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Numerical analysis of strong evaporation—condensation through the porous matter

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Abstract

Evaporation-condensation problem in the presence of porous body is investigated. The velocity distribution function of molecules is found from the direct numerical solution of the Boltzmann kinetic equation. New approach for calculation of the distribution function transformation as a result of gas molecules and porous body particles interactions is presented. The data of known theoretical and experimental works are compared with authors' results. Various approaches for the porous body description are discussed. © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: The Boltzmann kinetic equation; Numerical solution; Transformation of distribution function; Models of porous matter; Evaporation-condensation

1. Introduction

In the present work we investigate heat–mass transfer problem in porous layer.

The kinetic theory methods were used for the porous medium modelling in different works [1–4]. Most approaches for this problem investigation are based on the dust–gas models. In these models, the highly porous body is simulated by a homogeneous system of randomly distributed immovable spherical particles [1–3]. In [1] the conclusion about possibility of simulation in principle of porous matter by such manner was made. The coefficient of permeability was obtained on the basis of spherical particles model and using the Chapman–Enskog method for the kinetic problem solutions. Furthermore, in this paper authors have shown that the kinetic approach devoted by them gives the opportunity to estimate interface surface of porous body (specific surface).

In [2] gas flow in the porous body was investigated by using dust–gas model also. In analysis and corresponding calculations collision integral was replaced by external force. This force was regarded as the result of collective interaction of gas molecules with particles.

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In [3] the flow of a two-component gas mixture in the highly porous mater was investigated. The condensation of one component and absorption of another component have been taken into account. In this paper, the direct simulation Monte Carlo (DSMC) method was used for calculation.

In [4] the rarefied gas flow through the porous layer simulated by a set of parallel channels was studied. The considerable interaction between gas flows entering into porous body channels was discovered by computation. The DSMC method was used also.

Authors of the present work suggest an approach for calculation of gas velocity distribution function transformation as a result of gas molecules and immovable particle interactions and determination on the base of substantial characteristics of porous body including permeability coefficient. Here different models of porous body simulation are discussed. We believe that this approach can describe the large spectrum of processes and phenomena.

2. Problem, main equations and approach

The problem of evaporation—condensation in volume filled by porous substance is considered.

Nomen	clature	и х, у, z	mean (macroscopic) velocity of gas Cartesian co-ordinates
d D f J m	diameter of gas (vapour) molecule particle diameter velocity distribution function of molecules collision integral mass flux density mass of molecule	Greek sy ϵ χ ξ_x, ξ_y, ξ_z Subscript	porosity permeability coefficient of porous medium components of gas molecule velocity ξ
M	total quantity of velocity grid points	*	effective value
N	quantity of immovable particles per unit volume of porous body	1	type of molecule for which distribution function is determined
$\hat{N}_{ m d}$	quantity of immovable particles in 1 g of catalyst	11, 12 c	type of interaction cylinder
n	numerical density of gas	d1	individual solid particle
P_0	gas pressure on the porous body entrance	d	solid
R	individual gas constant	g	for pores in 1 g of catalyst
$S_{ m g}$	summed surface of pores in 1 g of catalyst	k, n	number of velocity grid points
S_{d1}	surface of one solid particle	x, y, z	Cartesian co-ordinates
T t T_0 $V_{\rm d1}$ $V_{\rm d}$	temperature of gas time temperature of interphase on which evaporation is realized volume of one solid particle volume occupied by particles in 1 g of catalyst	Superscri * ' k n r	after collisions with solid particle after interactions number of equations in system not interacted has been transformed

The statement of this problem is shown in Fig. 1. The evaporation of nitrogen takes place at surface x = 0. The domain x = 0–5 is occupied by porous matter. The surface x = 5 is absolutely cryogenic: the gas is absorbed completely. Further all densities and temperatures are given in relation to basis parameters: density $n = 4.83 \times 10^{24} \text{ l/m}^3$ and temperature T = 300 K, these values correspond to pressure p = 20 kPa. The co-ordinate x is given in mean free paths of vapour molecules at these parameters.

