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THE INFLUENCE OF GREAT DEPTHS ON THE CONDITIONS ACTIVATING DESORPTION OF ADSORBED METHANE GENERATED IN A VIRGIN COAL SEAM

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Abstract. It is generally accepted that when a coal seam is in a virgin rock massif for millions of years under the influence of rock pressure, its state is characterized by the absence of any physical processes in it.

In this article, it is stated that in reality, in these conditions relaxation processes can occur. These processes are caused by conformational rearrangements in the coal microstructure. They, in turn, create conditions for breaking the sorption equilibrium of adsorbed methane in coal due to the distance of methane molecules from the microstructure of the coal medium when the interlayer distances in the graphite-like layers of the coal seam expand.

The parameters are established, which determine the duration of the coal molecular structure relaxation, characterize ability of the structure to deform and the conditions under which the most significant changes occur. It is shown that the main parameters which affect the activation of desorption of adsorbed methane in a virgin coal seam are the energy of conformational rearrangements activation, the temperature of the seam and the interlayer distance in the graphite-like layers.

As a result of the research, it was found that with increase in the depth of the virgin coal seam, relaxation processes in it occur faster. These processes are caused by conformational rearrangements in the coal microstructure. They, in turn, create conditions for breaking the sorption equilibrium of adsorbed methane in coal due to the distance of methane molecules from the microstructure of the coal medium when the interlayer distances in the graphite-like layers of the coal seam expand.

The novelty of this research is in the establishment of the fact that with an increase in the depth of the virgin coal seam, all physical and mechanical processes associated with changes in the molecular structure of coal occur faster. At the same time, the energy of activation of these processes realization is less, including the energy of activation of adsorbed methane desorption. That is, the great depths of the coal seam accelerate and contribute to the physical processes that lead to the activation of the desorption of adsorbed methane.

Keywords: adsorbed methane, coal seam microstructure, activation energy, conformational rearrangements, graphite-like layers, interlayer distances, diffusion.

1. Introduction

Today, development of modern technologies for methane recovery and safe mining of coal seams is based on data on the volumes of methane in the rock-coal mass, which is generated in virgin coal seams for millions of years, and the conditions of its release. However, previously conducted studies that touch on these issues are mainly either on the level of hypotheses [1], or the researchers studied for years the results of methane release from the coal samples taken from wells without taking into account the peculiarities of the patterns of desorption processes in the seam [2–5]. It should be noted that today there are a lot of works for disturbed coal massif, which relate to studies of adsorbed methane in coal seams. For example, a complex of works carried out by the Institute of Rock Physics of the National Academy of Sciences of Ukraine [6–11]. However, these works do not make it possible to investigate the state of adsorbed methane from the moment of its generation and the conditions of activation of desorption in a virgin coal seam. Therefore, the work [12], which examines the process of generating adsorbed methane in a coal seam, which has been under the influence of rock pressure for millions of years, can be considered the most correct theoretical research in this direction. It is shown that as a result of the transformation of

the molecular structure of the coal seam, the separation of the methyl group and hydrogen from the aliphatic fringe occurs, which, being combined, create methane molecules [12]. These molecules immediately enter into a sorption relationship with coal. It was established that at depths deeper than 1000 m, more than 60 m^3/t of adsorbed methane can be generated in coal. Therefore, the process of activation of methane desorption can occur in the coal seam, which will lead to methane mass transfer to the environment. In order to establish the conditions for activation of desorption of methane generated in coal and the effect of great depths on them, the following goal was set for this work.

The goal of the work. To investigate the conditions for activation of methane desorption due to the breaking of sorption equilibrium of methane generated in a virgin coal seam for millions of years and to establish the influence of great depths on these conditions.

2. Methods

It is known [13, 14] that molecular structure of coal, where adsorbed methane is generated as a result of regrouping of trace elements, can be represented by the space of closed and open pores in coal, which are interconnected by graphite-like layers, as shown in Figure 1.

