

MICRODROPLET ABSORPTION BY TWO-LAYER POROUS MEDIA

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Microdroplet absorption by two-layer porous media is studied both theoretically and experimentally. A two-dimensional model for liquid flow from a droplet into a porous medium is presented and verified based on a simultaneous numerical solution of the Euler equations taking into account surface tension forces and the unsteady filtration equation. The effect of the structural parameters of the two-layer porous medium (pore size in the layers, and the thickness and porosity of the layers) on the droplet absorption is analyzed. It is shown that the presence of the second layer can have a significant effect on the droplet absorption rate and the liquid distribution in the medium. The pore size is found to be the main parameter that governs the effect of the second layer.

Key words: microdroplets, porous media, absorption.

Introduction. In studies of the various aspects of liquid transport in porous media, considerable recent attention has been focused on the absorption of small liquid droplets by one- and multi-layer porous media [1–3]. Interest in this problem has been stimulated by the development of high-quality inkjet printing [4, 5] and coating technologies [6].

An important feature of the process is the presence of free and contact surfaces [4–9]. Capillary spreading and the tendency of droplets to minimize the free surface area influence the droplet shape during absorption and the duration and final result of the process. Starov et al. [7] proposed an analytical model for liquid flow from a droplet to a thin porous bed. Davis and Hocking [5] considered a two-dimensional model in which the porous medium was modeled by alternating vertical slits permitting liquid flow only in the vertical direction. Alleborn and Raszillier [8] performed a numerical study of the effect of the permeability of the porous medium on droplet absorption in the case of two-layer media with identical pore sizes in the layers.

The present paper considers a two-dimensional model in which the layers of a two-layer porous medium are characterized by effective permeability coefficients dependent on porosity and pore size. Liquid flow in a porous medium is described by the unsteady filtration equation for an incompressible liquid. Liquid flow in a droplet on the surface of a porous medium is modeled by solving the Euler equations for an incompressible liquid taking into account surface tension forces. The numerical calculations were checked by comparing with experimental data on droplet absorption by porous media. The effect of the structural parameters of the two-layer porous medium (pore size in the layers and the thickness and porosity of the layers) on the duration of droplet absorption and on the liquid distribution in the porous medium is analyzed.

1. Formulation of the Problem. The subject of the present study is the absorption of a liquid droplet by one- or two-layer media with a pore size in the upper layer smaller than the droplet size. Initially, the droplet is assumed to be shaped like a spherical segment. The subsequent evolution of the droplet shape is governed by the capillary spreading, the tendency of the drop to minimize the free surface area due to surface tension forces,

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and droplet absorption. The indicated processes are modeled by simultaneous solution of the equations describing liquid flow in the droplet and in the porous medium.

1.1. *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 liquid and the continuity equation [10]. In the axial-symmetry approximation, these equations are written as

$$\frac{du}{dt} = -\frac{1}{\rho} \frac{\partial p}{\partial z}, \quad \frac{dv}{dt} = -\frac{1}{\rho} \frac{\partial p}{\partial r}, \quad \frac{\partial u}{\partial z} + \frac{\partial v}{\partial r} = -\frac{v}{r} \quad (1)$$

(u and v 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, the Darcy law is satisfied for steady-state flows (the flow velocity in the pores is directly proportional to the pressure gradient) [2, 3]. The proportionality coefficient is the filtration coefficient $k = k_0/\eta$, which is determined by the permeability coefficient of the medium k_0 (which depends on the porosity ε and the pore size d) and by the dynamic viscosity of the liquid η . Following a study [1], which considers experimental data for liquid flows in porous media of various structures, we use the following expression for the permeability coefficient:

$$k_0 = d^2 \varepsilon^3 / [150(1 - \varepsilon)^2]. \quad (2)$$

Under the adopted assumptions, the liquid flow in the porous medium is described by the equations of unsteady filtration of an incompressible liquid [2, 3], which in the axisymmetric case have the form

$$\frac{du}{dt} = -\frac{1}{\rho} \frac{\partial p}{\partial z} - \frac{1}{k} u, \quad \frac{dv}{dt} = -\frac{1}{\rho} \frac{\partial p}{\partial r} - \frac{1}{k} v. \quad (3)$$

Here $d/dt = \partial/\partial t + (u \partial/\partial z + v \partial/\partial r)/\varepsilon$ and u and v are the filtration velocity components.

