Development of the formalism of the discrete element method for the study of the mechanical behavior of liquid-saturated porous materials on a pore scale

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Abstract. In the framework of a single computational approach (particle method), we developed a coupled microscale computer model of liquid-saturated micropores in a solid-phase carrier. Within the model, the solid-phase and liquid-phase components of the contrast system are modeled on the basis of the formalism of homogeneously deformable discrete elements. For computer simulation of flow of complex liquids (including aqueous solutions of drugs) in the micropore volume of the carrier, a new implementation of discrete elements has been developed. This implementation borrows key advantages of the dissipative particle dynamics method. Using the system of adjacent slit-shaped micropores in hydroxyapatite filled with water as an example, we showed an adequacy of the developed discrete element based formalism and its applicability for microscale study of the coupled problems including deformation of solid-phase skeleton and flow of liquids in micropore space.

1. Introduction
The mechanical behavior of multiphase materials is determined by both the rheological properties of the components and the characteristics of their interaction. Special attention is drawn to the so-called contrast materials, some components of which are soft matter. These include, in particular, permeable (porous) liquid-saturated materials with solid skeleton. Their mechanical properties and physicochemical characteristics can be largely determined by the processes of spatial redistribution of highly mobile interstitial liquid in pore space [1]-[5]. This feature determines the ever-growing number of analytical and numerical studies of such kind of contrast systems in order to predict their properties in various thermodynamic conditions, as well as to design the pore structure of new promising materials with desired mechanical, filtration, adsorption and other characteristics. The solution of these problems is based on microscale description and modeling of contrast material as a complex of two interpenetrating and mutually related subsystems: hard matter and mobile soft matter (liquid).

The most fundamental approach to solving these problems is the use of numerical methods of a nanoscale. These methods treat the atomic and molecular structure of both the porous material and the interstitial liquid. The most widely used methods of nanoscale computer simulation are particle methods based on the formalism of the molecular dynamics (MD) method: nonequilibrium molecular dynamics (NEMD) [6][7], coarse-grained molecular dynamics (CGMD) [8], dissipative particle dynamics (DPD) [9], and other methods. Among them, the most promising in modeling the mechanical behavior of contrast systems is the dissipative particle dynamics method [10]. This method is based on the discrete element method (DEM) [11], which is a computer simulation method for the study of the behavior of macroscopic objects in the form of interconnected elements. The method of DEM allows taking into account the interaction of particles through real distances and interaction times. The dissipative particle dynamics method is a generalization of the DEM in the framework of the dissipative particle dynamics method.
dynamics (DPD) [9]-[10].

We specially note the latter method, which can be regarded as in some sense a simplified thermodynamic implementation of the method of coarse-grained molecular dynamics. It is widely used to model “soft matter” systems, such as complex fluids and polyatomic molecules (water with surfactants, proteins, etc.) [11]-[14]. The first key feature of this method is the introduction of the dissipative force the interaction of particles (beads) and the Brownian force. The coefficients of the Brownian and dissipative contributions to the bead-bead interaction force are not independent. Their values are uniquely related to each other to fulfill the condition of maintaining a constant predetermined temperature of an isolated ensemble of beads [10][15]. The second key feature of the method is the formulation of the potential, dissipative, and random constituents of the interaction force using weight coefficients depending on the distance between the centers of the beads. The introduction of these coefficients implies that the internal space of the particle is sufficiently free to allow for the rearrangement of low-atomic molecules or groups of atoms. The capabilities of the dissipative particle dynamics method make it possible to simulate the “soft matter” on a submicron spatial scale and on a time scale of tens of microseconds. DPD is often considered as a method that fills the gap between the nanoscale and microscale approaches to the description of soft matter including liquids.

The description of both solid-phase and liquid components of contrast material on a nanometer scale is carried out within a single (atomistic-molecular) formalism. However, the technical capabilities of such a direct approach make it possible to consider volumes with characteristic sizes less than a micrometer. Therefore, modeling of liquid-saturated porous materials on a microscopic scale (scale of typical pores) is usually carried out with use of numerical methods implying a continuum approximation to the description of the simulated system or its fragment. At the microscopic scale of consideration, the computer models normally treat individual pores of micron or submicron characteristic size, as well as conglomerates of such pores. These models include a pore volume filled with a liquid (for example, an aqueous solution of a drug) and adjacent areas of a solid-phase carrier matrix.

