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MECHANIC-CHEMICAL MODEL OF METHANE GENERATION IN THE DESTRUCTION OF CARBON SUBSTANCE

Skipochka S.I., Krukovskyi O.P., Palamarchuk T.A., Prokhorets L.V.

Institute of Geotechnical Mechanics named by N.Poljakov of National Academy of Sciences of Ukraine

Abstract. The subject of research presented in the article is the anomalous release of methane from coal seams during their development. The purpose of the development is to estimate the maximum possible volumes of the main phase components of methane containing the coal seam, substantiation and development of a methane generation model during the destruction of coal. The analysis and generalization of own and third-party results experimental and theoretical researches executed by methods of X-ray diffraction analysis, nuclear magnetic and electronic paramagnetic resonance, raster microscopy, infrared spectroscopy, physical and mathematical modeling are used in the work. It is established that the role of solid carbon-methane solution, which dominates in the theory of gas-dynamic phenomena, and its share in the total volume of methane is significantly exaggerated. The main inconsistencies of this theory relate to three provisions. Discrepancies in the volumes of methane in solid solution to the volumes registered during the gas-dynamic phenomenon. Incompatibilities of the velocities of diffusion-filtration processes during the decomposition of a solid solution and the course of a gas-dynamic phenomenon. Inconsistencies in the sequence of ejection and formation processes of mad flour. The developed model of methane anomalous manifestations in the destruction of coal, as one of the main, takes into account the process of methane generation. The model assumes three phases of the gas-dynamic phenomenon. Long-term, including the formation of the geological environment and the influence of technological factors. The main duration is 5-15 s, which occurs as a result of the coal destruction and includes all possible physical, chemical and mechanic-chemical processes, primarily the release of free methane, mechanic-chemical reaction of methane synthesis (methane generation), desorption of physically bound methane and strengthening of this component by mechanics-electrics processes, decomposition of solid carbon-methane solution and release of gas-hydrate component of gas. The quenching phase of the process for several hours, during which of solid carbon-methane solution decomposition continues and relaxation gas-dynamic and geomechanical processes take place. The work is directed at improving the safety of miners by clarifying the theory of gas-dynamic phenomena and improving on this basis methods and means to prevent them.

Keywords: coal mine, gas-dynamic phenomena, theories of phenomena, mechanics-chemical processes, methane generation, model.

Introduction. During the period of underground development of coal deposits in the world, more than thirty thousand sudden emissions of methane and coal were recorded, as a result of which almost sixty thousand people died. According to these indicators, sudden emissions in mines are among the most catastrophic phenomena of natural and man-made nature. A huge number of scientific works dating back to the second half of the nineteenth century are devoted to the study of the nature of these phenomena. However, the first generalizing results appeared only in the middle of the twentieth century.

It is known that the sudden release of coal and gas is a spontaneous, and fast-moving, process of fragile self-sustaining destruction of rocks, which is accompanied by the release of gas from the destroyed coal and its movement in the gas stream. The degree of this phenomenon danger depends on its scale and is determined by the amount of ejected rock mass, the intensity of the process and the parameter of gassiness.

The first thorough studies of sudden emissions are presented in [1, 2]. In particular, academician O.O. Skochynskyi logically and consistently reflected the nature of the process development, the forces involved in it, characteristics of the process and the consequences of the phenomenon. Further research was aimed at refining this model [3-6]. In particular, initially sudden emissions in mines, rock strikes and other dynamic manifestations of rock pressure were classified as phenomena of one nature. Subsequently, a number of hallmarks of sudden release were pointed out. They put the phenomenon in the category of unusual in relation to other manifestations of rock pressure. The peculiarity lies in the energy, which is realized as a shock wave energy, the work of coal removal from the reservoir over long distances, fine grinding of coal, abnormally high methane emissions and high potential energy of the rock mass.

As the amount of energy released was such that it significantly exceeded its estimated, there was a need to deepen research on the physical structure of coal. Work in this direction allowed us to reject a number of unsubstantiated hypothetical models based on the dominant role of gas energy and gas formation pressure, and to dwell on the hypothesis of a multi parameter mechanism of the phenomenon. The quintessence of this hypothesis is the model according to which the gas-dynamic phenomenon (GDP) is a loss of a geomechanical system stability in which the accumulated potential energy has exceeded some critical level. The phenomenon is accompanied by a sharp transition to the energy of mechanical destruction of molecular bonds and the kinetic energy of destroyed rock mass emission and gas. The catalyst for the development of emissions in this interpretation can be any, even minor, external influences, such as the launch of the mechanism in the face of the mine [7-9].

