into a porous material the Euler equations taking into account surface tension forces were used together with the unsteady filtration equation. The layers of porous medium were characterized by effective permeability coefficients dependent on porosity and pore size. The change in the droplet shape during absorption, the propagation of absorbed fluid in a porous medium, the fields of velocities and pressure are the output data of the problem. The effect of porous medium structure parameters and relative location of droplets on the rate of absorption and distribution of absorbed liquids is analyzed using the numerical experiment. It is shown that the presence of the second layer can significantly affect the duration and result of droplet absorption. The ratio of the pore size in the layers is found to be the main parameter that governs the effect of the second layer.

Keywords: Absorption of microdrops, Porous media, Rate of absorption,
Distribution of liquid

1. Introduction

The absorption of small size drops by porous media is a problem of both fundamental concern and practical interest. The deposition of liquid droplets on the surface of permeable solids accompanied by droplet spreading and imbibition serves as basic

Nomenclatures

a	Droplet radii, m
Bo	Bond number
d	Pore size, m
D_{dif}	Coefficient of diffusion, m^2/s
Fo	Fourier number
g	Gravitational acceleration, m/s^2
h	Thickness of porous layer, m
k	Permeability coefficient, m^2
L	Distance between the centers of droplets location, m
Oh	Ohnesorge number
p	Pressure, Pa
R_1,R_2	curvature radii of the droplet surface, m
Re	Reynolds number
S	Area of formed wet spot, m^2
t	Time, s
u, v, w	Velocity components, m/s
V	Volume of the droplet, m^3
We	Weber number
x, y ,z	Cartesian coordinates
Greek Symbols	
ε	Porosity
θ	Contact angle, grad
μ	Dynamic viscosity, Pa.s
ρ	Density, kg/m^3
σ	Surface tension coefficient, N/m
τ	Time scale, s
Subscripts	
a	Absorption
d	Droplet
in	Inertial
L	Layer
p	Pore
v	Viscous
1	First layer or first droplet
2	Second layer or second droplet
o	Initial

process of wide range of technologies such as textile processing, high-quality inkjet-printing, spray-painting and coating including the protection of softwoods and pharmaceutical tablets. It is the reason of considerable attention in the literature to various aspects of this process [1-9].

In respect to the description of the microdrop absorption as a physical phenomenon the following principal aspects of the problem should be distinguished. The important feature of the behavior of liquid drop on porous base is the presence of free and contact surfaces. Capillary spreading and the tendency of droplet to minimize the free surface determine the change in the droplet shape during absorption and formed pressure field in the liquid and, hence, the duration of process and resulting distribution of liquid in porous medium. In most applications the porous media have various structures including multilayer structure with different permeability of layers [2, 7, 8, 10-12]. Both spray coating and printing processes involve the stage of sequential deposition of fine droplets onto the surface of porous media. The presence of the porous sublayers and relative location of deposited droplets can significantly affect the dynamics of absorption and the liquid distribution in the medium.

To describe the absorption of the single droplet Starov et al. [7] proposed an analytical model for liquid flow from a droplet to a thin porous bed. Davis and Hocking [2] considered a two-dimensional model in which the porous media was modeled by alternating vertical slits permitting liquid flow in the vertical direction only. Alleborn and Raszillier [8] numerically studied the effect of the porous media permeability on droplet absorption in the case of two-layer media with identical pore sizes in the layers. Zadražil et al. [9] studied the droplet spreading, imbibition and solidification on porous base that is assumed to be a membrane composed of an array of pores having fixed width. In our previous work [13] a two-dimensional model was considered to describe the absorption of single droplet by double-layer porous media. The porous layers are characterized by effective permeability coefficients dependent on porosity and pore size.

With reference to the aforementioned technologies the present work focuses on numerical simulation of the sequential absorption of two microdrops by double-layer porous media. The practical possibilities of developed model are illustrated on the example of ink jet-printing processes [11, 14].

2. Theory - Formulation of the Problem

The subject of the present study is the sequential absorption of two droplets at its arbitrary location on the surface of double-layer porous media (Fig. 1). The case when a pore size in the upper layer is smaller than the droplet size ($d_1 << a$) is considered. It

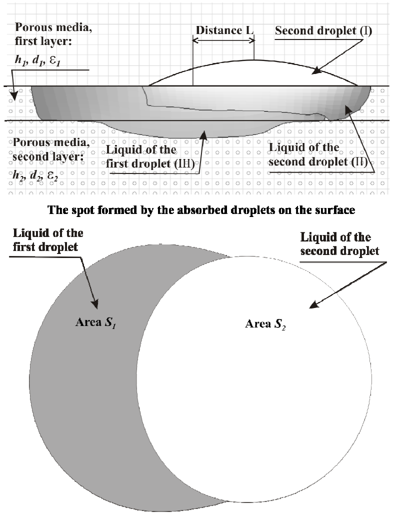

Fig. 1. Schematic of the Problem: Sequential Absorption of Two Droplets by Double Layer Porous Media.

is assumed that absorption of the second droplet starts when the first droplet has already been absorbed. Initially, the first and second droplets are assumed to be shaped like a spherical segment. The subsequent evolution of the droplet shape is governed by the absorption process, capillary spreading and tendency of the droplet to minimize the free surface area due to surface tension forces.