Solid-phase matrix on a microscopic scale is traditionally modeled using methods of continuum mechanics. The most commonly applied are the finite element method [16]-[19] in various implementations, including the extended finite element method [20][21] for modeling the growth and branching of cracks, as well as the finite difference method [22]. However, in recent years, particle-based methods have become increasingly active. The most common representative of this group of methods is the discrete element method (DEM) [23]-[26]. DEM is genetically derived from the molecular dynamics method, however, it is characterized by a number of fundamental differences. In particular, each element contains a sufficient number of atoms/molecules of a substance so that the classical thermodynamic relations can be applied to describe its state. The formulation of the potentials/forces of the element-element interaction is based on the classical (macroscopic) formulations of the defining relationships relating the stresses and strains in the simulated medium. The element is characterized by a certain shape and linear dimensions (in general, they may change both during the deformation process). There are some fundamental consequences of this feature. The first consequence is the use of the concept of contact interaction. The second one is the use of the concept of tangential interaction of elements and taking into account the rotational degrees of freedom of an element by supplementing the equation of translational dynamics with an equation of rotational dynamics. A key advantage of DEM over traditional implementations of numerical methods of continuum mechanics is the ability to directly model complex fracture processes, including multiple fracture, accompanied by relative sliding of fracture surfaces and mass mixing effects. This makes DEM an attractive and effective tool for microscale modeling of changes in the pore structure (pore collapse, dilatancy, etc.). It should be noted that the traditional DEM implementations are based on the use of the simplest approximations of the non-deformable element and the pair-wise interaction potentials. These simplifications strongly limit the scope of the method to brittle materials and granular media. However, the authors of the article recently developed an advanced implementation of the method, which considers discrete element as deformable and uses many-body formulation of
element-element interaction forces [27][28]. This allowed modeling of the behavior of solid-phase materials with complex rheological properties (elastic-plastic, viscoelastic and so on).

Despite the well-known advantages of DEM, its fundamental limitation as a tool for modeling contrast materials on a pore scale is the lack of implementations and models for description of the mechanical behavior of liquids. As a result, the fluid in the micropore space is typically modeled using continuum and meshless numerical methods: finite element realizations of hydrodynamic models and quasiparticle methods including smoothed particle hydrodynamics (SPH) [29][30], particle finite element method (PFEM) [31] and others. This results in the development of “hybrid” computational algorithms. The most pronounced problem in hybrid algorithms is connected with the need to match domains modeled by different numerical methods, to correctly specify their interaction and so on. It is important to note that although the methods of mechanical matching of solid-phase and liquid domains are well developed, the problem of taking into account the peculiarities of the state of the liquid in the near-wall region is still not solved in a general form.

In this regard, it is promising to build a generalized formalism of DEM, which would incorporate the advantages of dissipative particle dynamics for modeling the behavior of a liquid on a microscopic scale. In this case, the description of the mechanical interaction between the particles of the pore liquid, particles of the solid-phase matrix as well as the interaction between the "solid-phase" and "liquid" particles will be carried out within the framework of a single formalism (similar to the nanoscale description). Interface effects at the boundary between the solid and liquid phases can be effectively caught using, for example, adhesive interaction models. This paper is devoted to building such a generalized formalism of DEM based on the previously developed method of homogeneously deformable discrete elements [27][28][32].

2. Generalized formalism of DEM

2.1. DEM model of solid-phase skeleton

The applied formalism of DEM is based on the following principal approximations.