This hypothetical model is the basis of the "energy" theory of sudden emissions of coal and gas in mines. According to the GDP theory, its arise as a result of the simultaneous action of high mechanical stresses, high gas bearing capacity and low gas permeability of the formation, as well as the tendency of coal to mechanical destruction when the stress state changes. The model assumes that the main condition for initiating GDP is the release of the potential energy amount, which would be enough to perform the work of moving coal in the direction of mine working. For the above process to take place, the potential energy of the gas-saturated rock mass must be converted into kinetic emission energy. The impetus is external forces arising under the influence of technological processes. That is, the energy balance should be such that the sum of the accumulated energy components was sufficient to perform the work of destruction and movement of the gas mixture and crushed coal in mine working.

The model and theory are phenomenological. However, they do not raise special questions about their reliability.

It should be noted that the experience of using GDP prevention measures created on the basis of the "energy" model-theory has shown their insufficient effectiveness. A significant number of methane anomalous manifestations, which include gas-dynamic phenomena, and has not found convincing scientific explanations. This was the impetus for deepening and refining the theory of GDP in coal mines. Especially with regard to such key issues as the determination of methane phase states during geological formation and during coal seam development, as well as the reasons for the significant discrepancy between expected and actual methane emissions in mines.

With regard to the main phase components of methane in coal seams, the "energy" theory of GDP is based on the fact that in fossil coal methane already exists a priori, formed in different geological periods and is in free and sorbet states. Therefore, the processes of gas evolution in coal production are associated with its release from the cavities, desorption and filtration. Based on these phenomena, the estimated maximum volume of methane that can be released from one ton of coal is not more than 45 m^3 [10-12]. Therefore, in recent years, a number of researchers, not finding an answer to the question that explains the anomalous volumes of gas emissions, agree with the hypothesis that 70–85 % of methane is probably contained in the intermolecular space of coal in the form of solid carbon-methane solution [13-15]. This interpretation raises a number of doubts and requires additional, more in-depth studies of coal at the atomic-molecular level, taking into account the physical and chemical processes in the destruction of coal.

The aim of the work is to develop a mechanic-chemical model of methane generation during the destruction of coal matter.

Methods. The paper uses analysis and generalization of the results of own and third-party experimental and theoretical studies performed by X-ray diffraction analysis, nuclear magnetic and electronic paramagnetic resonance, scanning microscopy, infrared spectroscopy, physical and mathematical modeling.

Results and discussion. Regarding the hypothesis of the predominance of the methane solid phase over others, we note, first, that the main share of methane in the amount of 150–200 m^3 per ton of coal is released during the gas-dynamic phenomenon in the first seconds of the process. This does not agree with the parameters of solid-state diffusion (the diffusion coefficient in solids is from 10^{-9} to 10^{-14} cm^2/s). Secondly, if we talk about solid solutions, the proportion of crystalline phase in coal is an order of magnitude less than amorphous. Therefore, it is difficult to imagine that from 150 m^3 to 200 m^3 of methane contained in the volume of coal, not exceeding 0.08 m^3. Third, the analysis of this mechanism raises the question of the primary and secondary phenomena of diffusion and micron-dispersed grinding of coal in the process of emission. If the model of solid carbon-methane solution is taken as a basis, then micron-disperse grinding of coal substance should be primary. This is almost impossible to explain, because it is the processes of formation and release of methane that provoke the formation of so-called "crazy flour".

In order to deepen scientific knowledge about the nature and mechanisms of formation and provocation of gas-dynamic phenomena in the first stages of this work [16-20], a physical and mathematical model of methane formation processes in coal seams was developed and an analytical assessment of the methane contribution to the coal seams. In particular, the following is established.

1. The maximum volume of free methane in coal seams, depending on their geological properties, at depths up to 300 m can be up to 14 m^3 per ton of coal, and at depths up to 1000 m – up to 30 m^3/t.
2. The maximum possible volume of adsorbed methane can be up to 20 m^3/t.
3. The maximum volume of methane in solid solution can be 40 to 60 m^3/t.
4. The gas hydrate component of methane in the coal seam reaches 6 m^3/t.

The possible volume of methane in the pores may be within (10–50) m^3/t.

Thus, at the maximum possible average volume of methane in the coal seam of 100 m^3/t, the solid component should be about half of the total. The remaining part of methane, in a ratio of about two to three, is usually distributed between free methane inside macropores, microcracks and other defects in the integrity of coal and adsorbed gas (including volumetric filling of transition pores and macroscopic defects). This provision is quite justified, as confirmed by numerous direct measurements in mine conditions. The ratio of these phase components is illustrated by the diagram in Fig. 1.

