

FILTRATION OF DYNAMICALLY ADSORBED GAS IN A BIDISPERSE POROUS LAYER**¹Yelisieiev V., ¹Lutsenko V., ²Berkout V.**¹*M.S. Poliakov Institute of Geotechnical Mechanics of the National Academy of Sciences of Ukraine*²*MassTech Inc.*

Abstract. Among the problems related to filtration and mass transfer in porous bodies, the problems of the adsorption-desorption are currently of the greatest interest. This is due to a wide range of problems: the safety of mining and coal developments, the extraction of combustible gases, the storage of greenhouse gases in formations. At the same time, in theoretical terms, issues related to the multi-scale nature of fractured-porous media, in particular, mining and coal seams, are gaining wide popularity.

This paper considers the problem of filtration flow during gas injection into a porous formation, taking into account gas adsorption both on the surface of channels in macropores and during volumetric sedimentation in micropores. In the paper, it is assumed that the entire pore space of a coal seam, according to the results of recent studies, can be divided into two parts of mutually connected areas that differ from each other in their characteristic sizes of pore channels and, accordingly, with a large difference in permeability. The main attention is paid to the filtration features of the process of adsorbed gas flow in the reservoir. The developed model is based on the theory of inertialess gas motion, i.e. using the Darcy equation written in each zone separately. It is assumed according to the literature that the adsorption process obeys the Langmuir-type equation. The problem considers two stages of the process: gas injection into the reservoir and the process of filtration pressure establishment during reservoir blockage. Pressure changes in two different channel systems are shown depending on the intensity of gas flow from one pore branch of the system to another. With intensive overflows, pressure differences between the areas are insignificant. With weak overflows, the differences are large, which should lead to large internal stresses. The calculations performed qualitatively show the dynamics of the adsorption process during gas movement in the reservoir. It follows from the calculations that it largely depends on the structure of the pore space. With intensive overflows, the adsorption process practically follows the change in pressure in the entire pore space. For weak flows, the resulting large pressure differences between the two regions affect the adsorption process.

Keywords: porous medium, gas, diffusion, mass transfer, filtration, adsorption, microporous channels.

1. Introduction

Almost all rocks and coal seams are fractured-porous media with at least two conventional characteristic sizes of porous channels [1, 2]. In this regard, at present, when considering filtration problems with mass exchange processes in porous and fractured-porous media, the representation of the pore space of the medium in the form of bidisperse structures has become widely popular. The relevance of such problems is associated with the problems of oil production, natural gas production, carbon dioxide storage, and the retention of greenhouse gases in soils [3–6]. The number of publications considering theoretical formulations of problems and experimental results is growing. One of the interesting areas here is the consideration of mass transfer taking into account adsorption [5, 7] or desorption. It is believed that gas is adsorbed both on the internal surfaces of porous media in relatively wide channels or cracks, and in micropores, which, according to the theory of volumetric filling [8], are completely filled with it.

In this article, which is a continuation and clarification of the work [9], we will assume that the pore structure consists of two types of channels that differ significantly from each other in their characteristic diameters, as well as permeabilities. For example, according to measurements [1], these can be radii $R_{p1} = 20 \cdot 10^{-6}$ m and $R_{p2} = 10^{-7}$ m, and accordingly with [10], the gas permeability



coefficients can be 10^{-13} m^2 and 10^{-15} m^2 . Microporous structures that are volumetrically filled with adsorbed gas are adjacent to these channels. As a result, we will assume that gas is filtered through these channels, which is adsorbed both on the surface of the channels and in the volumes of the micropores.

Unlike works [5, 7], which consider the diffusion movement of gas in micropores, we will assume that adsorption, according to [8], is subject to both the theory of surface deposition and the theory of volume filling, i.e. we will use the known dependencies of the adsorption theory. This formulation is associated with the dynamic stage of adsorption [11], when as a result of mass exchange processes in the pore channels a quasi-equilibrium state is established. The time for establishing such equilibrium is comparatively short. The subsequent process is associated with the diffusion movement of the adsorbate in very narrow pores, but the time of this stage of the process can significantly exceed the time of the dynamic stage.