To formulate the possible hydrodynamic approaches for description of liquid flow in a droplet on the porous medium surface the order-of-magnitude analysis of problem conditions and time scaling lows of processes can be made. For the droplets with size $a \sim 10^{-4}$ m the Bond number Bo $= \rho a^2 g/\sigma << 1$, therefore the gravitational effects are negligible. For low viscous liquids ($\mu < 10^{-2}$ Pa·s) the values of Ohnesorge number Oh $= \mu/(\rho\sigma a)^{0.5} = \mathrm{We}^{0.5}/\mathrm{Re} << 1$ that corresponds to the case of hydrodynamic flows where viscous forces are small compared with surface tension forces. Follow to argumentation of Schiaffino and Sonin [15] the droplet spreading is driven by capillary force, and resisted by inertia. The inertial term, $\rho u/\tau_{in}$, is balanced by the capillarity-induced pressure gradient, which is the of order σ/a^2. The inertial motion has a time scale $\tau_{in} = (\rho a^3/\sigma)^{0.5}$, and the time scale for viscous damping is the viscous diffusion time, $\tau_v = \rho a^2/\mu$. The value of Reynolds number, Re = Oh^{-1}, and ratio $\tau_v/\tau_{in} = \mathrm{Oh}^{-1}$ are large for these conditions indicating underdamped liquid motion.

The time scale of droplet absorption can be estimated according to Darcy's equation [16, 17] as $\tau_a = z^2\mu/kp_p$. Here z is characteristic scale of liquid penetration into porous medium, that can be estimated as $a/\varepsilon^{1/3}$ and p_p is the capillary pressure inside the porous medium, which can be estimated as $4\sigma/d$. For most porous media ($\varepsilon < 0.4$ and $d \sim 10^{-6}$ m) the time of droplet absorption is significantly higher than the time of viscous damping and inertial spreading of droplet ($\tau_a > \tau_v > \tau_{in}$). The latter means that the penetration of liquid into porous medium is the defining process (slow process). The spreading of droplet over surface of porous medium during absorption, which results in a change of droplet shape (the size of droplet base and curvature of free surface) follows quickly. Thus, the approach of inviscid liquid flow can be used to describe the motion of liquid in the droplet during absorption.

The Fourier number estimation ($\mathrm{Fo}=D_{dif}\tau/d^2$) showed that Fo<<1 for molecules of liquid. Therefore, it is supposed that the liquids of the first and second droplets not mix during absorption.

In the case of layered porous media and two droplets absorption, the liquid motion in the medium is non-stationary unsteady flow. Therefore, the process of droplet absorption is modeled by simultaneous solution of the equations describing liquid flow in the droplet and equations of unsteady filtration in the porous medium.

2.1. Liquid flow equations

The liquid flow in a droplet on the surface of a porous medium is described using the flow equations for an incompressible, inviscid Euler fluid and the continuity equation [18]:

$$\frac{du}{dt} = -\frac{1}{\rho}\frac{\partial p}{\partial x},\ \frac{dv}{dt} = -\frac{1}{\rho}\frac{\partial p}{\partial y},\ \frac{dw}{dt} = -\frac{1}{\rho}\frac{\partial p}{\partial z},\ \frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} + \frac{\partial w}{\partial z} = 0 \qquad (1)$$

where $d/dt=\partial/\partial t+u\partial/\partial x+v\partial/\partial y+w\partial/\partial z$.

It is assumed that the absorbed liquid occupies a continuous region in the porous medium. The liquid flow in the pores is laminar and is described by the equations of unsteady filtration of an incompressible liquid [16, 17]:

$$\frac{du}{dt} = -\frac{1}{\rho}\frac{\partial p}{\partial x} - \frac{\mu}{\rho \cdot k}u,\ \frac{dv}{dt} = -\frac{1}{\rho}\frac{\partial p}{\partial y} - \frac{\mu}{\rho \cdot k}v,$$
$$\frac{dw}{dt} = -\frac{1}{\rho}\frac{\partial p}{\partial z} - \frac{\mu}{\rho \cdot k}w. \qquad (2)$$

Here $d/dt = \partial/\partial t+\varepsilon^{-1}(u\partial/\partial x+v\partial/\partial y+w\partial/\partial z)$. Following the study of Scheidegger [19], which considers experimental data for liquid flows in porous media of various structures, we use the following expression for the permeability coefficient:

$$k = \frac{d^2\varepsilon^3}{150(1-\varepsilon)^2} \qquad (3)$$

2.2. Boundary and initial conditions

At the free surface of a droplet, the following boundary conditions are imposed: a jump in the normal stress p_d due to capillary forces:

$$p_d = \sigma(1/R_1 + 1/R_2) \qquad (4)$$

At the droplet–medium interface, the condition of equality of the normal flow velocities in the droplet and in the porous media is used. At the part of outer surface of the porous medium that is filled by the absorbed liquid (the wet porous material – air boundary), the non-penetration condition is imposed, which implies zero normal velocity of the liquid flow in the porous medium.