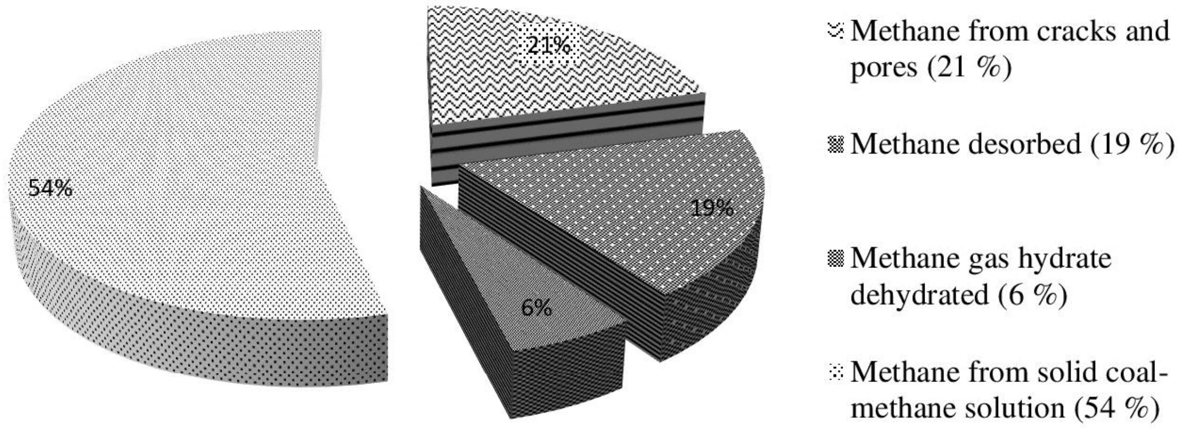

Figure 1 – Maximum possible release of the main natural phase constituents of methane from excavated coal for an unlimited period of time – (100) m^3/t

Note that all calculations were performed under the assumption that each of the accumulation processes and release of methane occurs under the most favorable thermodynamic conditions. The structure of the array, production technology and other factors and processes that coincide in time, contribute to the release of the maximum possible volume of gas. However, as we know, the probability of such a coincidence is almost impossible. In addition, the results of long-term observations of sudden methane emissions in coal mines at all deposits in the world indicate the following two key points.

First, it is the volumes of methane emissions registered by the practice of coal mining, which amount to more than 150–200 m^3/t (in some cases – up to 500 m^3/t). Taking into account this factor, the diagram of the maximum possible calculated ratio a priori values of the main phase components of methane in fossil coal will look like in Fig. 2. There is a very significant component of unknown origin methane.

Secondly, it is the duration of the gas-dynamic process, which is a maximum of 10 to 12 seconds. Based on this, as well as calculations indicating the duration of solid-state diffusion – from several hours to several days, the proportion of methane released from solid carbon-methane solution will be much less than shown in Fig. 1, 2. In Fig. 3 shows a diagram illustrating the rate of release of methane various components in the first minute of a sudden release of coal and gas.

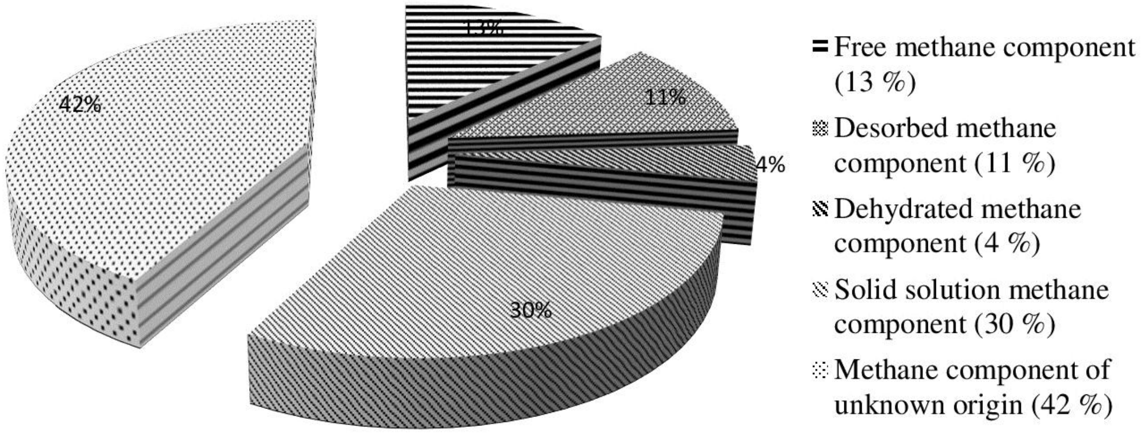

Figure 2 – Possible content of methane phase components in its total average volume (≈200 m^3/t) during GDP