Thus, the structure of the region associated with the dynamic stage of adsorption that we have adopted consists of three zones. The first two zones belong to a two-pore conjugated system with different, very different filtration characteristics, in which the gas is filtered, and the third is a micropore zone adjacent to both the first and the second. In the micropore zone, volume adsorption occurs with a known pattern. Such a division allows us to differentiate the pore space in more detail and, accordingly, better determine its characteristics and the influence of these areas on the entire process as a whole.

2. Theoretical part

We will assume that the pore space of the formation through which the gas is filtered consists of channels of two types with diameters that differ greatly from each other. At the same time, due to the low velocities, Darcy's law is realized in both zones, i.e.

$$u_J = -\frac{K_J}{\mu} \frac{\partial p_J}{\partial x}, J=1, 2, \quad (1)$$

where u is the filtration velocity, m/s; p is the pressure, Pa; K is the gas permeability, m^2 ; μ is the dynamic viscosity coefficient, Pa·s; index J refers to a particular zone. The equations of mass conservation in these two zones will have the form

$$\frac{\partial(\varepsilon_{PJ}\rho_J)}{\partial t} + \frac{\partial(\varepsilon_{PJ}\rho_J u_J)}{\partial x} = g_{AJ} + g_{PJ}, \quad (2)$$

where t is time, s; x is the coordinate, m; ε_{PJ} is the porosity associated with the channels; ρ is the gas density, kg/m^3 ; g_A is the flow rate associated with gas adsorption, $\text{kg}/(\text{s}\cdot\text{m}^3)$; g_P is the flow rate associated with gas flows from one porous structure to another, $\text{kg}/(\text{s}\cdot\text{m}^3)$.

Let us further assume that the formation is at a constant temperature T (for example, 293 K), and the injected pressure is relatively small, for example, $p = 50$ Pa, then it follows from the gas equation of state that

$$\rho_J = \frac{p_J}{RT}. \quad (3)$$

The flow rate for internal flows (in this case from the first to the second) is defined as

$$g_{P1} = -g_{P2} = S \frac{1}{2} (\rho_1 + \rho_2) (p_2 - p_1), \quad (4)$$

where S are the coefficients characterizing the flow rate per unit length, s m/kg. We will represent the adsorption component g_A for each of the regions as the sum of two values $g_{AJ} = g_{APJ} + g_{AVJ}$, g_{APJ} is associated with adsorption on the surface of the channels, and g_{AVJ} is associated with the volume adsorption in the micropores.

We will also assume that in addition to the porosity ε_{PJ} , there is porosity associated with the microporous structure, i.e. ε_{VJ} , which characterizes the volume of the micropores.

Thus, the values ε_{PJ} participate in both the filtration flow and the adsorption process, and ε_{VJ} - only in the adsorption process. According to [8], we will represent the surface adsorption in the form of the Langmuir dependence

$$a_{API} = A_{PJ} \frac{\gamma_{PJ}(p_J/p_A)}{[1 + \gamma_{PJ}(p_J/p_A)]}, \quad (5)$$

and volumetric - with the same expression

$$a_{AVI} = A_{VJ} \frac{\gamma_{VJ}(p_J/p_A)}{[1 + \gamma_{VJ}(p_J/p_A)]}, \quad (6)$$

where γ_{PJ} , γ_{VJ} are the values characterizing the adsorption rate, and A_{PJ} and A_{VJ} are the values indicating its maximum.

In [8], it is noted that, despite the more complex process of adsorption filling of micropores (the Dubinin-Radushkevich theory), the formal mathematical law (6) can be considered as a rough approximation, since the adsorption isotherm in micropores is close to the first type (Langmuir) according to the Brunauer classification. An important difference between the values of A_{PJ} and A_{VJ} is that A_{PJ} depends on the size of the surface onto which the gas is deposited, and A_{VJ} depends on the volume of micropores, so they can be roughly represented as: $A_{PJ} = \rho_{AD} \cdot \alpha \cdot \varepsilon_{PJ} (R_*/R_{PJ})$, $A_{VJ} = \rho_{AD} \cdot \beta \cdot \varepsilon_{VJ}$, where R_* is a certain characteristic radius, ρ_{AD} is the conditional density of