1.2. *Boundary Conditions.* At the free surface of the droplet, the following boundary conditions are imposed: zero shear stress and a jump in the normal stress p_d due to capillary forces:

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

(σ is the surface tension force and R_1 and R_2 are the principal curvature radii of the droplet surface).

At the droplet–porous medium interface, the condition of equality of the normal flow velocities in the droplet and in the porous medium is used. At the free surface of the porous medium occupied by the absorbed liquid, the nonpenetration condition is imposed, which implies zero normal velocity of the liquid flow in the porous medium.

The action of capillary forces at the absorption-front boundary (between the region of the porous medium filled with the liquid and the region free of the liquid) is characterized by the pressure discontinuity given by the Laplace formula [11]

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

(θ is the contact wetting angle).

1.3. *Initial Conditions.* The initial droplet shape (a spherical segment) is characterized by height and base diameter, from which the wetting angle for the substrate material can be determined [12]. 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 was set equal to p_p , in accordance with Eq. (5).

1.4. *Numerical Methods and Procedures.* The numerical solution of the problem was based on the use of the well-known moving-mesh procedure [13]. System (1), (3) was integrated using an explicit conservative scheme of first-order accuracy $O(\tau + h)$. In the construction of the finite-difference approximation, Eqs. (1) and (3) were converted for use in the moving-mesh procedure employing the method proposed and tested in [14]. New values of u , v , 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 boundaries defining the droplet shape on the surface of the medium and the advance of the absorption front. Once the liquid in the porous medium reached the free surface, the boundary condition given by (5) was replaced

by the nonpenetration condition. When the liquid reached the interface between the layers in the case of a two-layer medium, the pressure jump at the absorption-front boundary was changed [see Eq. (5)], and the permeability coefficient was adjusted to the layer parameters [see Eq. (2)]. As a result of the numerical solution, we obtained the evolution of the droplet shape in the absorption process, the liquid distribution in the porous medium, and the flow-velocity and pressure fields.

2. Experimental. The numerical results were verified by comparing with data of experiments with absorption of droplets deposited on one- and two-layer porous media. The experiments were carried out using stroboscopic imaging of stages of fast processes with their subsequent reconstruction [15]. The imaging was performed with a digital video-camera equipped with a microscope lens and a stroboscopic system with a light-pulse duration of 1.5 μsec .

Droplets about 1 mm in diameter were deposited by forcing the liquid through a capillary tube with an inner diameter of 0.2 mm. For the deposition of fine droplets (50 to 60 μm in size), an inkjet printer cartridge was used. The different stages of the process were identified by varying the time interval between the moment of droplet ejection and the stroboscopic flash. The minimum time interval was 10 μsec . The measured geometrical sizes of droplets were used to calculate the droplet volume versus time during the absorption process. The measurement error for the droplet base diameter and for the droplet height was 2 μm for fine droplets and 20 μm for droplets with a size of about 1 mm.

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%.

Droplets with a size of about 1 mm were deposited onto standard glass filters (nominal porosity class F, pore size 4.0 to 5.5 μm). The filter porosity $\varepsilon \approx 0.4$ was evaluated from the increase in the filter mass after immersion of the filter into water. Droplets of size 50 to 60 μm were deposited onto a two-layer paper. Optical microscopic measurements showed that the upper layer of the paper of thickness $h_1 \approx 25 \mu\text{m}$ was a porous medium consisting of grains with a size of 5 to 10 μm . The second layer of thickness $h_2 \approx 80 \mu\text{m}$ had a fibrous structure with void sizes ranging from 10 to 20 μm . During the deposition of fine droplets ($V_0 < 10^{-12} \text{ m}^3$), noticeable absorption of the liquid by the second paper layer was not observed within the microscope resolution of 2 μm . The liquid was distributed over the volume of the first paper layer, and this was taken into account in evaluating the porosity of this layer. From the results of measurements of the initial droplet volume before absorption, the diameter of the spot formed on the paper surface during absorption, and the thickness of the upper paper layer, the porosity was found to be $\varepsilon_1 \approx 0.2$.

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 simulation results were compared with the experimental data on droplet 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 medium, and the properties of the liquid. The wetting angle in the pores θ was set equal to the “steady-state” contact angle for the droplet on the surface, which, in turn, was determined from the height and the base diameter of the droplet. The varied parameter was the pore size. The results of numerical simulations for the absorption time and the size of the spot formed on the surface of the porous medium during absorption were compared with the experimental data. The difference between the experimental and calculated data not exceeding 10% was considered satisfactory.