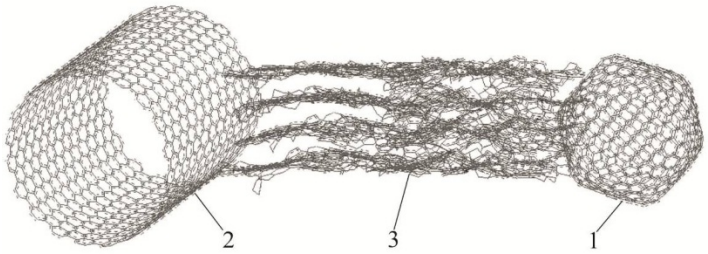

Figure – 1 Molecular model of closed (1) and open (2) micropores, which are connected by graphite-like layers of coal medium (3)

In the microstructure shown in Figure 1, the highest numerical values are reached by the sorption forces which, as it is known from [15, 16], bind methane molecules to the walls of closed pores, the microstructure of which is not compressed. Therefore, they are mostly filled with methane during its generation [17, 18]. But at the same time, the energy of activation of desorption from closed pores is significantly lower than from the interlayer space of graphite-like layers and from open pores [16]. Besides, the increase of interlayer distances in graphite-like layers saturated with adsorbed methane leads to a decrease in the sorption bonds between methane and coal. Therefore, these factors are the main ones when assessing the thermodynamic stability of these molecular structures. At the same time, their analysis shows that structural changes in the coal seam can occur only in graphite-like layers, since the microstructure of closed pores is not compressed. Besides, it should be taken into account that the energy of methane desorption activation in the microstructure of a coal seam is different in the micropores of coal and in graphite-like layers [19]. In the latter

case, it is 2–3 times greater and, according to [16], depends primarily on the conformational capacity of the coal microstructure, which is its invariable characteristic [20]. Therefore, the basic ground for achieving the goal set in this work is the regularity of distribution of the energy of activation of these conformations for different degrees of coal metamorphism, which is presented in Figure 2 [16].

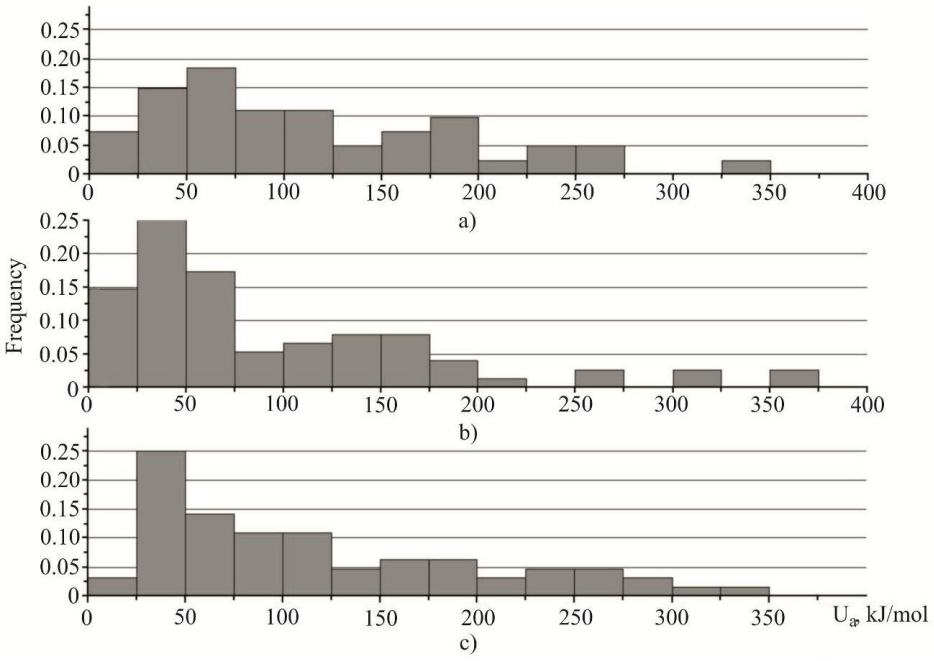

Figure – 2 Histogram of frequencies of conformational rearrangements energy in the molecular structure of coal - *Ua* with a carbon content of 87.7% (a), 89.1% (b) and 91.26% (c) [15]

It can be seen in the histograms shown in Figure 2 that the distribution of conformations in coal of all degrees of metamorphism has a similar character and occurs within the same limits of reaching their activation energy, which are from U_a=25 kJ/mol to U_a=300 kJ/mol. At the same time, the maximum frequency of manifestations of conformational rearrangements is realized at the level below U_a=200 kJ/mol. In this regard, the researches within the framework of the set goal can be carried out with one degree of coal metamorphism and for a limited interval of energy of conformations activation. For example, with the coal, which has the greatest bond of sorption interaction with methane and in which the largest volume of methane is generated. That is, for coal with a carbon content of 87.7% in the range of energy of conformations activation U_a=(25–200) kJ/mol.

