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ABSORPTION OF MICRODROPLETS BY LAYERED POROUS MEDIA

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ABSTRACT

The subject of this paper is the sequential absorption of two droplets at its arbitrary location on the surface of single- and double-layer porous media. The consideration is based on simultaneous solution of the equations describing liquid flow in the droplet and equations of unsteady filtration in the porous medium. 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 and the propagation of absorbed fluid in a porous medium 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.

INTRODUCTION

The absorption of small size drops by porous media serves as basic process of wide range of technologies such as high-quality inkjet-printing, spray-painting and coating (see for example the publications [1-9]). An important feature of the microdrop absorption is the presence of free and contact surfaces. In most applications the porous media have the various structures including multilayer structure with different permeability of layers. Both spray coating and printing processes involve the stage of sequential deposition of fine droplets onto the surface of porous media. The every subsequent droplet is deposited onto dry part of surface or surface saturated by liquid of previously deposited droplet. Adequate understanding these processes is essential to determining the relation between the structure of porous media, process parameters and qualitative adjectives of formed coatings.

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 & 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 & 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.

NOMENCLATURE

Bo		Bond number
d	[m]	Pore size
Fo		Fourier number
h	[m]	Thickness of porous layer
k	[m^2]	Permeability coefficient
L	[m]	Distance between the centers of droplets location
p	[Pa]	Pressure
R	[m]	Radii
S	[m^2]	Area of formed wet spot
t	[s]	Time
V	[m^3]	Volume of the droplet
$u, v.w$	[m/s]	Velocity components
Special characters		
ε		Porosity
η	[Pa·s]	Dynamic viscosity
θ	[grad]	Contact angle
ρ	[kg/m^3]	Density
σ	[N/m]	Surface tension coefficient
Subscripts		
a		Absorption
d		Droplet
L		Layer
1		First layer or droplet
2		Second layer or droplet
o		Initial

In our previous work [10] a two-dimensional model was considered to describe the absorption of single droplet by double-layer porous media.

The present work focuses on numerical simulation of the sequential absorption of two microdrops by porous media. A three-dimensional model is developed to describe the absorption of the droplets at its arbitrary location on the surface of porous media. The effect of the structure parameters of porous media (pore size in the layers and the thickness and porosity of the layers) and the relative location of droplets on the duration of absorption and distribution of absorbed liquids is analyzed using the numerical experiment. The practical possibilities of developed model are illustrated on the example of ink jet-printing processes [4, 11 - 13].

FORMULATION OF THE PROBLEM

The case when a pore size in the upper layer is smaller than the droplet size is considered. It 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.

For molecules of liquid the Fourier number Fo <<1. Therefore, it is supposed that the liquids of the first and second droplets not mix during absorption. For the droplets with size $a \sim 10^{-4}$ m the Bond number Bo << 1, therefore the gravitational effects are negligible. For low viscous liquids ($\eta < 10^{-2}$ Pa·s) the penetration of liquid into porous medium is the defining slow process. The spreading of droplet over surface of porous medium, which results in a change of droplet shape (the size of droplet base and curvature of free surface) follows quickly. Thus, the approaching of inviscid liquid flow can be used to describe the motion of liquid in the droplet during absorption. 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 [17]:

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

where $d/dt = \partial/\partial t + u\partial/\partial x + v\partial/\partial y + w\partial/\partial z$, u, v and w are the flow velocity components, p is the pressure, and ρ is the liquid density.

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 [15, 16]:

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

Here $d/dt = \partial/\partial t + \varepsilon^{-1}(u\partial/\partial x + v\partial/\partial y + w\partial/\partial z)$, η is the dynamic viscosity of the liquid, k is the permeability coefficient of the medium, u, v and w are the filtration velocity components and ε is the porosity of medium. Following a study of Scheidegger [18], which considers experimental data for liquid flows in porous media of various structures, we use the following expression for the permeability coefficient:

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

where d is the pore size of medium. At the free surface of a droplet a jump in the normal stress p_d due to capillary forces is imposed:

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

where R_1 and R_2 are the principal curvature radii of the droplet surface. 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 nonpenetration 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

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

where θ is the contact angle.