the adsorbed gas. Considering that $g_{APJ} = -\partial a_{APJ} / \partial t$, $g_{AVJ} = -\partial a_{AVJ} / \partial t$, combining the expressions written out, we can write the main equations as:

$$\left\{ 1 + \frac{RT}{p_A \varepsilon_{P1}} \left[A_{P1} \frac{\gamma_{P1}}{(1 + \gamma_{P1} p_A \sigma_1)^2} + A_{V1} \frac{\gamma_{V1}}{(1 + \gamma_{V1} p_A \sigma_1)^2} \right] \right\} \frac{\partial \sigma_1}{\partial \tau} + \frac{\partial^2 (\sigma_1^2)}{\partial \zeta^2} = \frac{p_A (\sigma_1 + \sigma_2)}{2 \varepsilon_{P1}} T_M S (\sigma_2 - \sigma_1), \quad (7)$$

$$\left\{ 1 + \frac{RT}{p_A \varepsilon_{P2}} \left[A_{P2} \frac{\gamma_{P2}}{(1 + \gamma_{P2} p_A \sigma_2)^2} + A_{V2} \frac{\gamma_{V2}}{(1 + \gamma_{V2} p_A \sigma_2)^2} \right] \right\} \frac{\partial \sigma_2}{\partial \tau} + \frac{K_2}{K_1} \frac{\partial^2 (\sigma_2^2)}{\partial \zeta^2} = -\frac{p_A (\sigma_1 + \sigma_2)}{2 \varepsilon_{P2}} T_M S (\sigma_2 - \sigma_1), \quad (8)$$

where $\sigma_J = p_J / p_A$; $\tau = t / T_M$, $\zeta = x / h$, $T_M = 2 \frac{h^2}{K_1 p_A} \mu$ – time scale, h – layer width, m.

The boundary conditions for equations (7) and (8) are the relations: when pumping a layer ($0 \leq \tau \leq \tau_*$)

$$\begin{aligned} \text{at } \zeta = 0 \quad \sigma_1 = \sigma_2 = 50, \\ \text{at } \zeta = 1 \quad \sigma_1 = \sigma_2 = 1; \end{aligned} \quad (9)$$

when the layer is blockage ($\tau \geq \tau_*$)

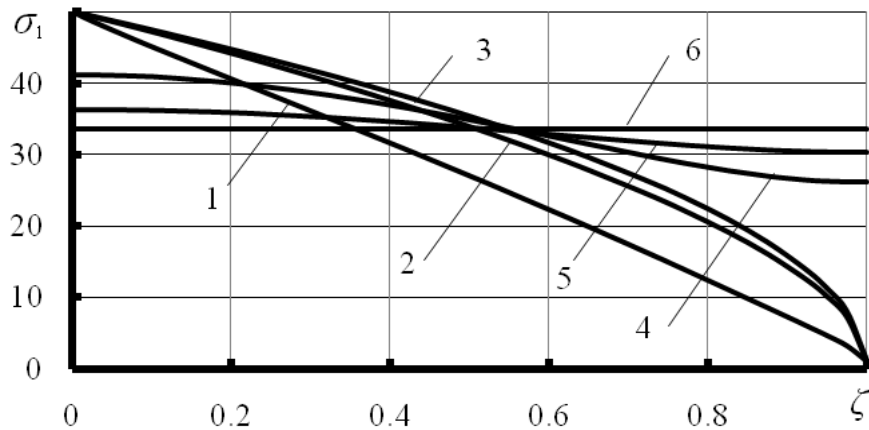
$$\left(\frac{\partial \sigma_1}{\partial \zeta} \right)_{\zeta=0} = \left(\frac{\partial \sigma_2}{\partial \zeta} \right)_{\zeta=0} = \left(\frac{\partial \sigma_1}{\partial \zeta} \right)_{\zeta=1} = \left(\frac{\partial \sigma_2}{\partial \zeta} \right)_{\zeta=1} = 0. \quad (10)$$

3. Results and discussion

In the calculations, it was assumed that: $\varepsilon_{P1} \varepsilon_{V1} = 0.01$; $\varepsilon_{P2} = \varepsilon_{V2} = 0.05$; $R_* = R_{P1}$, $\gamma = 10^{-5}$, $h = 1$ m, $T = 293$ K, $T_M = 3920$ s, $\rho_{AD} = 3000$ kg/m³. We will show the results of two calculation options for $S = 2 \cdot 10^{-8}$ and $S = 2 \cdot 10^{-9}$. Figures 1 and 2 demonstrate changes in the calculation parameters for the first option, and Figures 3 and 4 – for the second.