The action of capillary forces inside the porous medium at the absorption-front boundary (the boundary between the region of the porous medium filled by the liquid and the region free of the liquid) is characterized by the pressure discontinuity given by the Laplace formula [20]

$$p_p = -4\sigma\cos\theta/d \qquad (5)$$

The initial droplet shape (a spherical segment) is characterized by height and base diameter, from which the contact angle for the medium material can be estimated [21, 22]. The initial geometry of the porous region with the absorbed liquid under the drop is specified in the form of a thin disk whose diameter is equal to the droplet base diameter and the thickness is equal to the pore size. The initial liquid flow velocities in the droplet and the porous medium are set equal to zero. The initial pressure in the droplet is defined by Eq. (4), and the initial pressure in the porous medium region with the absorbed liquid under the droplet is set equal to p_p, in accordance with Eq. (5).

Depending on the distance L between the centers of first and second droplets location (see Fig. 1), the thin disk that used as initial approximation to describe the absorption of the second droplet may be fully or partially located inside the porous medium domain filled by the liquid of the first droplet. Correspondingly, the contact boundary of liquids of the first and the second droplets is formed. The condition of equality of normal components of velocities of the both liquids motion on the contact boundary is imposed. This implies that the liquid of the second droplet during its absorption forces the liquid of the first droplet out of the pores. The droplet on the surface of porous medium, the porous medium region filled by liquid of first droplet and the region that has been filled by liquid of second droplet during absorption are presented as separate mathematical bodies (Fig. 1). Initially, each body is a body of revolution. However, if the distance between the centers of first and second droplets location $L > 0$ the axial symmetry of each body would be disturbed in the course of absorption.

2.3. Numerical methods and procedures

The numerical solution of the problem is based on the use of the moving-mesh procedure [23]. System of Eqs. (1) and (2) was numerically integrated using an explicit conservative scheme of first-order accuracy. In the construction of the finite-difference approximation, Eqs. (1) and (2) were converted for use in the moving-mesh procedure employing the method proposed and tested by Meshcheryakov et al [24]. For each mathematical body (Fig. 1) a separate grid is constructed. The three-body configuration allows tracing the distribution of liquids of first and second droplets inside the layers of porous media.

According to used procedure, new values of u, v, w and p were found at each time step. In the first step, the calculation was performed for the specified initial and boundary conditions for the droplet and the liquid in the porous medium, and in the subsequent steps, for the current values of the indicated parameters. The obtained flow velocities were used to determine the new position of the droplet boundaries on the surface of the porous media and the advance of the absorption front. Once the liquid in the porous medium reaches the outer surface of medium (the porous material – air boundary), the boundary condition given by Eq. (5) was replaced by the non-penetration condition. When the liquid reaches the interface between the layers, the pressure jump at the absorption front boundary (5) was changed, and the permeability coefficient (3) was adjusted to the layer parameters. As a result of the numerical solution, we obtained the evolution of the droplet shape in the absorption process, the distribution of liquids of first and second droplets in the porous material, and the flow-velocity and pressure fields.

3. Experimental Procedure. Model Verification

For the case of single droplet absorption the validation of developed model was performed in the work [13]. The numerical results were verified by comparing with data of experiments with absorption of droplet deposited on various porous media (standard glass filters and double-layer paper). A comparison of numerical and experimental data on droplet shape change during absorption, the time scale of process and the fluid distribution in porous media showed that the developed model provides an adequate prediction of single droplets absorption process. The difference between the experimental and calculated data does not exceed 10%.

Analogously experiments were carried out to validate the model as applied to the sequential absorption of two droplets. A technique of the flash videography [25] and subsequent reconstruction of stages of fast repeated processes with a time resolution of 10 μs was used [13, 26]. The imaging was performed with a digital video-camera equipped with a microscope lens and a stroboscopic system with light-pulse duration of 1.5 μs. Droplets about 50 μm in diameter were deposited sequentially onto double-layer paper using the inkjet printer cartridges. The second droplet was deposited at instant when the first droplet has already been absorbed. The different stages of the process were identified by varying the time interval between the moment of droplet ejection and the stroboscopic flash. The identified stages were recorded, and their sequence was reconstructed. The measured geometrical sizes of droplets were used to calculate the time dependence of droplet volume change during the absorption process. The measurement error for the droplet base diameter and for the droplet height was 2 μm.