Figure 1 shows experimental and calculated data for droplet absorption by the glass filter: the variation of the droplet shape during absorption, the absorption-front propagation, and the variation of the droplet volume $V(t)/V_0$. The experiment consisted of ten series of measurements, in which droplets with controlled volume were deposited onto various regions of the surface of the same filter. As an example, Fig. 1b shows data from three series of measurements that illustrate the largest spread in the experimental droplet volumes at specified times; these data are compared with calculation results for three values of the pore size (curves 1–3). The calculated values (curve 2) are seen to agree with the experimental data, and the obtained pore size $d = 4.5 \mu\text{m}$ is in the range of the pore sizes typical of the filter used (4.0–5.5 μm).

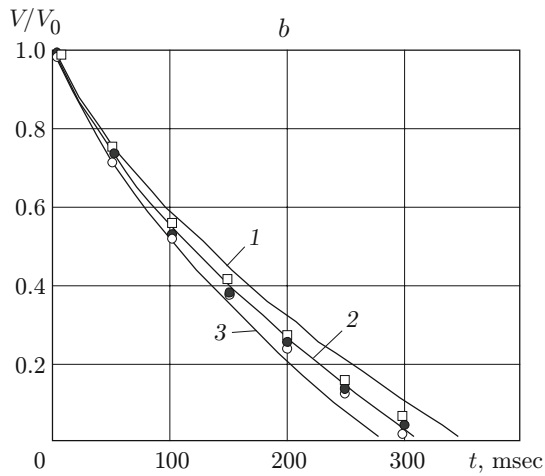
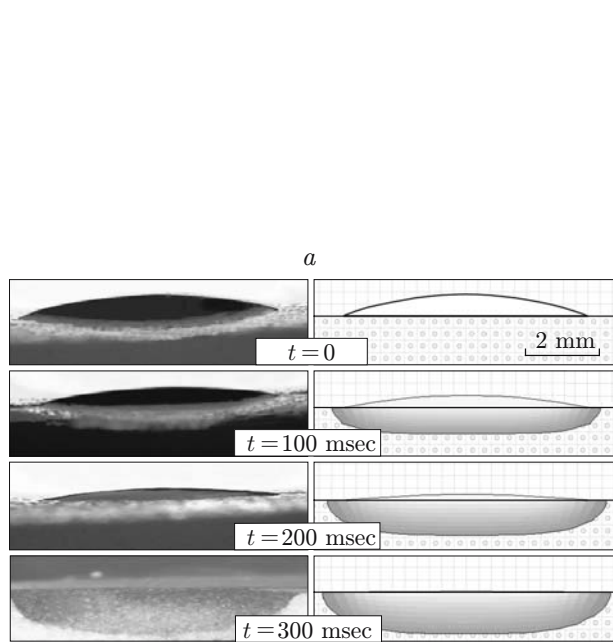


Fig. 1

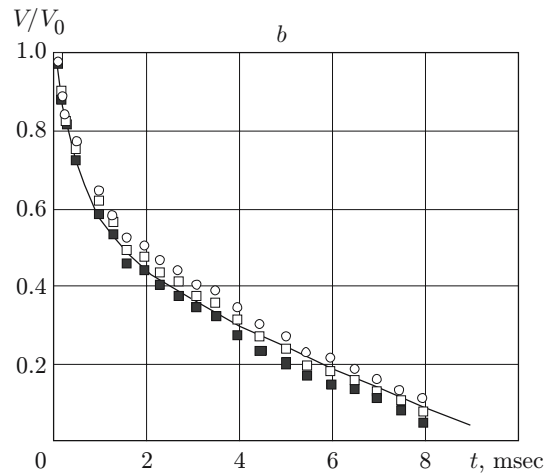
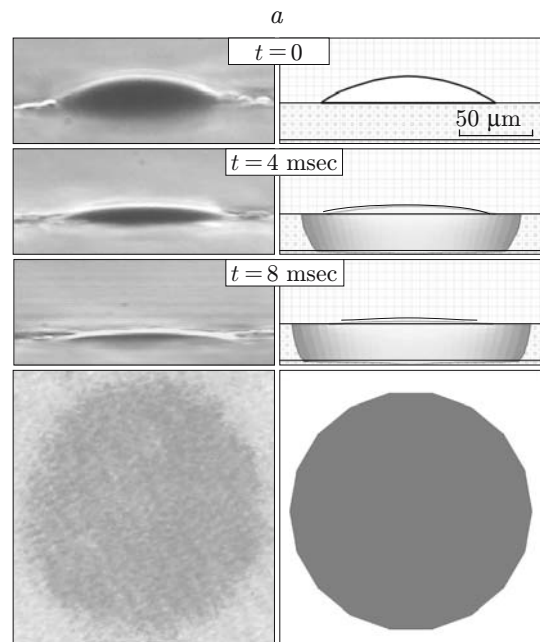