The study of the above-described porous substance is made by the Boltzmann kinetic equation (BKE) for onedimensional non-steady statement

$$\frac{\partial f}{\partial t} + \xi_x \frac{\partial f}{\partial x} = J_{11} + J_{12},\tag{1}$$

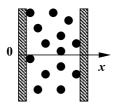


Fig. 1. Schematic illustration of the problem.

where $f = f(x, t, \xi)$ is the velocity distribution function, ξ (ξ_x, ξ_y, ξ_z) is molecular velocity, t is time, x is Cartesian co-ordinate, J_{11} and J_{12} are the collision integrals describing interactions between gas (vapour) molecules only and between gas (vapour) molecules and solid particles correspondingly.

In writing the expression for the collision integrals we have used the notations introduced by Kogan [5,6]

$$\begin{split} J &= \int_{\varOmega} \int \int \int \int \int \left(f' f_1' - f f_1 \right) |\xi - \xi_1| b \, \mathrm{d}\Omega, \\ \mathrm{d}\Omega &= \mathrm{d}b \, \mathrm{d}\varepsilon \mathrm{d}\xi_1. \end{split}$$

To solve BKE it is necessary to define initial and boundary conditions. It is assumed that gas molecules emitted from the surface x=0 have Maxwell distribution function with n=1, T=1 and zero flow velocity (diffuse scheme). All molecules arriving at the surface x=5 are condensed completely and because this surface is absolutely cryogenic no molecules are emitted from this interphase therefore $f\to 0$ for $\xi_x < 0$. In time t=0 $f\to 0$ for whole calculation region in configuration and velocity space.

The velocity distribution function is found as result of solving BKE. Macroparameters: density, temperature, pressure, mass flux, energy flux and others are moments of the distribution function and determined by the integration over three-dimensional space of molecules velocities. In particular

$$\begin{aligned}
 j_x &= \int \xi_x f \, \mathrm{d}\xi, \\
 N &= \int f \, \mathrm{d}\xi.
 \end{aligned}
 \tag{2}$$

To compare obtained results with data found by other authors it is necessary to calculate the permeability coefficient χ , which is determined by formulae (see, for example, [2])

$$\mathbf{j} = \chi \operatorname{grad} n, \tag{3}$$

where \mathbf{j} is the mass flux density.

The direct numerical solution of the Boltzmann kinetic equation (DNSBE) method [7] has been chosen to solve Eq. (1) and has been applied to the study of gassolid particles mixture.

At solving the BKE a rigorous numerical technique that includes conservative finite-difference approximation of differential part of equation and special Korobov's formulae for computing a multidimensional collision integral gas—gas (J_{11}) with high accuracy is used. The original Eq. (1) is reduced to a system of a large number (of the order of some hundreds or thousands) of finite-difference equations with a quadratic non-linearity in the right-hand side

$$\frac{\Delta f^k}{\Delta t} + \xi_x^k \frac{\Delta f^k}{\Delta x} = J_{11}^k + J_{12}^k.$$

This system is resolved by an iterative procedure.

The main steps of initial algorithm [7] are the following:

1. Motion without collisions

$$\frac{\Delta f^k}{\Delta t} + \xi_x^k \frac{\Delta f^k}{\Delta r} = 0.$$

2. Uniform relaxation

$$\frac{\Delta f^k}{\Delta t} = J_{11}^k + J_{12}^k.$$

- 3. Conservative correction.
- 4. Calculation of macroparameters.

The main part of the present work is devoted to the numerical technique of collision integral J_{12} calculation-described interaction between gas molecules and solid particles.

3. Models of porous matter

Some different methods are suggested in this study for simulation of real porous substance. In the first of them spherical particles model is used (Fig. 2(a)). At this it is supposed that spherical particles are distributed uniformly in physical space. In the second method the porous body is described by the set of cylinders with diameter D and length L_c (Fig. 2(b)). Cylinder axes are directed along x co-ordinate. Cylinders circular crosssections are distributed uniformly in the y-z plane. In the third method combined model is used (Fig. 2(c)). In this model the porous matter is replaced by the system of uniformly distributed particles also. But different from the first model these particles are the combination of spherical and cylindrical surface forms. Each particle consists of cylinder with diameter D and length L_c and two half sphere with diameter D. As in the second method axes of combined particles are oriented along x co-ordinate and circular cross-sections of these particles are distributed uniformly in y-z plane. The last model is the universal among presented methods at the simulation of porous body. At $L_c \rightarrow \infty$ porous matter is simulated by the set of cylinders and at $L_c \rightarrow 0$ porous matter is simulated by spheres.