As an example of experiments performed to validate the developed model, the data on sequential absorption of two droplets by the double–layer paper is presented. The upper layer of paper of thickness h_1= 25 μm had a granulated structure with an estimated size of grains 5 - 10 μm and porosity ε_1=0.2 [13]. The second layer of thickness h_2= 80 μm had a fibrous structure with void sizes up to 30 μm. The test liquid was water. The water was colored with ink to visualize the absorption region. On the addition of ink (volume fraction 0.1%) to water, the change in the properties of water (viscosity, density, and surface tension) was within 1%.

The experiments showed that no notable change in the droplet volume (within the measurement error of the droplet size) occurred during the time interval from the droplet impingement onto the surface to the moment the droplet took the shape of a spherical segment. The contribution of liquid evaporation to the subsequent reduction in the droplet volume was within 10%.

The calculations were compared with the experimental data on first and second droplets absorption as follows. The initial data in the calculations were the droplet shape defined by droplet base diameter and height, the thickness and structural parameters of the porous media, and the properties of the liquid. The contact angle θ was set equal to the 'steady–state' contact angle for the droplet on the media surface, which, in turn, was determined from the height and the base diameter of the droplet. Initially, the results of numerical simulations for the duration of first droplet absorption and the size of the spot formed on the surface of the porous media after droplet absorption were compared with the experimental data. The varied parameter was the pore size. The difference between the experimental and calculated data not exceeding 10% was considered satisfactory.

Further, the value of pore size, which gave the best approximation of experimental data on first droplet absorption, was used at calculation and comparison with experimental data on second droplet absorption. Fig. 2 shows the data on sequential absorption of two droplets of volume $V_o = 0.9 \cdot 10^{-13}$ m^3 by the double–layer paper. The example of comparison in Fig. 2a and Fig. 2b corresponds the case when the distance between the centers of first and second droplets location $L = 0$. The variation of duration of second droplets absorption t_2 with the distance L between the centers of the first and second droplets location on surface of porous medium is presented in Fig. 2c. The experimental data and results of calculation are plotted in the relative coordinates t_2/t_1 and L/R_{d1}. Here t_1 and t_2 are the durations of first and second droplet absorption and R_{d1} is the radius of spot formed on the surface of porous medium by absorbed liquid of the first droplet. From the data in Fig. 2, it follows that the predicted droplet volume evolution and the size of the wet spot formed on the surface of paper are in good agreement with the experimental data. It should be noted that the adopted value of the pore size in the upper layer of the paper d_1 = 3.5 μm agrees with the data of optical microscopic measurements of the void sizes between paper grains (2-6 μm). The microscopic measurements and calculation data also showed that if the pores in the second layer have sizes $d_2 >> d_1$, the liquid of droplet of volume $V_o < 10^{-13}$ m^3 does not penetrate into this layer and is distributed over the volume of the first paper layer.

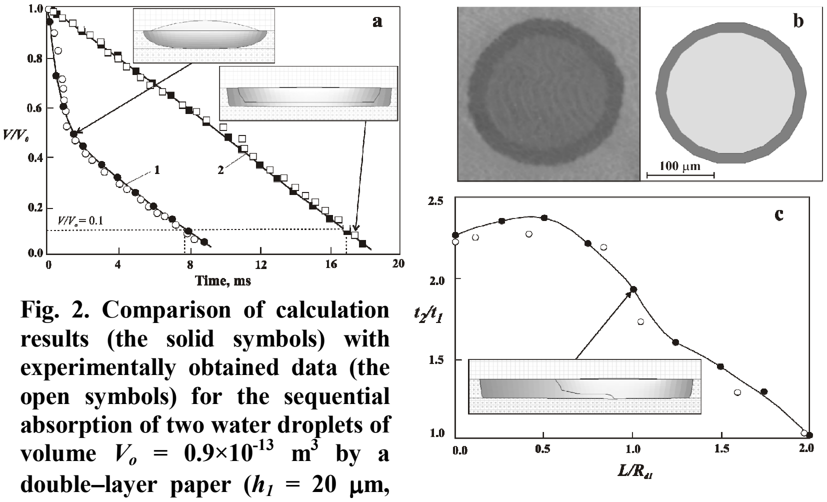

Fig. 2. Comparison of calculation results (the solid symbols) with experimentally obtained data (the open symbols) for the sequential absorption of two water droplets of volume $V_o = 0.9\times10^{-13}$ m^3 by a double–layer paper (h_1 = 20 μm, ε_1 = 0.2 and d_1 = 3.5 μm):

- **(a) the temporal evolution of the volume $V(t)/V_o$ of the first (1) and second (2) droplets;**
- **(b) the spot formed by the absorbed liquids of two droplets on the paper surface: experimental data (on the left) and calculated data (on the right);**
- **(c) the variation of duration of second droplets absorption t_2 with the distance L.**

4. Results of Numerical Experiments

Numerical experiments were carried out to examine the effect of the structure parameters of the double-layer porous media and relative location of the first and second droplets on its absorption. The volume of the test droplet was $V_o = 10^{-13}$ m^3. For the materials in the first and second layers, the contact angle was set equal to 30°. The varied parameters were the pore size in the layers, porosity, and the thickness of the upper layer in the porous media. The absorption process was characterized by the time t_a required for adsorption of 90% of the droplet volume. The distribution of the absorbed liquid in the porous media was characterized by the area S of the wet spot formed on the porous surface by the absorbed liquid. The procedure of droplet absorption time t_a determination is illustrated in Fig. 2a. For the case of first droplet absorption the time point when absorption front arrives at the interface between the layers is denoted on dependency 1 of Fig. 2a.