It should be noted that the ability of coal to undergo conformational rearrangements, which are realized without destroying chemical bonds between atoms, creates the mechanism of deformation of the molecular structure of coal as a high-molecular medium [21, 22]. That is, when a virgin layer is under the influence of rock pressure for many years, not only methane is generated in it, but also its stress-deformed state is not stable, since the macro and microstructure of coal is deformed over time. Deformation of this structure is characterized by a strong influence of loading speed, stress relaxation during constant deformation, and other features [22]. In this case,

relaxation processes in high-molecular media, including the coal molecular structure, can be described by the Arrhenius equation [22]:

$$\tau_p = \tau_0 \, exp\left(\Delta U / RT\right), \tag{1}$$

where τ_p is duration of the segments relaxation in the macromolecule; τ_0 is the constant of microstructure of the medium, which depends on the frequency of oscillations of atoms in the molecule and for coal it is equal to τ_{p0}=–(12–13) s [16]; ΔU is the energy of the relaxation process activation; R is gas constant; T is the temperature of the seam, which is determined by the depth of its bedding.

In equation (1), the energy of relaxation process activation $-\Delta U$ is determined by the energy of conformational rearrangements activation in the coal microstructure – E_a^{κ}. Therefore, in this case, ΔU is equal to E_a^{κ}. In turn, conformational rearrangements in the coal structure change the interlayer distances in the graphite-like layers due to the reorientation of carbon chains in the aliphatic fringe of the coal substance. The theoretical regularity of these changes, which was established in the work [16] by calculation of potential energies of rotation of torsional angles of the conformer in the microstructure of coal, is presented in Figure 3.

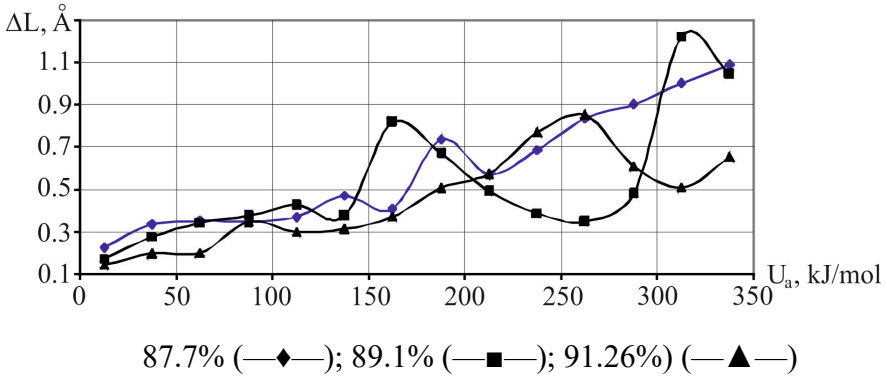

Figure – 3 Changes of average values of the interlayer distance increase in the ranges (ΔL) of the energy of conformational rearrangements activation (U_a), 25 kJ/mol, at different degrees of coal metamorphism [16]

It should be noted that the interlayer distance increase in Figure 3 reduces the sorption bond between methane and molecular structure of coal due to the reduction of the Lennardo-Jones potential energy, which describes the repulsion or attraction of molecules among themselves depending on the distance between them.

Besides, such a parameter as the coal seam temperature, which is limited by the temperature of great depths, is of great importance. Today, it is accepted that these depths begin at H=800 m, where coal is a classic high-molecular substance, and reach approximately H=1400–1500 m. This is due to the fact that with further increase in temperature, signs of the viscous-fluid state of coal can appear [21, 22]. Therefore, it is advisable to carry out the research within the specified depths of the coal seam

bedding.

The temperature of a coal seam, which is determined by the depth of its bedding, is calculated by geothermal gradient according to the formula [23]:

$$T_{gr.} = 100 \cdot \frac{(T_2 - T_1)}{(h_2 - h_1)}, \quad (2)$$

where: $T_{gr.}$ is geothermal gradient of rock temperature change for every 100 m of depth of coal bedding; T_2 and T_1 are temperatures at depths h2 and h1, respectively.