The porosity ϵ of studied body is determined as the ratio of volume of all pores to whole volume occupied by porous body. This definition of porosity and simulation method connected with solid particles geometry gives the possibility to deduce the relationship among particles concentration and their sizes.

$$\epsilon = 1 - NV_{\rm d1},\tag{4}$$

where $V_{\rm d1}$ is volume of one solid particle.

One can see in (4) that it is possible to obtain many models of porous matter with same ϵ but with different solid particle forms. Below the calculations results are given for different sizes and solid particle concentrations.

Taking into account experimental data presented in paper [8] namely volume of pores and their summed surface per 1 g of catalyst the real porous body can be replaced by equivalent model. Volume occupied by particles in 1 g of catalyst is equal to $V_d = V_{d1} \hat{N}_d$, where

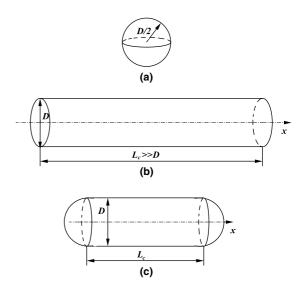


Fig. 2. Models of porous body particles.

 \hat{N}_d is general quantity of particles in 1 g of catalyst. Correspondingly summed pores surface is determined by the correlation $S_g = S_{d1} \hat{N}_d$, where S_{d1} is surface of one solid particle, S_g is summed surface of pores in 1 g of catalyst.

Formulae for $V_{\rm d1}$ and $S_{\rm d1}$ are determined by geometry of solid particles set which simulates the real porous body. For combined sphere–cylinder particle (Fig. 2(c)) $V_{\rm d1} = \pi D^2/2(L_{\rm c}/2 + D/3)$ and $S_{\rm d1} = \pi D(D + L_{\rm c})$. On the base of definition (4) taking into account that volume of 1 g catalyst V is equal to sum of porous volume $V_{\rm g}$ and solid particles volume per 1 g catalyst namely: $V = V_{\rm g} + V_{\rm d}$, sizes of solid particles and their concentration depending on $V_{\rm d}$ and $S_{\rm g}$ can be obtained. Values of $V_{\rm d}$ and $S_{\rm g}$ for real concrete catalysts presented in paper [8] are used for calculation in this paper below.

4. Solution method and input data

As it was noted above the direct numerical solution of the BKE method [7] is used in the present paper in the version developed for gas–solid particles mixture [9]. To realize this way collision integral J_{12} is replaced by certain numerical procedure describing reflection of the gas molecules from solid particles. Therefore, the distribution function transformation caused by interactions between gas molecules and solid particles are considered below.

It is assumed inside this discrete model that gas molecules accept only those values of the velocities ξ_k , which are determined by velocity grid.

During timestep Δt only part n^{r} of the all gas molecules n has been reflected by solid particles. In such a manner velocity distribution function f_k is composed by two parts: the first invariable portion f_k^{n} corresponding to gas molecules which has not been interacted with porous matter; the second part f_k^{r} of the distribution function which has been transformed as a result of the reflection. Index k refers to k-point in velocity space

$$f_k = f_k^{\rm n} + f_k^{\rm r}.$$

Reflection of the gas molecules from solid particles is described by diffuse scheme. The molecules which had identical velocities ξ_k before collision will have different velocities after it. Fig. 3(a) illustrates the process of transformation of the distribution function. The values of distribution function for reflected gas molecules are determined by non-penetration condition at complete accommodation. This procedure should be done for each point ξ_k of velocity grid.

The velocity distribution function for gas molecules after collisions with porous substance $f_k^{r^*}$ is found as a result of summing of all reflected distribution function $f_n^{r^*}(\xi_k)$ obtained for each grid velocity point ξ_n :

$$f_k^{{f r}^*} = \sum_{n=1}^M f_n^{{f r}^*}({f \xi}_k), \quad (^*),$$

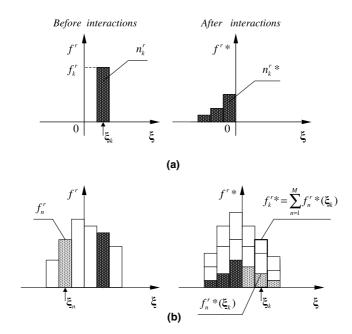


Fig. 3. The velocity distribution function transformation at interaction of gas molecules with solid particles.

where M is the total quantity of velocity grid points. Expression (*) shows that each group of molecules having before collision the velocity ξ_n makes its own contribution in the determination of distribution function f_k^{**} after reflection (Fig. 3(b)).