The first one is the most common approximation in DEM. When describing dynamics of a discrete element, its shape is approximated by an equivalent ball (3D problem statement) or disk (2D problem) [24][27][33]. An important consequence is that the equations of motion of a discrete element can be written in simplified Newton-Euler notation:

\[
\begin{align*}
\frac{d^2 \vec{r}_i}{dt^2} &= m_i \frac{d\vec{v}_i}{dt} = \sum_{j=1}^{N_i} \left( F_{ij}^{\text{norm}} + F_{ij}^{\text{tang}} \right), \\
\frac{d\vec{v}_i}{dt} &= \sum_{j=1}^{N_i} \frac{M_{ij}}{J_i} \frac{d\vec{\omega}_i}{dt},
\end{align*}
\]

(1)

where \( \vec{r}_i, \vec{v}_i, \) and \( \vec{\omega}_i, \) are radius vector, velocity vector and pseudovector of the angular velocity of a discrete element \( i, \) \( m_i \) is element mass, \( J_i \) is moment of inertia of an equivalent disk or sphere, \( F_{ij}^{\text{norm}} \) and \( F_{ij}^{\text{tang}} \) are forces of central and tangential interaction of the element \( i \) with the neighbour \( j, \) \( M_{ij} \) is moment of the tangential force \( F_{ij}^{\text{tang}}, \) \( N_i \) is number of neighbours of the element \( i. \) Note that in the framework of this approximation, the central and tangential interaction forces are not formally related to each other (the central force does not cause either tangential movement or rotation, and vice versa).

The second one is the approximation of the uniform (homogeneous) distribution of stresses and strains in the volume of an element. This is similar to the widely used approximation of linear interpolation of displacement distribution in finite elements. In the framework of this approximation, the stress-strain state of a discrete element is characterized by tensors of averaged stresses and strains. The averaged values of the components of the stress tensor in the element are determined through the interaction forces of the element with neighbours [27]:

where \( \tau_{ij}, \sigma_{ij}, \) and \( \theta_{ij}, \) are components of the stress tensor, the displacement vector, and the strain tensor, respectively, \( N_i \) is the number of neighbours of the element \( i. \) Note that in the framework of this approximation, the central and tangential interaction forces are not formally related to each other (the central force does not cause either tangential movement or rotation, and vice versa).
\[
\sigma^i_{\alpha\beta} = \frac{R_i}{V^0_i} \sum_{j=1}^{N_j}\left[\cos\theta_{i,\alpha}\cos\theta_{j,\beta}\sigma_{ij} \pm \cos\theta_{j,\alpha}\sin\theta_{j,\beta}\tau_{ij}\right].
\]

where \( R_i \) is radius of equivalent disc/sphere, \( S^0_j \) is an area of the surface of interaction of unstrained elements \( i \) and \( j \), \( V^0_i \) is a volume of unstrained element, \( \alpha,\beta = x,y,z \) (XYZ is a laboratory coordinate system), \( \cos\theta_{i,\alpha} \) is projection of a unit vector connecting the centers of elements \( i \) and \( j \) onto the axis \( \alpha \) of the laboratory coordinate system, \( \sigma_{ij} \) and \( \tau_{ij} \) are specific values of the forces \( \vec{F}_{ij}^{\text{norm}} \) and \( \vec{F}_{ij}^{\text{tang}} \) respectively. The components of the tensor of averaged strains in the volume of a discrete element are calculated by analogy with the tensor of averaged stresses.

The third approximation, which follows from taking into account the deformability of elements, is the use of a many-body formulation of the relations for the forces of element-element interaction. For locally isotropic materials, the structure of the expression for the element-element central interaction force in the many-body approximation implies taking into account not only pairwise strain (the normalized value of the change in distance between mass centers of elements), but also volume strain or stress. One of the general formulations of the relations for the central and tangential components of the response force of element \( i \) on the mechanical action from the element \( j \) have the following form:

\[
\begin{align*}
\sigma_{ij} &= \sigma_{ij}^{\text{pair}}(\varepsilon_{ij}) + A_i \sigma_{ij}^{\text{mean}}, \\
\tau_{ij} &= \tau_{ij}^{\text{pair}}(\gamma_{ij}),
\end{align*}
\]

where the superscript “pair” denotes the pair component of the corresponding force, \( \sigma_{ij}^{\text{mean}} = (\sigma_{xx}^i + \sigma_{yy}^i + \sigma_{zz}^i)/3 \) is the mean stress in the element \( i \), \( A_i \) is the material parameter, \( \varepsilon_{ij} \) and \( \gamma_{ij} \) are the normal and shear strains of element \( i \) in the pair \( i-j \). A more detailed description is given in [27][28].