The effect of the thickness of the first layer of porous medium and pore size in the second layer on the duration of first and second droplets absorption is illustrated in Fig. 3. The dependencies of the area of the wet spots formed on the porous surface by the absorbed liquid of the first and first and second droplets versus thickness of the first layer of porous medium are presented in Fig. 4. The calculations are performed for the case when the distance between the centers of first and second droplets location $L = 0$. The inserts in Figs. 4 and 5 illustrate the distributions of liquids realized in the porous media. The calculation data are plotted in the relative coordinates t_a/t_{L1} and S/S_{L1}. Here t_{L1} and S_{L1} are the values of duration of droplet absorption and the area of formed wet spot that correspond to the limiting case of droplet absorption by a semi-bounded media with parameters equal to those in the first layer. The second layer does not influence the droplet absorption process. For zero thickness of the first layer, we have the second limiting case of droplet absorption by a semi-bounded media with parameters corresponding to those of the second porous layer.

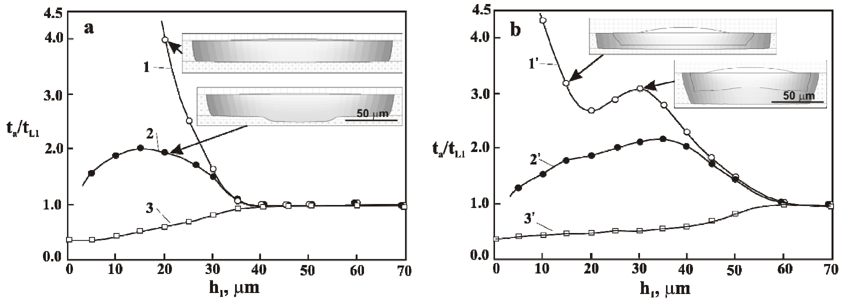

Fig. 3. Variation of duration of first (a) and second (b) droplets absorption with the thickness of the first (upper) layer in the double–layer porous media for $d_2 = 30$ μm (curves 1 and 1'), 8 μm (curves 2 and 2'), and 1 μm (curves 3 and 3'); $\varepsilon_1 = 0.2$, $d_1 = 2$ μm, and $\varepsilon_2 = 0.3$.

For intermediate thicknesses of the first porous layer, the duration of droplet absorption and the liquid distribution in the media depend appreciably on the ratio of the structural parameters of the layers (see Figs. 3 – 4). For large pore sizes in the second layer ($d_2 >> d_1$), almost no absorption by the second layer is observed, as is indicated by the large areas of the spot formed on the surface of the porous media (curves 1 and 1' in Fig. 4). The processes of absorption of first and second droplets are characterized by long times (curves 1 and 1' in Fig. 3). As the thickness of the layer increases, the duration of first and second droplets absorption and the area of the wet spot decrease (curves 1 and 1' in Figs. 3 – 4), approaching the values corresponding to the case of droplet absorption by a semi-bounded media ($t_a/t_{L1} \rightarrow 1$ and $S/S_{L1} \rightarrow 1$). However, in contrast to the case of first droplet absorption the duration of the second droplet absorption is decreased non-monotonically (curve 1' in Fig. 3). The curve of the second droplet absorption time versus the first layer thickness has characteristic inflection.

As the pore size in the second layer decreases ($d_2 > d_1$ and $d_2 \sim d_1$), a liquid flow to the second porous layer arises, which accelerates the droplet absorption process (curves 2 and 2' in Fig. 3). The fraction of the liquid absorbed by the second layer decreases with increasing thickness of the first layer. The curves of the droplet absorption time and the size of the wet spot versus the thickness of the first layer exhibit characteristic maxima (curves 2 and 2' in Figs. 3 – 4). With a further increase in the thickness of the first layer, the droplet absorption time and the area of the wet spot decrease (curves 2 and 2' in Figs. 3 – 4), approaching the values for droplet absorption by a semi-bounded media ($t_a/t_{L1} \rightarrow 1$ and $S/S_{L1} \rightarrow 1$).