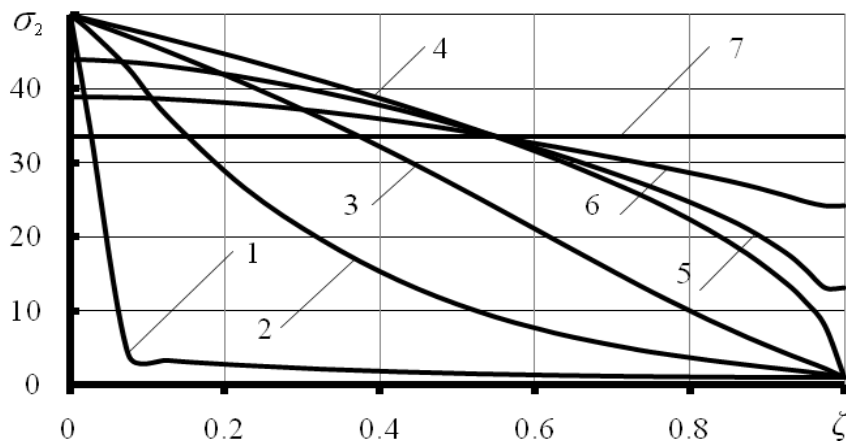
In Figure 1, curves 1–3 refer to the first stage of the process (layer pumping), and the following curves 4–6 refer to the second (layer blockage). We interrupt the first stage, which should end with the establishment of a flow in the channel with crossflows (in our case, it is close to establishment), at $\tau = 0.1$. The second stage ends

with the establishment of an average pressure along the formation (curve 6 approximately corresponds to this).



Curves: 1 – $\tau = 0.002$, 2 – 0.026, 3 – 0.098, 4 – 0.102, 5 – 0.108, 6 – 0.148

Figure 1 – Changes in relative pressure in the first region. $S = 2 \cdot 10^{-8}$, $T_M = 3920$ s



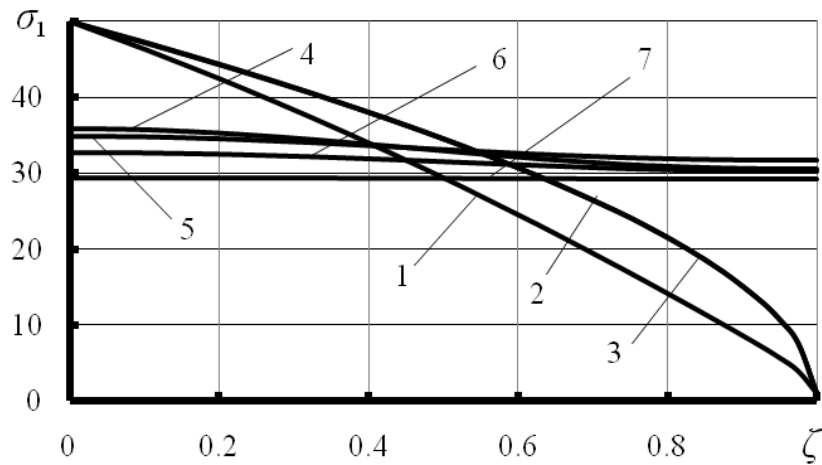
Curves: 1 – $\tau = 0.002$, 2 – 0.008, 3 – 0.016, 4 – 0.098, 5 – 0.102, 6 – 0.106, 7 – 0.148

Figure 2 – Changes in relative pressure in the second region. $S = 2 \cdot 10^{-8}$, $T_M = 3920$ s

It follows from Figure 2 that the pressure, with some delay (this delay is clearly visible on curves 1 and 2), follows the pressure in the first region. Curve 4 in Figure 2 practically coincides with curve 3 in Figure 1. The following curves 5–7 clearly show the establishment of the average pressure in the formation. The adsorption values (not shown here) qualitatively coincide with Figure 2.