Hence the general form of the relations for calculating the forces of interaction of the elements \( i \) and \( j \) (taking into account the need to fulfill the third Newton's law):

\[
\begin{align*}
\left\{\begin{array}{l}
\sigma_{ij} = \sigma_{ij}^{\text{pair}}(\varepsilon_{ij}) + A_i \Delta \sigma_{ij}^{\text{mean}} = \sigma_{ij}^{\text{mean}_{ij}} + A_i \Delta \sigma_{ij}^{\text{mean}}, \\
\Delta h_{ij} = \Delta \varepsilon_{ij} R_i + \Delta \varepsilon_{ij} R_j,
\end{array}\right.
\end{align*}
\]

\[
\begin{align*}
\left\{\begin{array}{l}
\tau_{ij} = \tau_{ij}^{\text{pair}}(\gamma_{ij}) = \tau_{ij}^{\text{pair}}, \\
\Delta l_{ij} = \Delta \gamma_{ij} q_{ij} + \Delta \gamma_{ij} q_{ij},
\end{array}\right.
\end{align*}
\]

Here, the symbol \( \Delta \) denotes the increment of the value of the corresponding parameter per time step of integration of the motion equations, the indices “n” and “n-1” mark the values of the specific response forces of the element \( i \) or \( j \) at the current and previous steps of the numerical scheme, \( \Delta h_{ij} \) is the change of the distance between the centers of the elements over the integration step, \( \Delta l_{ij} \) is the relative shear displacement of the elements of the pair \( i-j \). Equations (4)-(5) are solved for the increments of the central and shear strains of the elements in the pair \( \Delta \varepsilon_{ij} \) and \( \Delta \gamma_{ij} \). The found values are then substituted to determine the current specific values of the forces of element-element interaction \( \sigma_{ij}^{\text{mean}_{ij}} \) and \( \tau_{ij}^{\text{pair}} \). The calculated values of the central and tangential interaction forces are used at the current step of integrating the equations of motion (1). In particular, when modeling an isotropic linear-elastic solid-phase material, the expressions for the increment of specific force in (4)-(5) are formulated as defined by Hooke’s law:
\[
\begin{align*}
\Delta \sigma_{ij} &= 2G_i \Delta \epsilon_{ij}^l + \left( 1 - \frac{2G_i}{3K_i} \right) \Delta \epsilon_{ij}^\text{mean}, \\
\Delta \tau_{ij} &= 2G_i \Delta \gamma_{ij}^l
\end{align*}
\]  

(6)

where \( G_i \) and \( K_i \) are shear modulus and bulk modulus of the material of the element \( i \).

Pairs of elements that model consolidated material are chemically bonded. Local fracture is modeled by breaking a chemical bond in a pair (which corresponds to the occurrence of a local crack). The condition for breaking the bond is the fulfillment of the specified fracture criterion calculated on the interaction surface of the pair [27][28]. Elements in the unbonded pair may be non-interacting or contacting. Mechanical contact of elements generally includes adhesive interaction [34][35] and resistance to relative slippage of the elements in the plane of contact (dry or viscous friction [33]).

2.2. DEM-based model of interstitial liquid

To simulate a liquid in the micropores of a carrier, we developed the particle method, which incorporates some important features of the dissipative particle dynamics method into the formalism of the homogeneously deformable elements. In the framework of this method, the pore liquid is modeled by an ensemble of elements (hereinafter referred to as the liquid particle). Each particle simulates a some volume of liquid. We believe that the size of the liquid particle is large enough so that the main thermodynamic characteristics (pressure, temperature, viscosity, etc.) are applicable to it.