The quantity of the gas molecules in volume unit interacting with spherical solid particles during time Δt is given by the formula

$$n_k^{\rm r} = \frac{1}{4} N n_k \pi D_*^2 |\mathbf{\xi}_k - \mathbf{v}| \Delta t.$$

In analogy for immovable cylindrical solid particles

$$n_k^{\rm r} = Nn_k D_* L_{\rm c} \sqrt{\xi_{ky}^2 + \xi_{kz}^2} \Delta t.$$

Here $D_* = D + d$, D is the diameter of the solid spherical or cylindrical particle, d is diameter of the gas molecule, $|\xi_k - \mathbf{v}|$ is relative velocity of moving for solid particle and gas molecule, ξ_{ky} , ξ_{kz} , y- and z- components of the ξ_k -vector, N is numerical density of the solid particles, $n_k = f_k \Delta \xi^3$ is numerical density of gas molecules having velocity ξ_k and $\Delta \xi^3$ is the volume of cell in velocity space.

Correspondingly for part of the distribution function f_k^{T} the following expressions are valid:

$$f_k^{\rm r} = \frac{1}{4} f_k N \pi D_*^2 |\xi_k - \mathbf{v}| \Delta t \quad \text{spherical particles},$$

$$f_k^{\rm r} = f_k N D_* L_{\rm c} \sqrt{\xi_{ky}^2 + \xi_{kz}^2} \Delta t \quad \text{for cylindrical particles}.$$
(5)

Calculation $f_k^{r^*}$ in accordance with the above-described algorithm gives the possibility to determine velocity distribution function of gas molecules after interactions with solid particles:

$$f_k^* = f_k^{\mathrm{n}} + f_k^{\mathrm{r}^*}.$$

To obtain the distribution function in whole velocity space it is necessary to repeat this procedure for all velocity grid points.

Values D and N calculated on the base of spherical particles model are presented in Table 1. The spherical particle sizes are given in the first column and corresponding numerical density in the second column of this table.

Table 1 Parameters of spherical particles model

D (m)	$N ({ m m}^{-3})$
5.0×10^{-10}	7.18×10^{25}
5.0×10^{-10}	1.16×10^{27}
14.8×10^{-10}	7.18×10^{25}
20.0×10^{-10}	7.18×10^{25}
29.6×10^{-10}	7.18×10^{25}
90.6×10^{-10}	0.39×10^{24}
90.6×10^{-10}	0.77×10^{24}

Table 2
Parameters of system of cylinders and combined particles model

D (m)	$L_{\rm c}$ (m)	$N~(\mathrm{m}^{-3})$
93.4×10^{-10}	17.0×10^{-7}	4.38×10^{15}
93.4×10^{-10}	17.0×10^{-7}	2.22×10^{21}
93.8×10^{-10}	1.7×10^{-7}	2.43×10^{22}

Same data for cylindrical particles are presented in the second and the third rows of Table 2. Corresponding values for combined particles are given in the fourth row of this table.

5. Results and discussion

Solution results of the problem with evaporation condensation for different porous matter models are presented in Tables 3, 4 and in Fig. 4. It was noted above and was shown in Tables 1 and 2 that porous body with same parameter ϵ can be simulated by some (not one) ways. For example, the porosity $\epsilon \cong 0.7$ in this paper is simulated by the following sets: $D = 20 \times 10^{-10}$ m at $N = 7.18 \times 10^{25} \text{ m}^{-3}$ and $D = 90.6 \times 10^{-10} \text{ m}$ at N = $0.77 \times 10^{24} \text{ m}^{-3}$ for spherical particles and $D = 93.8 \times 10^{12} \text{ m}^{-3}$ 10^{-10} m, $L_{\rm c} = 1.7 \times 10^{-7}$ m, $N = 2.43 \times 10^{22}$ m⁻³ for combined model of particles. Vapour density distribution along co-ordinate x is shown in Fig. 4 for spherical $D = 0.5 \times 10^{-9}$ model at $N = 7.18 \times 10^{25} \text{ m}^{-3}$. Here the result obtained without taking into account collisions between molecules is presented by solid line: $J_{11} = 0$. In this figure results in presence of molecule collisions are shown by dots: $J_{11} \neq 0$.