Fig. 2

Fig. 1. Absorption of a water droplet of volume $V_0 = 1.1 \cdot 10^{-8} \text{ m}^3$ by a glass filter ($\varepsilon = 0.4$): (a) variation of the droplet shape and the porous region filled with water during the absorption process: experimental data (on the left) and simulation data for $d = 4.5 \text{ } \mu\text{m}$ (on the right); (b) variation in the droplet volume $V(t)$ during absorption; the points and curves refer to the experimental and calculated data, respectively, obtained for $d = 4$, 4.5 (2), and $5 \text{ } \mu\text{m}$ (3).

Fig. 2. Absorption of a water droplet of volume $V_0 = 0.9 \cdot 10^{-13} \text{ m}^3$ by a two-layer paper ($\varepsilon_1 = 0.2$ and $d_1 = 3.5 \text{ } \mu\text{m}$): (a) variation of the droplet shape during absorption and the spot formed by the absorbed water on the paper surface: experimental data (on the left) and calculated data (on the right); (b) variation of the droplet volume $V(t)$ during absorption: the points refer to experimental data from three series of measurements and the curve refers to the calculated data.

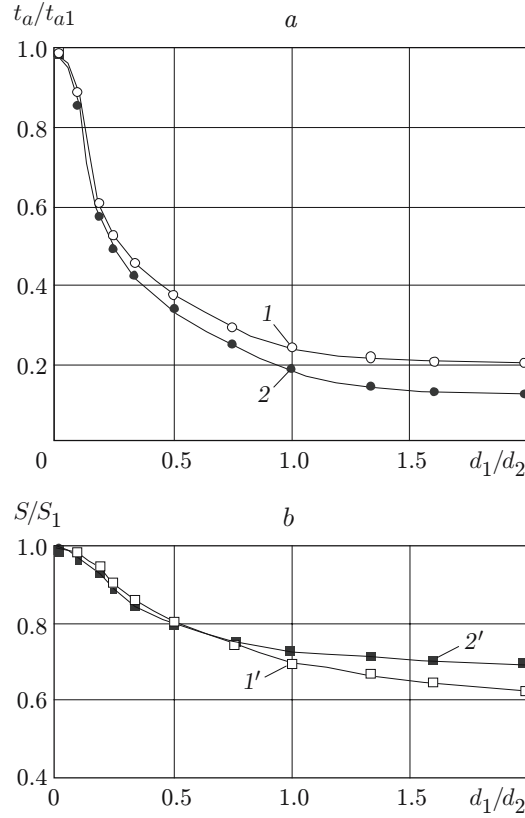


Fig. 3. Droplet absorption time (a) and the area of the wet spot (b) versus the ratio of the pore sizes d_1/d_2 in the first and the second layer of the porous medium for $\varepsilon_2 = 0.2$ (1 and 1') and 0.4 (2 and 2'); $V_0 = 10^{-13} \text{ m}^3$, $h_1 = 20 \text{ }\mu\text{m}$, $d_1 = 2 \text{ }\mu\text{m}$, and $\varepsilon_1 = 0.2$.

Figure 2 shows experimental and simulation data for absorption of water droplets of volume $V_0 = 0.9 \times 10^{-13} \text{ m}^3$ by the two-layer paper. The data of Fig. 2b on the variation of the droplet volume during absorption obtained in independent experiments with droplet deposition onto various regions of the test paper illustrate the effect of the structural inhomogeneity of the porous medium on droplet absorption. From the data in Fig. 2, it follows that the predicted time variation of the droplet shape, the size of the wet spot formed (Fig. 2a), and the droplet volume during absorption (Fig. 2b) 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 $d_1 = 3.5 \text{ }\mu\text{m}$ agrees with the data of optical microscopic measurements of the void sizes between paper grains (2–6 μm). The calculation data show that if the pores in the second layer have sizes $d_2 > 10 \text{ }\mu\text{m}$, the liquid does not penetrate into this layer, which is confirmed experimentally.