The liquid is assumed to be compressible. Due to the lack of macroscopic elastic resistance of the liquid to shear deformation, only the forces of the central interaction of the particle with the neighbours are taken into account. In this case, by analogy with the dissipative particle dynamics method, the displacement of the particle \( i \) of a liquid is described by the classical Newton’s equation:

\[
m_i \frac{d^2 \mathbf{r}_i}{dt^2} = m_i \frac{d \mathbf{v}_i}{dt} = \sum_{j=1}^{N_i} \mathbf{F}_j^{\text{nom}}.
\]

(7)

As in the DPD, the central force of interaction of fluid particles \( \mathbf{F}_j^{\text{nom}} \) contains three components:

- potential \( \mathbf{F}_j^{\text{pot}} \),
- viscous (dissipative) \( \mathbf{F}_j^{\text{visc}} \), and
- random (Brownian) \( \mathbf{F}_j^{\text{rand}} \).

\[
\mathbf{F}_j^{\text{nom}} = \mathbf{F}_j^{\text{pot}} + \mathbf{F}_j^{\text{visc}} + \mathbf{F}_j^{\text{rand}}.
\]

(8)

Further we will consider liquid as linearly compressible. The potential force of response of the particle \( i \) to the impact by the neighboring particle \( j \) is formulated as a limiting case of the first relation in (6):

\[
\Delta \sigma_{ij} = \Delta \epsilon_{ij}^l = 3K_i \Delta \epsilon_{ij}^\text{mean},
\]

(9)

where \( \sigma_{ij} \) is the specific value of the potential force \( \mathbf{F}_j^{\text{pot}} \), \( \Delta \epsilon_{ij}^l = \left( \epsilon_{ij}^{xx} + \epsilon_{ij}^{yy} + \epsilon_{ij}^{zz} \right)/3 \) is mean strain in the particle. Corresponding diagonal components of averaged strain tensor are calculated as follows:

\[
\epsilon_{i\alpha\alpha}^l = \frac{R_i}{V_i} \sum_{j=1}^{N_i} S_{ij} \epsilon_{ij} \left( \cos \theta_{ij,\alpha} \right)^2.
\]

(10)

Non-diagonal components \( (\alpha \neq \beta) \) are assumed to be zero \( (\epsilon_{i\alpha\beta}^l = 0) \) inasmuch as tangential force of interaction of fluid particles is zero \( (\tau_{ij} = 0) \).

One can see from (10) that averaged strains in the volume of the particle \( i \) are calculated through
particle strains in all pairs i-j. The “limiting” formulation of Eq. (4) is used to calculate these pair strains:

\[
\begin{align*}
K_i \Delta \varepsilon_{ij} &= K_j \Delta \varepsilon_{ji} \\
\Delta h_{ij} &= \Delta \varepsilon_{ij} R_i + \Delta \varepsilon_{ji} R_j.
\end{align*}
\]  

(11)

Eq. (11) is solved for each pair of the elements. Calculated current values of the strains \( \varepsilon_{ij} \) and \( \varepsilon_{ji} \) are then used in (10) to determine the mean strain and mean stress (\( \sigma^i_{\text{mean}} = 3K_i \varepsilon^i_{\text{mean}} \)) in the volume of the particle \( i \). Given the need to comply with the third Newton’s law, the potential force of interaction of particles \( i \) and \( j \) is calculated as the average of the response forces of each of the particles:

\[
\sigma_{ij}^{\text{cur}} = \sigma_{ji}^{\text{cur}} = \sigma_{ij}^{\text{pre}} + \frac{\Delta \sigma_{ij}^i + \Delta \sigma_{ji}^j}{2} = \sigma_{ij}^{\text{pre}} + \frac{\Delta \sigma_{\text{mean}}^i + \Delta \sigma_{\text{mean}}^j}{2}.
\]

(12)