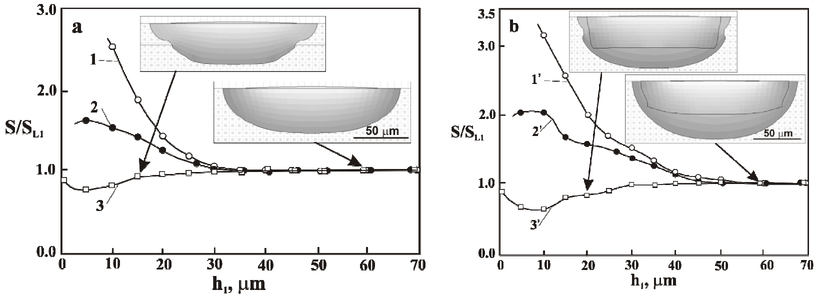

Fig. 4. Variation of area of spot formed at absorption of the first droplet (a) and first and second droplets (b) with the thickness of the first (upper) layer in the double–layer porous media for d_2 = 30 μm (curves 1 and 1'), 8 μm (curves 2 and 2'), and 1 μm (curves 3 and 3'); ε_1 = 0.2, d_1 = 2 μm, and ε_2 = 0.3.

The presence of the second layer with a pore size $d_2 < d_1$ accelerates the process of first and second droplets absorption (curves 3 and 3' in Fig. 3). For a small thickness of the first layer, the droplet absorption time is shorter than that in the case of droplet absorption by a semi-bounded media with parameters corresponding to those of the first porous layer ($t_a/t_{L1} < 1$). The second layer absorbs a considerable amount of the liquid and $S/S_{L1} < 1$. The effect of the second layer reduces with increasing thickness of the first layer. In this case, the durations of first and second droplets absorption and the spot areas increase (curves 3 and 3' in Figs. 3 – 4).

To highlight the practical possibilities of modeling predictions the effect of the structure porous media and relative location of droplets can be analyzed applying to problems of ink-jet printing. At ink-jet printing the every dot in the image is formed by sequential deposition of different color droplets. If the total size of spot formed by deposited droplets on the surface of media characterizes the resolution and local contrast of image then the ratio of differently colored areas in the spot defines the tone colour of dot. In the case of two droplets the ratio of areas S_1/S_2 can be used as parameter to characterize the tone colour of dot. Here S_1 is the part of total area of spot colored by liquid of the first droplet and S_2 is the part of spot area formed by liquid of second droplet. The effect of the thickness of the first layer of porous medium and pore size in the second layer on the value of parameter S_1/S_2 is illustrated in Fig. 5. It is seen that at all analyzed variations of porous medium structural parameters, the ratio of areas $S_1/S_2 < 1$. The smallest values of parameter S_1/S_2 are realized in the media the pore size in the second layer of which is smaller than the pore size in the first layer ($d_2 < d_1$). At small thickness of first layer the ratio of areas $S_1/S_2 << 1$ (curve 3 in Fig. 5).

The change in the parameter S_1/S_2 at variation of distance L between the centers of the first and second droplets location is shown in Fig. 6. The values of porous media structural parameters used in calculations are given in the Fig. 6 caption. The calculation data are plotted in the relative coordinates L/R_{d1} where R_{d1} is the radius of spot formed on the surface of porous medium by absorbed liquid of first droplet before absorption of second droplet. Curve 1 in Fig. 5 corresponds to the case of media a pore size in the second layer of which is much larger than the pore size in the first layer. For these media the absorption by the second layer is insignificant. The examples of droplets absorption by media with comparable pore size in the first and second layers and by semi-bounded media are presented in Fig. 6 by curves 2 and 3, respectively. As one would expect, at the large distances $L \rightarrow 2R_{d1}$ the value of parameter S_1/S_2 approaches 1. The second droplet is absorbed as a single droplet. At distances $L < 2R_{d1}$ the distribution of liquids depends on the ratio of structure parameters of the layers.

5. Discussion

The process of first droplet absorption by a double-layer porous media can be divided into two stages: the stage of liquid absorption by the first porous layer until the absorption front arrives at the interface between the layers and the stage in which the liquid flows in both layers (see for example Fig. 2). In the case of second droplet the process of absorption becomes complicated. The effect of relative location of the first and second droplets on absorption process takes place (see Fig. 6). If the distance between the centers of first and second droplets location $L \geq 2R_{d1}$ (where R_{d1} is the radius of spot formed at single droplet absorption) the process of second droplet absorption is similar to absorption of the first droplet. In the case of $L = 0$ (the case of axial symmetry) the region of porous media under the second droplet has been fully filled by the liquid of the first droplet. At intermediate values of distance L the contact boundary between liquids of first and second droplets is formed on the various stages of absorption. The symmetry of pressure field and as result the preferred directions of liquid flow in porous media are changed.

In the case of first droplet absorption if the pore size in the second layer is much larger than the pore size in the first layer ($d_2 >> d_1$), almost no absorption of the liquid by the second layer is observed. Having reached the interface between the layers, the liquid moves predominantly in the radial direction and fills the volume in the first layer. This case is characterized by the largest values of the absorption time and the size of the spot formed on the medium surface (Figs. 4–5). As the thickness of the first layer increases the absorption time and the area of spot decrease approaching to the values corresponding the case of absorption by a semi-bounded medium ($t_a/t_{L1} \rightarrow 1$ and $S/S_{L1} \rightarrow 1$).