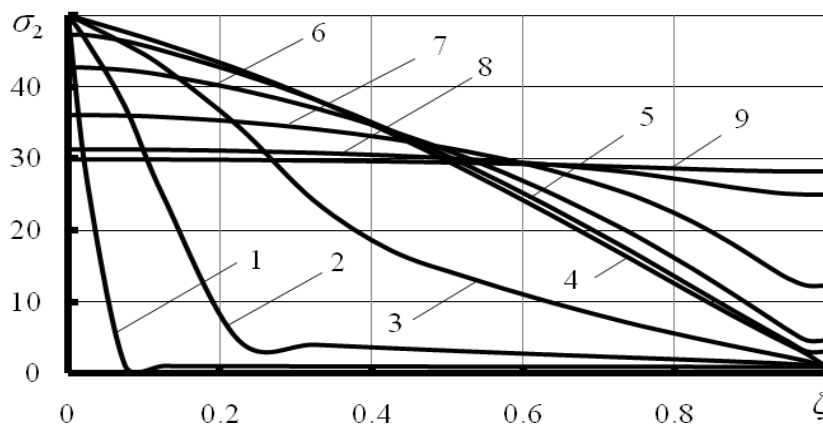
Let us show the results of the second variant, when $S = 2 \cdot 10^{-9}$. The curves in Figure 3 in the first stage are similar to the curves of the same stage in Figure 1, but they differ in the second stage of the process. At the same time, the average level here is somewhat lower, due to the fact that the adsorbed mass of gas during the time $t = T_M \cdot \tau^*$ turned out to be lower here, which is natural. According to the curves in region

2 in Figure 4, it is noticeable that the delay from the first region is stronger. This is due, first of all, to the lower value of S in contrast to the first variant.



Curves: 1 – $\tau = 0.002$, 2 – 0.004 , 3 – 0.098 , 4 – 0.102 , 5 – 0.108 , 6 – 0.12 , 7 – 0.190

Figure 3 – Changes in relative pressure in the first region. $S = 2 \cdot 10^{-9}$, $T_M = 3920$ s



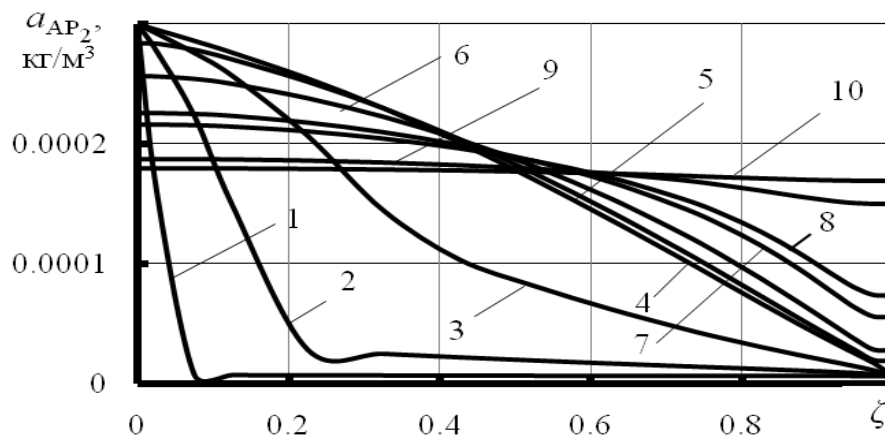
Curves: 1 – $\tau = 0.002$, 2 – 0.024 , 3 – 0.06 , 4 – 0.098 , 5 – 0.102 , 6 – 0.108 , 7 – 0.126 , 8 – 0.16 , 9 – 0.190

Figure 4 – Changes in relative pressure in the second region. $S = 2 \cdot 10^{-9}$, $T_M = 3920$ s

In this case, the final curve 9 in Figure 4, corresponded in time to the final curve 7 in Figure 3, is close to it, but there is still a difference, i.e. it is somewhat lower and more heterogeneous.