From formula (2), we have:

$$T_2 = T_1 + (T_{gr.}/100)(h_2 - h_1). \quad (3)$$

Therefore, the algorithm for assessing the diffusion of methane adsorbed in the microstructure of coal, when it is generated in a virgin coal seam, at the degree of coal metamorphism of 87.7% and H=800 m;1400 m, includes the following stages:

1. Determination the coal seam temperature at depths of 800 m and 1400 m by formula (3).

2. Determination by formula (1) of the relaxation duration corresponding to the energy of confirmation rearrangements activation in the molecular structure of a coal seam with a carbon content of 87.7% in the interval E_a^{κ}=(25–200) kJ/mol (Fig. 2a).

3. Establishment and analysis of graphical dependencies $\tau_p = \tau_p(E_a^{\kappa})$ for the depths of the coal seam bedding under the research.

4. Determination of the values of interlayer distances in the molecular structure of the coal seam at different energies of conformational rearrangements activation with taking into account increase of interlayer distances, which is presented in Figure 3.

5. On the basis of the dependencies determmined in paragraphs 3 and 4, establishment of the relationship between duration of relaxation of the coal molecular structure deformation due to its conformational rearrangements over many years and the interlayer distances in the graphite-like layers of the coal seam.

6. According to the results of the research in paragraphs 2–5, analysis of simultaneous influence of the energy of conformational rearrangements activation in the microstructure of coal and the relaxation duration on changes in its interlayer distance in a graphite-like layer.

3. Results

According to the data of the work [23], it is known that geothermal gradient in the Donetsk basin is usually within 2.2–2.6°C per 100 m [23], and, hence, averages 2.4°C per 100 m. At the same time, temperature T_1 of this basin is 20.5°C at a depth of 400 m [23]. Therefore, the temperature at the depths, which are studied and calculated according to the equation (3), will be equal to T=303.1 K° for H=800 m and T=317.5 K° for H=1400 m. Then the influence of the depth on the duration of the

coal molecular structure relaxation, which is calculated by equation (1), with the energies of conformational rearrangements activation within the range E_a^{κ}=(20–200) kJ/mol, can be represented by the graphical dependence shown in Figure 4.

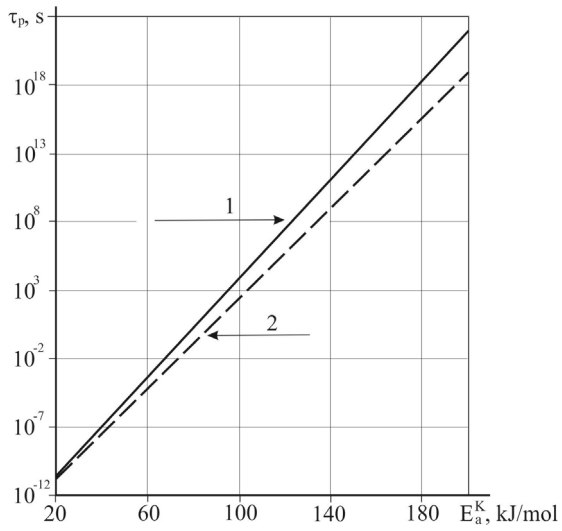

Figure 4. The regularity of changes in the relaxation duration τ_p depending on the energy of conformational rearrangements in the molecular structure of the coal seam E_a^{κ} at different depths of its bedding, where 1 is H=800 m, 2 is H=1400 m

It can be seen in Figure 4 that, as the energy of conformational rearrangements increases, the relaxation duration increases according to a linear law in the entire interval of changes of this energy at each depth of the coal seam. At the same time, the graphic dependence in Figure 4 demonstrates a rather strong influence of temperature and, as a result, the depth of the coal seam bedding on the duration of the coal molecular structure relaxation. For example, range of the energy of conformational rearrangements activation of 112.5 kJ/mol, the relaxation time at a depth of 800 m is equal to τ_p=2.54·10^5 s, and at a depth of 1400 m is equal to τ_p=3.3·10^4 s. That is, the difference corresponds to an almost tenfold decrease of the relaxation duration when the depth of coal seam bedding increases by 600 m.