The initial droplet shape (a spherical segment) is characterized by height and base diameter. 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 equation (4), and the initial pressure in the porous medium region with the absorbed liquid under the droplet was set equal to p_p, in accordance with equation (5).

Depending on the distance L between the centers of first and second droplets location, 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. 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. Initially, each body is a body of revolution. The numerical solution of the problem was based on the use of the moving-mesh procedure [19].

MODEL VERIFICATION

For the case of single droplet absorption the validation of developed model was performed in the work [10]. 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). The difference between the experimental and calculated data not exceeds 10%.

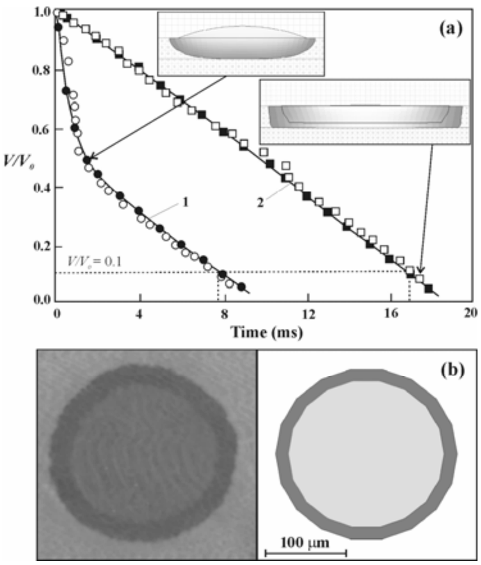

Figure 1 Comparison of calculation results with experimentally obtained data for the sequential absorption of two water droplets of volume $V_o = 0.9 \cdot 10^{-13}$ m^3 by a double–layer paper (h_1 = 20 µm, ε_1 = 0.2 and d_1 = 3.5 µm):
(a) the temporal evolution of the volume $V(t)/V_o$ of the first (1) and second (2) droplets during its absorption: the open symbols - experimental data and the solid symbols - calculation. The distance between droplets location L = 0. Inserts illustrate the distribution of liquid in the porous media in the time moment when absorption front of first droplet arrives at the interface between the layers and when the absorption of two droplets takes place;
(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).

Analogously experiments were carried out to validate the model as applied to the sequential absorption of two droplets. A technique of the flash videography and subsequent reconstruction of stages of fast repeated processes with a time resolution of 10 µs was used [10]. Droplets about 50 µm in diameter were deposited sequentially onto double-layer paper using the inkjet printer cartridges. 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 [10]. 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. From the data in Figure 1, it follows that the predicted the droplet volume evolution and the size of the wet spot formed on the surface of paper are in good agreement with the experimental data.

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 (d_1 and d_2), porosity (ε_1 and ε_2), and the thickness of the upper layer (h_1) in the porous media. Here the subscripts 1 and 2 indicate the number of the layer (1 for the first (upper) layer and 2 for the second layer). The absorption process and the distribution of the absorbed liquid in the porous media were characterized by the time t_a required for adsorption of 90% of the droplet volume (see Figure 1(a)) and the area S of the wet spot formed on the porous surface by the absorbed liquid.