Relations (11) determine the increments of strains in a pair i-j in a linearly elastic approximation (linear compressible liquid). They are unconditionally valid under compression: \( \sigma_{ij}^{\text{cur}} \leq 0 \). Due to the surface tension, a pair of particles simulating a liquid is able to remain adhered even under tensile loading. As the pair stretches, the particles begin to separate at a certain maximum positive (threshold) value of the potential interaction force \( \sigma_{ij}^{\text{cur}} = \sigma_{\text{adh}} \geq 0 \) (at some value of pair strain \( \varepsilon_{ij} = \varepsilon_{\text{crit}} \geq 0 \)). The separation of particles (the transition of the state of a pair from interacting to non-interacting one) occurs gradually as the distance between them further increases. The interaction of particles in the region \( \varepsilon_{ij} > \varepsilon_{\text{crit}} \) is described by adhesion (attracting) potential force, which, unlike the linear-elastic force, does not increase as the distance between the particles increases. An attractive force linearly decreasing with distance (like in DPD method), or a constant adhesion force (Dugdale’s model) can be used as such an adhesion force:

\[
\begin{align*}
\sigma_{ij} &= \sigma_{ji} = \sigma_{\text{adh}} \left( 1 - \frac{R_{ij} - R_{\text{crit}}}{R_{\text{adh}}} \right),
\text{or}
\sigma_{ij} &= \sigma_{ji} = \sigma_{\text{adh}},
\end{align*}
\]

(13)

Here, \( \sigma_{\text{adh}} \) is adhesive strength (or Dugdale’s adhesive stress), \( R_{\text{adh}} \) is the length of action of adhesion potential, \( R_{ij} \) is a distance between mass centers of the elements. The magnitude of the work of adhesion is directly determined by the magnitude of the surface energy of the simulated liquid. Note that similar to compression, at \( \varepsilon_{ij} < \varepsilon_{\text{crit}} \), the interaction between particles is determined by relations (11).

In the case of Newtonian fluid, the dissipative component of the force of interaction of particles \( i \) and \( j \) has the formulation traditional for particle methods:

\[
\vec{F}_{ij}^{\text{visc}} = -\eta_{ij} \vec{V}_{ij}^{\text{norm}},
\]

(14)

where \( \eta_{ij} \) is the viscosity coefficient, \( \vec{V}_{ij}^{\text{norm}} \) is the normal component of the vector of the relative velocity of the particles in the pair \( ij \). Note that viscosity coefficient \( \eta \) is proportional to the dynamic viscosity of fluid and the particle size (it has the dimension [Pa·s·m]).

The random (Brownian) component of the potential force of interaction of particles \( i \) and \( j \) is formulated by analogy with the methods of coarse-grained molecular dynamics and dissipative particle dynamics [10]:
where \( \zeta_{ij} \) is the noise amplitude, \(-1 \leq \zeta_{ij} \leq 1\) is randomly fluctuating variable that should meet the following Gaussian white noise requirements:

\[
\begin{align*}
\left\langle \zeta_{ij}(t) \right\rangle &= 0 \\
\left\langle \zeta_{ij}(t)\zeta_{ij}(t') \right\rangle &= (\delta_{ij} + \delta_{ij} \delta_{tt}) \delta(t - t')
\end{align*}
\]

where \( \delta_{tt} \) is Kronecker delta, \( \delta(t - t') \) is a Dirac delta function of time.

By analogy with dissipative particle dynamics, in order to maintain thermal equilibrium of an isolated liquid system, the amplitude of the random force must be related to the viscosity of the fluid and a specified temperature as follows [10]:

\[
\theta = A \sqrt{\eta T} ,
\]

where \( T \) is a temperature, \( A \) is a coefficient of proportionality. Note that since the viscosity coefficient \( \eta \) in (14) is proportional to the particle size, the contribution of the random component of the interaction force of particles simulating a liquid decreases with an increase in the particle size \( (\theta \propto R^{-1/2}) \) and becomes insignificant when the characteristic scale of the size \( R \) is of the order of \( 10^2 \) nm.

Similar to the above ratios describe the interaction of liquid particles with discrete elements that simulate solid-phase walls of micropore. The specific values of the coefficients and constants for the interaction forces (adhesive strength, the length of the adhesive potential, the viscosity coefficient, the amplitude of the random force, etc.) differ from similar values for the pairs of liquid particles.