The second important step for assessing conditions for activation of adsorbed methane desorption during its generation in a coal seam at great depths is the analysis of the change in interlayer distances in the graphite-like layers of a coal seam at different energies of conformational rearrangements activation in the coal microstructure. Figure 3 shows the theoretical regularity [16] of the change of the increase of the specified parameter Δl depending on the energy of conformational rearrangements activation of the coal molecular structure. It can be seen in this Figure that the given regularity has a wave-like character. As it is known from [16], it is due to the peculiarities of the mechanism of rearrangements of the structural chains of the coal substance, which is realized when the energy potential of the molecular structure of coal is changed as a result of the step-by-step accumulation of potential energy. This, in turn, leads to a change in the energy of valence bonds in the microstructure and, as a

result, to a change in interlayer distances in graphite-like layers. It should be noted that the described deformation mechanism also determines the essence of the diffusion jump of desorbed methane in coal, which is described in [16]. Therefore, the analysis of the influence of depth was carried out based on the maximum values of the interlayer distances increase, presented in Figure 3. It was taken into account that the interlayer distance of the coal with a carbon content of 87.7% in the absence of disturbance of the coal microstructure is equal to l_0=0.39 nm [16]. That is, the full values of interlayer distances were established, which, in turn, contribute to the activation of adsorbed methane desorption. It should be emphasized that the main parameter, on which the interlayer distance ($\Delta l+l_0$) depends, is the energy of conformational rearrangements activation E_a^{κ}. In turn, this parameter is closely related to the relaxation duration and depth resulting from formula (1) by the following relationship:

$$E_a^{\kappa} = RT\,ln\left(\tau_p/\tau_0\right).$$

Therefore, when establishing the regularity of the change of the interlayer distance at different depths of the coal seam bedding, such parameter as the energy of conformational rearrangements activation in the molecular structure of coal E_a^{κ} was matched with such parameters as the duration of this microstructure relaxation at different depths of the coal seam. As a result, the regularity of the change of interlayer distances ($\Delta l+l_0$) due to the E_a^{κ} and τ_p at different depths of the coal seam is presented in Figure 5. It should be noted that in this figure the three horizontal axes of the abscissa indicate that, for example, the interlayer distance $\Delta l+l_0$=0.426, which corresponds to point A on the ordinate and is reached when E_a^{κ}=50 kJ/mol and τ_p=10^{-5} s at H=800 m and when τ_p=10^{-6} s at H=1400 m.

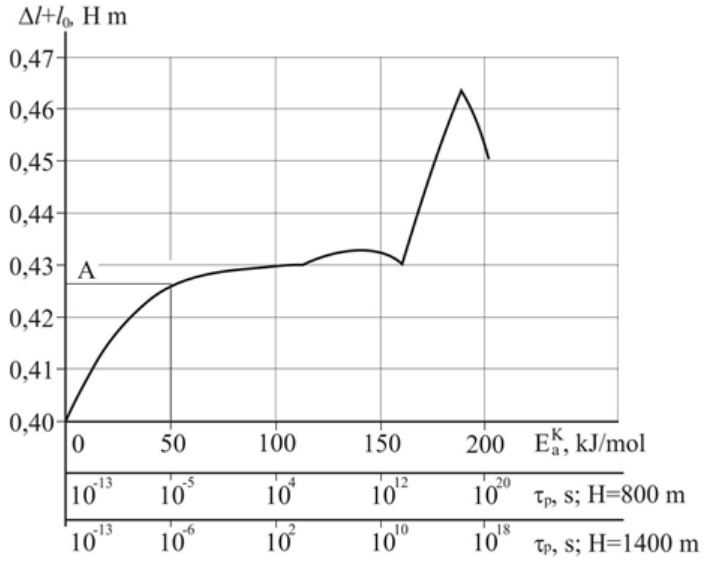

Figure – 5 Dependence of the change of interlayer distances in the graphite-like layers of a coal

seam on the energy of conformational rearrangements activation E_a^{κ} and relaxation duration τ_p in the molecular structure of the coal seam at its bedding depths of 800 m and 1400 m