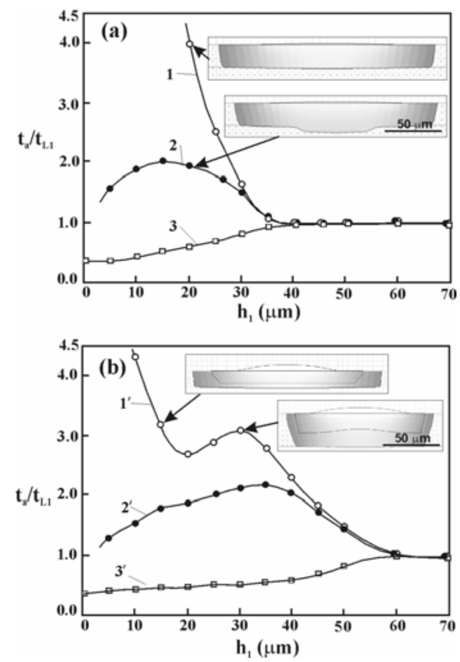

Figure 2 Variation of duration of first (a) and second (b) droplets absorption with the thickness of the first (upper) layer in the double–layer porous media for d_2 = 30 µm (curves 1 and 1'), 8 µm (curves 2 and 2'), and 1 µm (curves 3 and 3'); ε_1 = 0.2, d_1 = 2 µm, and ε_2 = 0.3. The distance between the centers of droplets location L = 0. Inserts illustrate the realizable distributions of liquid in the porous media.

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 Figure 2. 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 Figure 3. The inserts in Figures 2 and 3 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 spot that correspond to the limiting case of droplet absorption by a semibounded media with parameters equal to those in the first layer. 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 Figures 2 – 3).

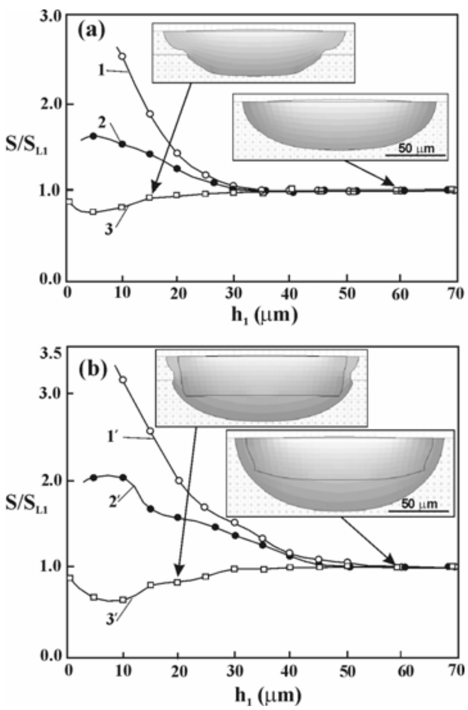

Figure 3 Variation of area of spot formed at absorption of the first droplet (a) and first and second droplets (b) with the thickness of the first (upper) layer in the double–layer porous media for d_2 = 30 μm (curves 1 and 1'), 8 μm (curves 2 and 2'), and 1 μm (curves 3 and 3'); ε_1 = 0.2, d_1 = 2 μm, and ε_2 = 0.3. The distance between the centers of first and second droplets location L = 0. Inserts illustrate the realizable distributions of liquid in the porous media.

At ink-jet printing the every dot in the image is formed by sequential deposition of different color droplets. 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 Figure 4. 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 Figure 5. 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.

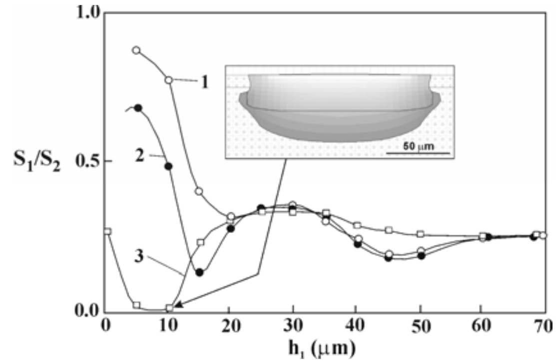

Figure 4 The ratio of areas S_1/S_2 in the spot formed by liquids of first (S_1) and second (S_2) droplets plotted against the thickness of the first layer in the double–layer porous media for d_2 = 30 μm (curve 1), 8 μm (curve 2), and 1 μm (curve 3); ε_1 = 0.2, d_1 = 2 μm, and ε_2 = 0.3. The distance between the centers of first and second droplets location L = 0. Inserts illustrate the realizable distributions of liquid in the porous media.