3. Modeling of water filtration in slit-shaped micro pore

The results of testing of DEM-DPD hybrid formalism showed that the proposed particle-based model of a liquid allows one to adequately describe redistribution of a liquid in closed volumes of different geometries as well as liquid flow through channels. To demonstrate the adequacy of the model, below we provide the results of computer simulation of liquid transfer in a system of two adjacent slit-shaped micropores. These micropores are considered as part of an interconnecting network of micropores (filtration volume of a carrier). The density, bulk modulus and dynamic viscosity of liquid particles correspond to pure water. The adhesive strength and the characteristic length of action of the adhesive potential were evaluated on the basis of the magnitude of the surface energy (with an accuracy of the order of magnitude). Dugdale's adhesive potential was used. The mechanical characteristics of the material of micropore walls were specified in accordance with the typical values of the mechanical characteristics of hydroxyapatite (HAP), which is a biocompatible material used in different biomedical applications including transplantology. The key mechanical characteristics of solid-phase discrete elements and liquid particles are shown in Table 1.

Figure 1 shows the structure and loading scheme of a model of a pair of adjacent slit-shaped micropores filled with liquid. The analyzed horizontal slit-shaped micropore is connected to a wider (“main”) vertical slit-shaped micropore. The thickness (opening) of the horizontal micropore is \( 1 \) \( \mu \)m. The model takes into account the regions of the HAP solid phase matrix adjacent to the walls. The thickness of the modelled channel wall is \( 0.3 \) \( \mu \)m. Discrete elements simulating micropore walls are shown in black in the figure. The diameter of the vertical channel is \( 3 \) \( \mu \)m. The size of the liquid particles, which model liquid in the pore space, and the solid-phase discrete elements were assumed to be the same and equal to \( 15 \) nm. The discrete elements that form the outer surface of the model walls were rigidly fixed (\( V_x = V_y = 0 \)).
Table 1. Mechanical characteristics of elements/particles modeling solid-phase and liquid-phase components of the system

| Characteristic / Material | Hydroxyapatite (HAP) | Liquid |
|--------------------------|-----------------------|--------|
| Density (kg·m⁻³)         | 3160                  | 1000   |
| Elastic modulus (GPa)    | 100 (Young’s modulus) | 2.2 (bulk modulus) |
| Poisson ratio            | 0.2                   |        |
| Adhesive strength (MPa)  | 0.03 (for pairs “HAP-liquid”) | 0.06 (for pairs “liquid-liquid”) |
| Length of action of adhesive potential (m) | 1·10⁻⁹ | 3·10⁻⁹ |

Figure 1. The structure and the loading scheme of a model of adjacent slit-shaped micropores with a liquid. The discrete elements that form the micropore walls are shown in black. Liquid particles are shown in light gray. Some liquid particles are colored brown to track local flows and the general flow of a liquid.

A liquid is compressed in the vertical channel by the “pistons” moving at a constant speed $V_y$ (Figure 1). This loading imitates an increase in pressure as a result of the action of some external factor. We analyzed the conditions for flow beginning in the horizontal micropore (including the "threshold" value of applied pressure on the piston).

Simulation results show that the flow of liquid in the analyzed horizontal micropore does not occur instantaneously with increase in liquid pressure in the "main" (vertical) micropore. Figure 2 shows an example of typical dependence of the specific value of the compressive resistance force recorded on the upper piston (piston pressure) as a function of time at $V_y=0.5$ m/s. At the initial stage of the piston motion, the resistance force grows nonlinearly up to a some maximum value. When a certain (threshold) piston pressure is reached, the collective motion of particles simulating a liquid begins along the horizontal channel (fluid flow), and the magnitude of the piston pressure decreases by an order of magnitude. A further motion of the pistons in the vertical channel does not cause an increase
in the force of water resistance. This indicates a steady state regime of liquid flow in the micropore. Note that the magnitude of the resistance force in the steady state regime is determined by the viscosity of the liquid. Figure 3 shows flow of liquid in the channel at different stages of loading.