3. Results of Numerical Experiments. Numerical experiments were carried out to examine the effect of the structure of the two-layer porous medium on the absorption of small droplets. The volume of the test droplet was $V_0 = 10^{-13} \text{ m}^3$. For the materials in the first and second layers, the edge wetting 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 medium. The absorption process and the distribution of the absorbed liquid in the porous medium were characterized by the time t_a required for adsorption of 90% of the droplet volume and the area S of the wet spot formed on the porous surface by the absorbed liquid.

Figure 3 shows the droplet absorption time t_a (curves 1 and 2) and the area of the wet spot S (curves 1' and 2') versus the ratio of the pore sizes in the first and second porous layer d_1/d_2 . The results are plotted in the relative coordinates t_a/t_{a1} , S/S_1 , and d_1/d_2 (t_{a1} and S_1 are the droplet absorption time and the area of the wet spot, respectively, formed during liquid absorption only in the first porous layer of thickness h_1).

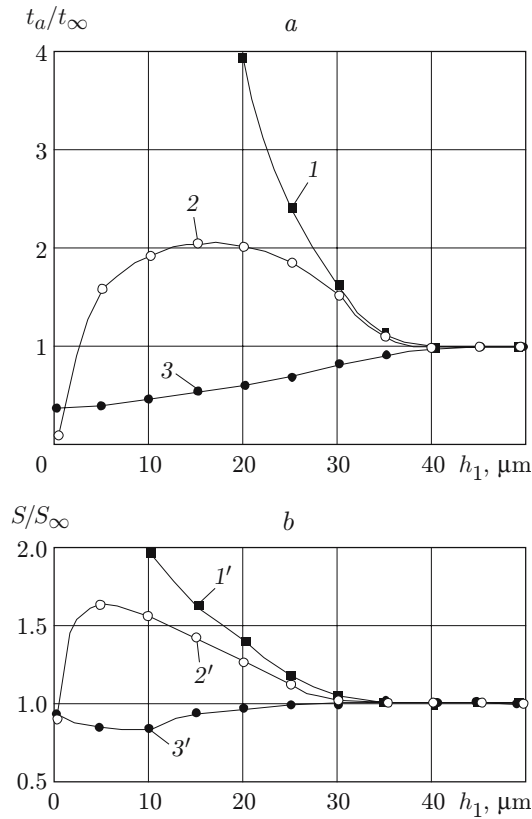


Fig. 4. Droplet absorption time (a) and the wet-spot area (b) versus the thickness of the upper layer in the two-layer porous medium for $d_2 = 30$ (1 and 1'), 8 (2 and 2'), and 1 μm (3 and 3'); $\varepsilon_1 = 0.2$, $d_1 = 2 \mu\text{m}$, and $\varepsilon_2 = 0.3$.

It is seen from Fig. 3 that as the pore size in the second layer d_2 decrease, the absorption time (curves 1 and 2) and the size of the wet spot formed on the surface (curves 1' and 2') also decrease. An increase in the porosity of the second layer also results in decreased absorption time (curves 1 and 2).

The effect of the thickness of the first layer on the duration of droplet absorption by the porous medium is illustrated in Fig. 4. The calculation data are plotted in the relative coordinates t_a/t_∞ and S/S_∞ (t_∞ and S_∞ are the total time of droplet absorption and the area of the wet spot formed during liquid absorption by the first porous layer whose thickness is greater than the absorption length). Under these conditions, the absorption front does not reach the interface between the layers, and the second layer does not influence the droplet absorption process. This is the limiting case of droplet absorption by a semibounded medium with parameters equal to those in the first layer. For zero thickness of the first layer, we have the second limiting case of droplet absorption by a semibounded medium with parameters corresponding to those of the second porous layer.

For intermediate thicknesses of the first porous layer, the droplet absorption time and the liquid distribution in the medium depend appreciably on the ratio of the structural parameters of the layers (Fig. 4). For large pore sizes in the second layer ($d_2 \gg d_1$), almost no absorption by the second layer is observed, as is indicated by the large sizes of the spot formed on the surface of the porous medium (curve 1' in Fig. 4). The process is characterized by long absorption times (curve 1 in Fig. 4). As the thickness of the layer increases, the droplet absorption time and the area of the wet spot decrease (curves 1 and 1' in Fig. 4), approaching the values corresponding to the case of droplet absorption by a semibounded medium ($t_a/t_\infty \rightarrow 1$ and $S/S_\infty \rightarrow 1$).