Similar regularity takes place at absorption of the second droplet (curves 1' in Figs. 3 – 4). The liquid of second droplet forces the liquid of first droplet out to the periphery. As the thickness of the first layer increases the cross-sectional area of the first layer through which the liquid propagates in the radial direction also increases. This accelerates the absorption process. However, at some values of first layer thickness the rate of displacement of liquid under central part of droplet falls (see the inserts in Fig. 3b that illustrates the character of motion and distribution of liquid in porous medium). As result, the characteristic inflection on the curve of the second droplet absorption time versus the layer thickness is observed (curve 1' in Fig. 3).

As the pore size in the second layer decreases, the liquid flow due to the absorption by the second layer becomes substantial. The absorption time and the size of the spot formed on the porous surface decrease (Figs. 3 – 4). In the model, the effect of the second layer on the liquid flow depends on the permeability coefficient of the porous media (3) and on the pressure field, which, in turn, depends on the droplet surface tension forces (4) and the pressure jump at the absorption front (5). With reduction in the pore size in the second layer, the permeability coefficient decreases; this might be expected to decelerate the absorption process. Since the numerical experiment yields the opposite result (see Fig. 3), it can be concluded that the main factor responsible for the reduction in the absorption time with decreasing pore size in the second layer is an increase in the pressure jump at the absorption front.

As the pore size in the second layer decreases ($d_2 > d_1$ and $d_2 \sim d_1$), liquid flow to the second porous layer arises, which accelerates the droplet absorption process (curves 2 and 2' in Fig. 3). The effect of the second layer reduces with increasing thickness of the first layer. However, the fraction of the liquid absorbed by the first layer before the arrival of the absorption front at the interface between the layers increases, and this reduces the total absorption time. The cross-sectional area of the first layer through which the liquid propagates in the radial direction also increases; this also accelerates the absorption process. As a result, the curves of the first and second droplets absorption time and the wet spot size exhibit maxima (curves 2 and 2' in Figs. 3 – 4).

If the pore size in the second layer is smaller than the pore size in the first layer ($d_2 < d_1$), the pressure jump formed when the liquid reaches the second layer is greater than the pressure jump at the liquid propagation front in the first layer. As a result, having reached the interface between the layers, the liquid is absorbed primarily by the second layer. In this case, the time of absorption and the size of the wet spot are small ($t_a/t_{L1} < 1$ and $S/S_{L1} < 1$). The effect of the second layer reduces with increasing thickness of the first layer ($t_a/t_{L1} \rightarrow 1$ and $S/S_{L1} \rightarrow 1$).

When the distance between the first and second droplets location on the surface of porous media $L = 0$ the liquid of the first droplet is forced out in all directions by liquid of the second droplet. Therefore, the part of liquid of the first droplet is distributed in the medium under the layer of liquid of second droplet. As result the ratio of areas in the spot takes on values $S_1/S_2 < 1$ at all variations of structural parameters of porous media (see Fig. 5). At increasing the thickness of first layer the effect due to the second layer weakens. The largest values of parameter S_1/S_2 are achieved in the double-layer porous media with thin first layer and large pore sizes in the second layer (curve 1 in Fig. 5). In these media the penetration of liquid into a second layer is inconsiderable and the main part of liquid of the first droplet is forced out on the periphery. In the case of media in which the pore size in the second layer is smaller than the pore size in the first layer ($d_2 < d_1$), the considerable part of liquid of the first droplet overflows into second layer. The parameter S_1/S_2 can take on values close to 0 (curve 3 in Fig. 5).

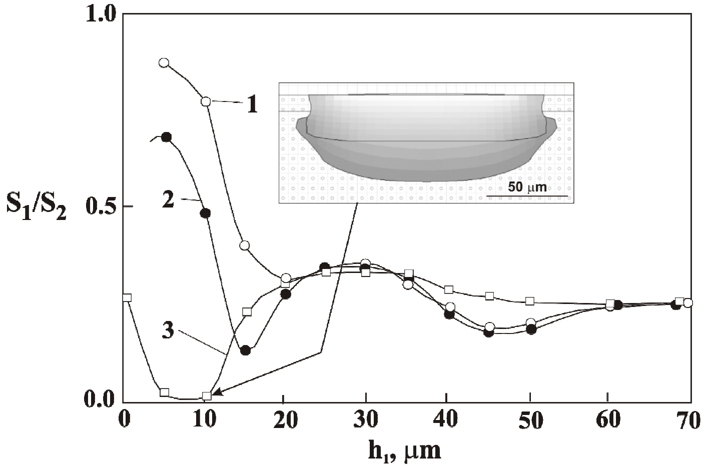

Fig. 5. The ratio of areas S_1/S_2 in the spot formed by liquids of first (S_1) and second (S_2) droplets plotted against the thickness of the first layer in the double–layer porous media for $d_2 = 30$ μm (curve 1), 8 μm (curve 2), and 1 μm (curve 3); $\varepsilon_1 = 0.2$, $d_1 = 2$ μm, and $\varepsilon_2 = 0.3$. The distance $L = 0$.