The analysis of regularity presented in Fig. 5 shows that when the energy of conformational rearrangements activation increases to 162 kJ/mol the interlayer distance increases by a parabolic law, and then, with increase of E_a^{κ}, increases sharply to the energy of conformational rearrangements activation, which is equal to E_a^{κ}=187 kJ/mol. In this case, when H=800 m then τ_p=1018 s, and when H=1400 then τ_p=1016 s. That is, at a greater depth, the same values of interlayer distances are reached faster. In this case, at a depth of 1400 m, which is 600 m greater, the same interlayer distances are reached 100 times faster, starting with the energy of conformational rearrangements activation of 100 kJ/mol. Up to this value, when E_a^{κ}≤100 kJ/mol, this effect occurs at a tenfold difference in relaxation duration τ_p. In addition, it can be seen in Figure 5 that at the beginning, when the energy of conformational rearrangements activation in the molecular structure of coal E_a^{κ} increases, the potential energy also increases, accumulates and then is realized in the form of a sharp jump in the interlayer distance in the graphite-like layer of the coal seam. At the moment of this jump, as it is known from [16], the process of adsorbed methane desorption in the coal seam is activated. Moreover, this effect takes place both at the depth of the coal seam H=800 m and the depth H=1400 m only with different relaxation duration that correspond to them. That is, at all relaxation durations, all the parameters that are most important for activation of adsorbed methane desorption, namely the energy of confirmation rearrangements activation and interlayer distances in graphite-like layers, will change faster at greater depths of the coal seam. This creates favorable conditions for activation of methane desorption.

4. Discussion

As it is determined in [12], the volumes of methane generated in virgin coal seams over millions of years can reach values of up to 60 m^3/t and more. This volume of gas is much greater than the coal can hold in its structure. Therefore, many questions arise related to the assessment of physical state of this gas in coal, conditions of its equilibrium breaking, influence of the coal microstructure and mining and geological conditions of the coal seam bedding on gas and other related factors. The conducted research presented in this work answers many questions in this area. Thus, now it becomes clear how this gas is related to the microstructure of coal, why it changes over many years. As well as which factors and how they affect the state of methane in coal and which conditions break its equilibrium. However, this is only the beginning of the research, what one needs to know in order to get answers to the main questions. That is how much methane generated in the coal seam remains in its microstructure, how methane will be released into the environment. It is also important to know in which direction the optimal ways of redistribution of methane generated in the rock massif will be directed and how depth of the coal seam bedding influences

on these processes.

The results of the research carried out in this work can serve as a basic ground for establishing the answer to the above questions.

Knowing these answers is very important for the development of safe and efficient technologies for the extraction of minerals and methane at great depths of the coal massif.

5. Conclusions

So, when a coal seam is in a virgin rock massif for millions of years under the influence of rock pressure, its state can only be imagined as an equilibrium, which is characterized by the absence of any physical processes occurred in it. However, this is only wishful thinking. In reality, the massif does not only generate adsorbed methane as a result of separation from the aliphatic fringe of methyl groups and hydrogen, which, when combined, create methane molecules, as established in [12]. In these conditions, other processes also occur, which are considered in the studies presented above. Thus, the conducted research showed that the main characteristic of the rock massif bedded at great depths is that relaxation processes take place in it as well. These processes are caused by conformational rearrangements in the coal microstructure. They, in turn, create conditions for breaking the sorption equilibrium of adsorbed methane in coal due to the distance of methane molecules from the microstructure of the coal medium when the interlayer distances in the graphite-like layers of the coal seam expand. That is, the current state of the virgin coal massif is only a function of time.

In turn, duration of the coal molecular structure relaxation will be determined by such independent parameters as temperature of the coal seam, which is determined by the depth of its bedding - H and the energy of conformational rearrangements activation in the microstructure of coal - E_a^{κ}. The latter characterizes the ability of the coal structure to deform. It was established that the range of energy E_a^{κ}, at which the most significant conformational rearrangements take place in the coal molecular structure , is in the range from 50 kJ/mol to 200 kJ/mol. Range of this energy does not depend on the depth of bedding and other parameters. Only the duration of the coal molecular structure relaxation at different depths of the coal seam bedding will change in it significantly.

So, the established regularity of changes of the relaxation duration τ_p due to the E_a^{κ} showed that with an increase in the energy of conformational rearrangements, the relaxation duration increases according to a linear law in the entire interval of this energy changes. At the same time, the depth of the coal seam bedding has a rather strong influence on the relaxation duration. For example, for the energy of conformational rearrangements activation of 112.5 kJ/mol, the relaxation duration is $2.54 \cdot 10^5$ s at a depth of 800 m and $3.3 \cdot 10^4$ s at a depth of 1400 m. That is, the difference corresponds to an almost tenfold decrease in relaxation duration when the depth of the seam bedding increases by 600 m. This indicates that with an increase of the depth of

the coal seam bedding, the relaxation processes in it proceed faster.