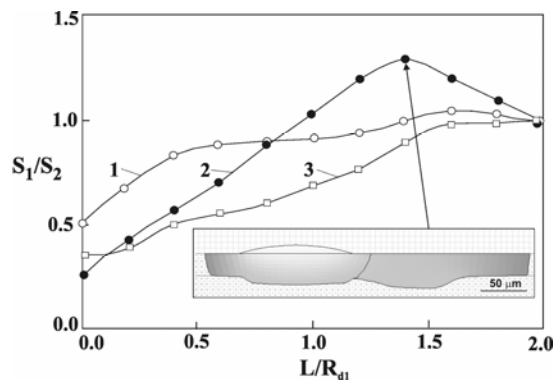

Figure 5 The ratio of areas S_1/S_2 in the spot formed at sequential absorption of liquids of first (S_1) and second (S_2) droplets plotted against the distance L between the centers of droplets location on surface of porous media. The structure parameters of porous media: d_1 = 1 μm, ε_1 = 0.15, ε_2 = 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. Inserts illustrate the realizable distributions of liquid in the porous media.

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. In the case of second droplet the effect of relative location of the first and second droplets on absorption process takes place (see Figure 5). 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.

The data (see Figures 2–4) show that the preferred directions and the rate of liquid propagation realized in the second stage of first droplet absorption and at absorption of second droplet depend appreciably on the ratio of the structural parameters of the layers. In the case of first droplet absorption if $d_2 >> d_1$, 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 (see Figures 2–3). An increase in the thickness of the first layer leads, on the one hand, to an increase in the fraction of the liquid absorbed by the porous medium before the arrival of the absorption front at the interface between the layers, and, on the other hand, it results in an increase in the cross-sectional area of the first layer through which the liquid propagates in the radial direction. Both factors reduce the droplet absorption time and the wet-spot area (curves 1 in Figures 2–3). The absorption time and the area of spot decrease approaching to the values corresponding the case of absorption by a semibounded medium.

Similar regularity takes place at absorption of the second droplet (curves 1' in Figures 2–3). 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 Figure 2(b)). As result, the characteristic inflection on the curve of the second droplet absorption time versus the layer thickness is observed (curve 1' in Figure 2).

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 (see Figures 2–3). In the model, the effect of the second layer on the liquid flow depends on the permeability coefficient of the porous media (equation (3)) and on the pressure field, which, in turn, depends on the droplet surface tension forces (see equation (4)) and the pressure jump at the absorption front (equation (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 Figure 2), 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 Figure 2). 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 Figures 2–3).

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 first and second droplets absorption time and the size of the wet spot are smaller (curves 3 and 3' in Figures 2–3) than those in the case of droplets absorption by semibounded medium ($t_a/t_{L1} < 1$ and $S/S_{L1} < 1$). Since the increase in the thickness of the first layer weakens the effect due to the second layer, the absorption time and the area of the wet spot increase ($t_a/t_{L1} \to 1$ and $S/S_{L1} \to 1$).

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 Figure 4). At increasing the thickness of first layer the effect due to the second layer weakens. The largest values of this parameter are achieved in the double-layer porous media with thin first layer and large pore sizes in the second layer (curve 1 in Figure 4). The penetration of liquid into a second layer is inconsiderable. 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 Figure 4).

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. 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 nonmonotonic character and depends appreciably on the ratio of the pore size in the layers (see Figure 5). 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 Figure 5).

CONCLUSION

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 were performed to analyze the effect of the porous media structural parameters on the rate of droplets absorption and distribution of absorbed liquids.

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. The liquid of first and second droplets fills the space in the first layer. 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. In those media, the second layer absorbs the liquid having reached the interface between the layers, predominantly. At absorption of second droplet the considerable part of liquid of the first droplet overflows into second 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 nonmonotonic and depends on the ratio of the pore size in the layers. At small distances of relative location of the droplets the contact boundary between liquids of first and second droplets is formed on initial stage of second droplet absorption. The absorption of the second droplet is determined by displacement from the pores the liquid of first droplet by liquid of second droplet. At all analyzed variations of structural parameters of porous media 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 to a values greater then 1.

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