![Figure 2. Typical dependence of piston pressure on time \(V_y=0.5 \text{ m/s}, \sigma_{\text{adh}}=0.03 \text{ MPa}\).](image)

![Figure 3. Liquid flow in the horizontal micropore at various stages of loading: a) the moment of reaching the maximum piston pressure; b) the beginning of the flow of liquid; c) steady state flow. To visualize the flow of the liquid, a certain vertical section of the horizontal micropore was selected at the initial moment of time, and the liquid particles belonging to this section were determined (these particles are considered as section identifiers). In the pictures we displayed only particles that are to the left of the section identifiers. Liquid particles to the right of the section are not displayed (white region inside the channel).](image)

The key factors determining the behavior of a liquid in a micropore are: 1) the magnitude of the rate of pressure gradient increase; 2) the value of the force of adhesion of a liquid to the micropore walls.

Under the considered loading conditions, the first factor is determined by the speed of motion of the pistons in the vertical slit-shaped micropore. We carried out a special study to analyze the influence of the velocity of motion of the pistons \(V_y\) (loading rate) on the dynamics of fluid transfer in a horizontal slit-shaped micropore. Figure 4a shows the dependence of the piston pressure on time for
two different values of \( V_y \). One can see that the loading rate has only time effect on the dynamics of the process of fluid flow in the micropore. The maximum value of the force of liquid resistance to compression (piston pressure) remains nearly constant with a twofold increase in the strain rate, while the “time position” of the pressure peak shifts in proportion to the loading rate. The value of the "residual" pressure (at the stage of steady-state flow) therewith remains at the same level. Note that when a certain (threshold) magnitude of compression rate is exceeded, the magnitude of the maximum pressure begins to increase sharply, which indicates a transition to the hydrodynamic shock regime.

![Figure 4. The dependences of the piston pressure on time for different values of the loading rate: (a) “reference” adhesive strength \( \sigma_{adh}=0.03 \) MPa; (b) two different values of adhesive strength (0.03 MPa and 0.01 MPa).](image)

The second key factor influencing the dynamics of liquid flow in a micropore is the adhesive strength of the bonds between the particles that simulate the liquid and the discrete elements that simulate hydroxyapatite (solid-fluid interface). To study the effect of this factor on the process of liquid flow in a slit-shaped micropore, a series of numerical experiments were carried out, similar to those given above. In these simulations, the adhesive force in the pairs of particles "liquid-hydroxyapatite" and the characteristic length of action of adhesive potential in these pairs were reduced by 3 times in comparison with those given in Table 1. Figure 4b shows the dependences of the piston pressure on time for different loading rates (piston speeds) and adhesive strength.

The simulation results showed that the magnitude of liquid adhesion to hydroxyapatite slightly affects the dynamics of the liquid flow process in the considered range of variation of \( \sigma_{adh} \) and \( r_{adh} \). Namely, a three-fold change in the magnitude of adhesive stress leads to a change in the magnitude of the maximum piston pressure within 30%. The magnitude of the force of resistance to compression at the steady-state stage of the liquid flow weakly depends on the magnitude of the adhesive strength. In other words, at the stage of steady flow, the resistance of the pore liquid is determined mainly by its viscosity. This qualitatively corresponds to the experimentally established regularities of the steady-state flow of liquids in microscale channels.

4. Conclusions
In the paper, we developed a particle-based model of the mechanical behavior of a liquid in the micropore space of a solid-phase material. The developed model combines the key features of the two particle-based methods: the discrete element method and the dissipative particle dynamics method. In the framework of the model, the liquid is modeled as an ensemble of elements (particles). Each of them simulates some volume of fluid that interacts with the surrounding volumes of fluid (particles) and the walls of the solid-phase skeleton. The model takes into account compressibility of liquid, surface energy and the influence of dissipative and random (Brownian) forces. Similar to dissipative particle dynamics, the values of the Brownian force amplitude and dynamic viscosity coefficient are
uniquely related to each other to maintain the specified temperature of the modeled liquid.

The results of the test simulation showed the adequacy of the proposed model of the mechanical behavior of a liquid in the solid-phase skeleton and its applicability for solving various problems concerning fluid-solid interaction at the microscale. One of promising applications of the developed formalism is the modeling of drug transport in the pore space of microporous drug carriers.

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