Figure 5 shows the obtained adsorption values for the second variant. It is evident from the figure that adsorption follows the pressure changes in the second region with a small delay. The volume adsorption in the second region is close in nature to the curves of Figure 5 (it is not shown in the figure), but due to the selected coefficients, it is significantly smaller in value, shown by a_{P2} . The process of gradual filling of the pore space with the adsorbed mass is also clearly visible, which serves as a clarification of the process picture presented in [11]. Thus, the filtration dynamics of

the adsorbed gas largely depends on the flow system, i.e. on the value of S . This is physically justified, since this value determines the hydrodynamic interaction of two zones of the pore system.



Curves: 1 – $\tau = 0.002$, 2 – 0.024, 3 – 0.06, 4 – 0.098, 5 – 0.102, 6 – 0.108, 7 – 0.12, 8 – 0.126, 9 – 0.16, 10 – 0.19

Figure 5 – Surface adsorption in the second region of a_{AP2} . $S = 2 \cdot 10^{-9}$, $T_M = 3920$ s

4. Conclusions

The calculations performed qualitatively show the dynamics of the adsorption process. It follows from the calculations that it largely depends on the structure of the pore space (in our case, to which zone the adsorption capacity belongs). The characterizing adsorption coefficients themselves undoubtedly play an important role in the dynamics and at certain process times can significantly affect the final result. The further picture of the process is associated with the diffusion restructuring of the adsorbed gas mass, but this process has a different time scale.

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ФІЛЬТРАЦІЯ ДИНАМІЧНО АДСОРБУЮЧОГО ГАЗУ У БІДИСПЕРСНОМУ ПОРИСТОМУ ШАРІ

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Анотація. Серед завдань, пов'язаних з фільтрацією та масообміном у пористих тілах найбільшого інтересу нині набувають завдання адсорбційно-десорбційного напрямку. Це пов'язано з широким спектром проблем: безпекою гірничих та вугільних розробок, видобутком горючих газів, зберіганням парникових газів у пластах. Разом з тим у теоретичному плані набувають широкої популярності питання, пов'язані з різномасштабністю тріщинувато-пористих середовищ, зокрема, гірських та вугільних пластів.

У цій роботі розглядається завдання про фільтраційну течію при закачуванні газу в пористий пласт з урахуванням адсорбції газу, як на поверхні каналів у макропорах, так і при об'ємному осадженні у мікропорах. У роботі приймається, що весь поровий простір вугільного пласта, згідно з результатами останніх досліджень, може бути поділено на дві частини взаємно пов'язаних областей, що відрізняються одна від одної своїми характерними розмірами порових каналів і, відповідно, з великою відмінністю проникностей. Основна увага приділяється фільтраційним особливостям процесу течії газу, що адсорбується в пласті. У основу розробленої моделі покладено теорію безінерційного руху газу, тобто, з використанням рівняння Дарсі, записаного у кожній зоні окремо. Передбачається, що адсорбційний процес підпорядковується, згідно з літературними даними, рівнянню ленгмюрівського типу. У задачі розглянуто дві стадії процесу: закачування газу в пласт і процес встановлення фільтраційного тиску при закупорці пласта. Показано зміни тиску у двох різних канальних системах залежно від інтенсивності перетікання газу з однієї порової гілки системи до іншої. При інтенсивних перетіканнях перепади тиску між областями незначні. При слабких перетіканнях, перепади великі, що повинно призводити до великих внутрішніх напруг. Проведені розрахунки якісно показують динаміку адсорбційного процесу під час руху газу в пласті. З розрахунків випливає, що вона великою мірою залежить від структури порового простору. При інтенсивних перетіканнях процес адсорбції практично слідує зміні тиску у всьому поровому просторі. Для слабких перетоків великі перепади тиску, що виникають між двома областями, позначаються на процесі адсорбції.

Ключові слова: пористе середовище, газ, дифузія, масообмін, фільтрація, адсорбція, мікропористі канали.