It is known that coefficient of permeability is the substantial characteristic of porous body. Also analysis of papers [1,2,8] shows that coefficient χ for concrete body is a constant value with accuracy of some quota of percentage. Therefore there is no necessity to solve Eq. (1) in domain with larger value along co-ordinate x size (which could be comparable with real macroscopic sizes of porous body). Thus coefficient of permeability can be

Table 3
Porosity and coefficient of permeability for spherical particles model

D (m)	ϵ	$N ({ m m}^{-3})$	$\chi (m^2/s)$
5.0×10^{-10}	0.99	7.18×10^{25}	1.12×10^{-6}
5.0×10^{-10}	0.92	1.16×10^{27}	6.24×10^{-8}
14.8×10^{-10}	0.88	7.18×10^{25}	4.16×10^{-8}
20.0×10^{-10}	0.70	7.18×10^{25}	2.68×10^{-8}
29.6×10^{-10}	0.03	7.18×10^{25}	1.52×10^{-8}
90.6×10^{-10}	0.85	0.39×10^{24}	5.92×10^{-7}
90.6×10^{-10}	0.70	0.77×10^{24}	2.50×10^{-7}

Table 4		
Porosity and coefficient of permeability	y for system of cylinders and	combined particles model

D (m)	$L_{\rm c}$ (m)	ϵ	$N ({ m m}^{-3})$	$\chi \ (m^2/s)$
93.4×10^{-10}	17×10^{-7}	$\rightarrow 1$	4.38×10^{15}	6.69×10^{-4}
93.4×10^{-10}	17×10^{-7}	0.74	2.22×10^{21}	5.83×10^{-4}
93.8×10^{-10}	1.7×10^{-7}	0.70	2.43×10^{22}	1.42×10^{-6}

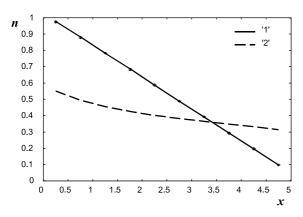


Fig. 4. The dependencies of gas density n on x inside domain of calculation: (1) evaporation through the porous body; (2) evaporation from an open surface.

determined on the base of solution results obtained in the region with size along $x L \gg D$, where D is solid particle diameter.

Coefficient of permeability calculation results for different porous bodies simulated by spherical particles are presented in Table 3. Corresponding results for system of cylinders are given in Table 4. In the last row of Table 4 calculation results for combined particles model are shown.

Analysis of Tables 3 and 4 data results in conclusion about the influence of solid particle geometry on the value of coefficient of permeability. Such kind of behaviour can be explained by different system of cylindrical or spherical surfaces simulating pores interface surfaces influence. As it was noted above the character and peculiarities of gas flow through the porous matter are determined mainly by interactions of gas molecules with solid particles of porous body. Quantity of molecules which collide with pore boundaries determined in accordance with correlation (5) can be different and these differences can be large enough. This peculiarity is due to position that for same porous body parameter sizes and concentration of solid particles depend on their geometry.

It should be emphasized that in the present paper the authors do not try to make the direct simulation of porous body but propose the method of porous body description adapted to the direct numerical solving of the Boltzmann equation. Prescribing solid particles ge-

ometry (spheres, cylinders and so on) we determine part of molecules from general quantity of these molecules which interacts with solid particles. Basically model operates not with real forms of particles but with the system of different oriented surfaces which can reflect molecules. Comparison of spherical and cylindrical surfaces leads to conclusion that in sphere case solid particles more often meet molecules during their motion. Correspondingly in this case directions of molecules motions are varied more often also. Therefore coefficient of permeability for spherical solid particles is smaller than for cylindrical particles at same body porosity.