At the distances between the first and second droplets location on a surface of porous media $L > 0$ the liquid of second droplet may fully or partially contact with the liquid of the first droplet initially distributed in the porous media. In any case, the axial symmetry of arising pressure gradients is disturbed that leads to change of preferred directions of liquid flow. At increasing the distance between the droplets location the contact boundary is formed on the later stages of second droplet absorption. The process of second droplet absorption becomes similar to absorption of the first (single) droplet. The ratio of areas S_1/S_2 approaches 1. However, this tendency has non-monotonic character and depends appreciably on the ratio of the pore size in the layers (see Fig. 6). In particular if the pore size in the second layer $d_2 \sim d_1$ (or $d_2 < d_1$) the ratio of areas S_1/S_2 can take to values greater than 1 (see the curve 2 in Fig. 6). The significant part of liquid of the second droplet is absorbed by the second layer of porous medium.

6. Conclusions

A three-dimensional model of sequential absorption of two droplets at its arbitrary location on the surface of double-layer porous media was considered. The numerical experiments based on developed model were performed to analyze the effect of the porous media structural parameters on the rate of droplets absorption and distribution of absorbed liquids. As an example of practical application the effect of relative location of two droplets on the distribution of absorbed liquids was analyzed with reference to the problems of inkjet-printing. The ratio of differently colored areas in the spot formed at absorption of two droplets was used as the parameter characterizing the local color transfer.

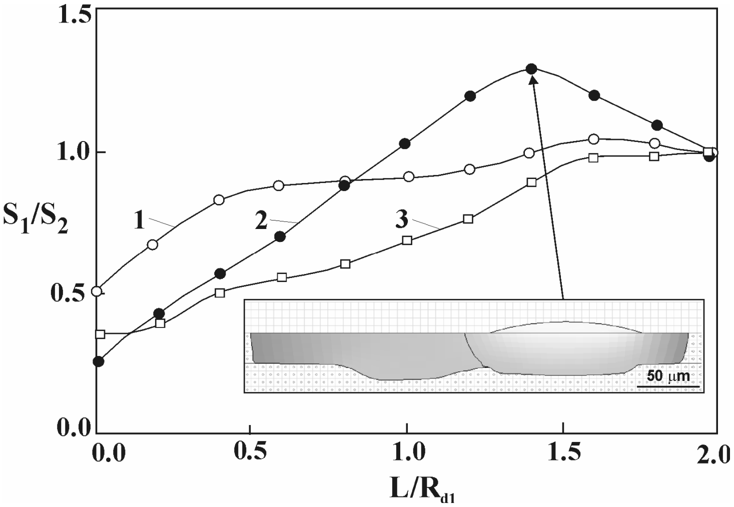

Fig. 6. The ratio of areas S_1/S_2 in the spot formed at sequential absorption of liquids of first (S_1) and second (S_2) droplets plotted against the distance L between the centers of droplets location. The structure parameters of porous media: $d_1 = 1$ μm, $\varepsilon_1 = 0.15$, $\varepsilon_2 = 0.3$, $h_2 = 100$ μm; curve 1: $h_1 = 25$ μm, $d_2 = 30$ μm; curve 2: $h_1 = 25$ μm $d_2 = 2$ μm; curve 3: $h_1 = 100$ μm.

The results of numerical study have indicated that the presence of the second layer could lead to a significant change in the liquid distribution in the medium. The main parameter governing the effect of the second layer is the pore size, which determines the capillary forces acting at the absorption front boundary and the pressure field in the liquid. For media with a pore size in the second layer much larger than the pore size in the first layer, the absorption by the second layer is insignificant. Such media are characterized by the largest droplet absorption time and the largest size of the wet spots formed on the surface. The shortest duration of droplet absorption and the smallest spot size are observed for media with a pore size in the second layer smaller than the pore size in the first layer. For media with a pore size in the second layer larger than but comparable to the pore size in the first layer the curves of the droplet absorption time and the wet-spot size versus the thickness of the first layer are found to have characteristic maxima.

This study shows that the dependency of the parameter characterizing the local color transfer versus the distance of relative location of the droplets is non-monotonic and depends on the ratio of the pore size in the layers. At small distances the ratio of differently colored areas in the spot is less than 1. The largest values of the parameter characterizing the local color transfer are achieved in the media with thin first layer and large pore size in the second layer. For media with a pore size in the second layer smaller than the pore size in the first layer the ratio of differently colored areas in the spot can take on values close to 0.

At large distances of relative location of the droplets the process of second droplet absorption becomes similar to absorption of the single droplet. The ratio of differently colored areas in the spot approaches 1. At intermediate values of distance the contact boundary between liquids of first and second droplets is formed on various stages of second droplet absorption. The symmetry of pressure field and as result the preferred directions of liquid flow in porous media are changed. For media with a pore size in the second layer comparable to or smaller than the pore size in the first layer the ratio of differently colored areas in the spot can take values greater than 1.

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