In turn, since the relaxation duration is related to the energy of conformational rearrangements activation in the microstructure of the coal, the change in the relaxation duration leads to a change in the microstructure of the coal, which is represented by graphite-like layers. Their main parameters are interlayer distances in this microstructure. Therefore, when the relaxation duration is changed, this parameter will be the first to change. In this regard, the regularity of the change of interlayer distances in the graphite-like layers of a coal seam due to the energy of conformational rearrangements activation and duration of the coal molecular structure relaxation at the depths of its bedding of 800 m and 1400 m was established, which showed the following.

When the energy of conformational rearrangements activation increases to 162 kJ/mol the interlayer distance increases according to the parabolic law, and then, with an increase of E_a^{κ}, it sharply increases to the energy of conformational rearrangements activation, which is equal to E_a^{κ}=187 kJ/mol. In this case, $\tau_p=10^{I8}$ s when H=800 m and $\tau_p=10^{16}$ s when H=1400 m. That is, at a greater depth, the same values of interlayer distances are reached faster. In this case, at a depth of 1400 m, which is 600 m greater, interlayer distances are reached 100 times faster, starting with the energy of conformational rearrangements activation of 100 kJ/mol. Up to this value, when E_a^{κ}<100 kJ/mol, this effect occurs with a tenfold difference in τ_p. At the same time, when the energy of conformational rearrangements activation increases, the potential energy in the molecular structure of coal first increases, accumulates and then is realized in the form of a sharp jump in the interlayer distance in the graphite-like layer of the coal seam. At the moment of this jump, as it is known from [16], the desorption process in the coal seam is activated.

Thus, with an increase in the depth of a virgin in coal seam, all physical and mechanical processes associated with changes in the molecular structure of coal occur faster. The energy of activation of these processes realization is less, including the energy of activation of adsorbed methane desorption. That is, the great depths of the coal seam bedding accelerate and contribute to all physical processes which lead to the activation of the desorption of adsorbed methane generated in the coal seam.

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ВПЛИВ ВЕЛИКИХ ГЛИБИН НА УМОВИ АКТИВАЦІЇ ДЕСОРБЦІЇ АДСОРБОВАНОГО МЕТАНУ, ГЕНЕРОВАНОГО У НЕЗАЙМАНОМУ ВУГІЛЬНОГО ПЛАСТІ

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Анотація. В статті показано, що коли вугільний пласт знаходиться у незайманому гірничому масиві мільйоні років під дією гірничого тиску, то його стан характеризується відсутністю протікання у ньому яких-небудь фізичних процесів.

В реальності в цих умовах реалізуються також інші процеси, пов’язані з реалізацією в ньому релаксаційних

процесів. Ці процесі обумовлені конформаційними перебудовами у мікроструктурі вугілля. Вони, в свою чергу, створюють умови порушення сорбційної рівноваги адсорбованого метану у вугіллі за рахунок віддалення молекул метану від мікроструктури вугільного середовища при розширенні міжшарових відстаней у графітоподібних шарах вугільного пласта.

Встановлені параметри, що визначають час релаксації молекулярної структури вугілля, характеризують її спроможність до деформування та умови при котрих здійснюються самі значні її перебудови. Показано, що основними параметрами, які впливають на активізацію десорбції адсорбованого метану у незайманому вугільному пласті є енергія активації конформаційних перебудов, температура пласта та міжшарова відстань у графітоподібних шарах.

В результаті досліджень встановлено, що при збільшенні глибини залягання незайманого вугільного пласта ньому релаксаційних процесів. Ці процесі обумовлені конформаційними перебудовами у мікроструктурі вугілля. Вони, в свою чергу, створюють умови порушення сорбційної рівноваги адсорбованого метану у вугіллі за рахунок віддалення молекул метану від мікроструктури вугільного середовища при розширенні міжшарових відстаней у графітоподібних шарах вугільного пласта.

Новизна виконаних досліджень полягає в тому, що встановлено, що при збільшенні глибини залягання незайманого вугільного пласта усі фізико-механічні процеси, які пов'язані зі зміною молекулярною структури вугілля відбуваються швидше. При цьому енергія активації реалізації цих процесів менше, у тому числі, і енергія активації десорбції адсорбованого метану.

Ключові слова: адсорбований метан, мікроструктура вугільного пласта, енергія активації, конформаційні перебудови, графітоподібні шари, міжшарові відстані, дифузія.