The quantitative influence to geometry of the particles by which porous body is modelled on value of permeability γ is seen from results presented in Tables 3 and 4. Permeability χ is defined by value of mass flux density through porous body and mentioned density gradient which were obtain by present calculation. These results show that, for example, at approximately same diameters of the sphere (D = 9.06 nm) and cylinder (D = 9.34 nm) gas percolates considerably better (the coefficient of permeability $\chi \approx 10^{-4} \text{ m}^2/\text{s}$) through the porous body model from cylinders with length $L_{\rm c} = 1700 \text{ nm}$ than through the model from spheres $(\chi \approx 10^{-7} \text{ m}^2/\text{s})$ at same porosity ϵ , pore specific volume $V_{\rm g} = 4.68 \times 10^{-7} \, \, {\rm m}^3/{\rm g}$ and specific $S_g = 86 \text{ m}^2/\text{g}$. Thereby the cylindrical elements provide greater value of γ and this value should be larger at the rise of the cylinders length. Cases when the particles of the porous body are formed by the system of spherical and cylindrical surfaces are investigated also. The result of calculation shows that the coefficient of permeability (when the $\epsilon, V_{\rm g}, S_{\rm g}$ are prescribed) depends substantially on the cylinders length.

The coefficient of permeability can been obtained also from expression presented by [1] for different schemes of molecular reflection from pores surfaces. It should be noted that results [1] were got as solutions of the Boltzmann equation for gas–dust mixture in linearized problem. At this the model of hard spheres was used for gas–gas and gas–dust interactions description. For example, for D=2 nm, $N=7.18\times10^{25}$ m⁻³ ($\epsilon=0.7$ and nitrogen gas) the values of permeability coefficient equal $\chi=4.81\times10^{-6}$ m²/s for mirror reflection scheme and $\chi=3.278\times10^{-6}$ m²/s for diffuse reflection scheme were obtained. The calculations for the equal parameters based on the hard spherical particles

model and approach which has been presented in Section 4 of this paper give $\chi = 2.68 \times 10^{-8} \text{ m}^2/\text{s}$.

In paper [2] the porous body is simulated by system of randomly distributed immovable spherical particles. Approach [2] for diffuse reflection scheme and nitrogen gas at $\epsilon = 0.7$ gives $\chi = 0.893 \times 10^{-6}$ m²/s.

The analogous calculation can be made for the combined model which is described in Section 3. If it is assumed that at $\epsilon=0.7$ and $N=2.43\times 10^{22}$ m⁻³ the length of cylinders $L_{\rm c}=1.7\times 10^{-7}$ m and diameter $D=93.8\times 10^{-10}$ m, respectively, the procedure of the Section 4 gives value $\chi=1.42\times 10^{-6}$ m²/s.

It is well clear that value of χ obtained on the basis of various approaches can differ essentially. However at the present stage there is no opportunity to make conclusion about correctness of description based on some model.

For the decision of problem about adequacy of model to reality it is necessary to carry out experiment with well-controllable geometrical characteristics of porous body and not at a level ϵ , $V_{\rm g}$, $S_{\rm g}$ but with definition of the sizes of an individual solid particle. The data of known experimental work [8] do not give good enough response on this problem.

The results presented in the paper were obtained statistically and accurately in the frames of diffuse scheme of molecular reflection from porous body surfaces. This note is necessary because often the description of the porous body is realized on the basis of correlations and formulae for flows through a single element: single capillary (channel) for the model of cylindrical particles, single cell with hard spherical particle in centre for the model of spherical particles and others. After obtaining information about single cell, parameters and characteristics of whole porous body are calculated just as sum of corresponding values for separate elements. However this operation can give large errors. Paper [4] shows that the result of calculation for one channel and system channels can differ more then on 50%.

6. Conclusions

 Procedure describing the transformation of velocity distribution function for gas (vapour) molecules at their interactions with immovable particles of porous body in accordance with diffuse reflection scheme is suggested. Using this procedure in the direct numerical solving of the Boltzmann equation gives the possibility to calculate gas flows through model po-

- rous bodies and to determine macroscopic characteristics concerning these flows and bodies including the coefficient of permeability.
- The efficiency of gas flow through the porous body at the gas percolation depends strongly on the model of porous body simulation as it was supposed.
- At the equal porosity, specific interface surface: gas—solid and specific volume of solid the model of cylindrical elements gives approximately 100-fold larger value of permeability coefficient than the model of spherical particles.

Acknowledgements

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