As the pore size in the second layer decreases ($d_2 > d_1$ and $d_2 \approx d_1$), a liquid flow to the second porous layer arises, which accelerates the droplet absorption process (curve 2 in Fig. 4). 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. 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 Fig. 4), approaching the values for droplet absorption by a semibounded medium ($t_a/t_\infty \rightarrow 1$ and $S/S_\infty \rightarrow 1$).

The presence of the second layer with a pore size $d_2 < d_1$ accelerates the droplet absorption process (curves 3 and 3' in Fig. 4). For a small thickness of the first layer, the droplet absorption time is shorter than that in the case of droplet absorption by a semibounded medium with parameters corresponding to those of the first porous layer ($t_a/t_\infty < 1$). The second layer absorbs a considerable amount of the liquid, and $S/S_\infty < 1$. The effect of the second layer reduces with increasing thickness of the first layer. In this case, the absorption time and the spot area increase.

4. Discussion of Numerical Results. The process of droplet absorption by a two-layer porous medium 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. The results of the numerical experiments (see Figs. 3 and 4) show that the processes occurring in the second stage depend appreciably on the ratio of the structural characteristics of the layers.

If the pore size in the second layer is much larger than the pore size in the first layer ($d_2 \gg d_1$), 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 corresponds to the largest values of the absorption time and the size of the spot formed on the porous surface (see Fig. 3); these values far exceeds the values for the case of droplet absorption by a semibounded medium with parameters equal to those of the first layer (curves 1 and 1' in Fig. 4).

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 Fig. 3). In the model [see Eqs. (2), (4), and (5)], the effect of the second layer on the liquid flow depends on the permeability coefficient of the porous medium and on the pressure field, which, in turn, depends on the droplet surface tension forces and the pressure jump at the absorption front. With reduction in the pore size in the second layer, the permeability coefficient [see Eq. (2)] 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.

The effect of the permeability coefficient of the second porous layer on droplet adsorption can be evaluated by comparing results of calculations for various porosities of the second layer (curves 1 and 2 in Fig. 3). For instance, for a pore size in the second layer $d_2 = 4 \mu\text{m}$, a two-fold increase in the porosity (from $\varepsilon_2 = 0.2$ to $\varepsilon_2 = 0.4$) results in a more than an order of magnitude increase in the permeability coefficient [see Eq. (2)]. In this case, the droplet absorption time (curves 1 and 2 in Fig. 3) decreases insignificantly (by approximately 10%). These data also indicate that the effect of the second layer on the droplet absorption rate is defined primarily by the pressure jump at the absorption front that arises when this front arrives at the interface between the layers.

In the case where the pore size in the second layer $d_2 \gg d_1$ and no absorption by the second layer occurs, 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 leads to 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 and 1' in Fig. 4).

As the pore size in the second layer decreases ($d_2 > d_1$ and $d_2 \approx d_1$), liquid flow to the second porous layer arises, which accelerates the droplet absorption process (curve 2 in Fig. 4). 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. In this case, 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 droplet absorption time and the wet-spot size exhibit maxima (curves 2 and 2' in Fig. 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 layer, the liquid is absorbed primarily by

the second layer. In this case, the droplet absorption time and the size of the wet spot are smaller (curves 3 and 3' in Fig. 4) than those in the case of droplet absorption by a semibounded medium with parameters corresponding to those of the first porous layer ($t_a/t_\infty < 1$ and $S/S_\infty < 1$). Because increasing 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, approaching the values typical of the case of droplet absorption by a semibounded medium ($t_a/t_\infty \rightarrow 1$ and $S/S_\infty \rightarrow 1$).

Conclusions. A two-dimensional model of droplet absorption by one- and two-layer media was considered in which the porous medium is characterized by an effective permeability coefficient dependent on the porosity and the pore size. A comparison of the numerical results and experimental data showed that the model provides an adequate prediction of droplet absorption by porous media.

The numerical experiments showed that the presence of a second layer can 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 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, no absorption by the second layer is observed and the liquid 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 is observed for media with a pore size in the second layer smaller than the pore size in the first layer. In those media, the liquid, having reached the interface between the layers, is absorbed predominantly by the second layer. Unlike in the case of media with the indicated ratio of pore sizes, where an increase in the thickness of the first layer reduces the effect of the 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 have characteristic maxima.

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