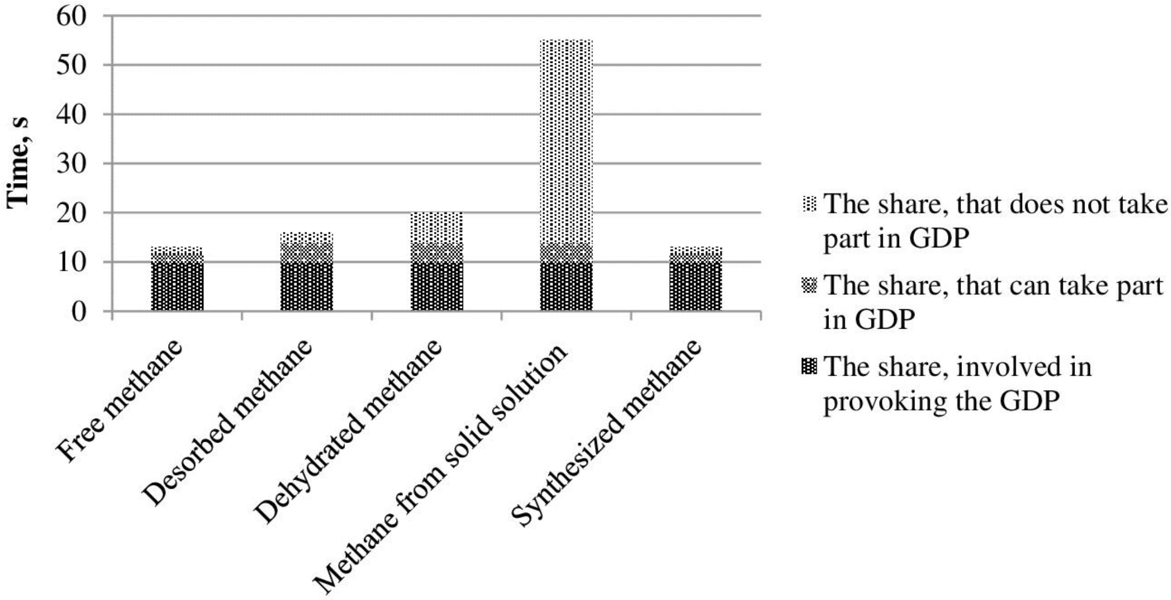

Figure 3 – Estimated fraction of methane release in the first 60 seconds after the GDP

As follows from the diagram, all the main phase components of methane are involved in the gas-dynamic process, but significantly different. This is especially true of the component released from the solid solution. In the first minute after of a sudden release, only 20 % of this component is involved. This calls into question the dominant role of the solid carbon-methane component in anomalous gas formation in the sudden release of coal and gas.

Thus we have a situation where a significant proportion is methane of unknown origin. From this we can conclude that a more accurate idea of the processes that take place in the system "coal – methane" can be obtained only by a deeper study of this system structure at the atomic and molecular level, taking into account thermodynamic, mechanic-tribe-electric, emission and other processes that occur both at the stages of metamorphism and man-made intervention in the coal seam.

Previous studies [16-20] indicate that the most likely disintegration of coal involves mechanic-chemical processes, which take a few seconds to synthesize methane in the aliphatic part of the coal substance. Especially in the presence of catalysts (various metals), which are sufficient in fossil coal. This methane increases the total volume of gas. Experiments on electric-hydrodynamic destruction of coal in a vacuum chamber confirmed the validity of this hypothesis model.

Before constructing a model of anomalous manifestations of methane in the destruction of coal and the sudden emission provoked by this, let us dwell on some known and established by us scientific positions.

1. The most variable component of coal is hydrogen.

2. At the molecular level, coal consists of nuclei (crystallites) and a side fringe, which is characteristic of coal aliphatic groups, connects the ordered part and plays the role of bridges connecting the primary elements.

3. Layers and crystallites are interconnected by hydrogen bonds, van der Waals forces and, sometimes, structural (covalent) bridges, such as essential oxygen and polymethylene groups.

4. The temperature range from 30 to 35 ^{0}C is critical for the specific heat of coal and is quite important for chemical processes. That is, there is a critical region of temperatures, below which not all degrees of freedom of thermal oscillating motion are carried out by carbon atoms and their corresponding radicals.

5. To break the C-C connection requires 60 to 70 kcal. Since the connection usually does not need to be broken, but simply to switch it, the activation energy for coal is about 35 kcal.

6. The mechanism of abnormal methane formation in coal seams is closely related to the sudden emissions mechanism, so it is the area of sudden emissions of coal to be considered as a "boiler" of methane generation.

7. External signs of emission (scattering of coal on micron-dispersed particles) indicate in favor of participation in the process of valence chemical bonds. Additional evidence may be the time of emission, which is characteristic of the induction period of rupture and rearrangement of chemical bonds.

8. Coal has many honeycomb nodes that contain only carbon atoms, each of which is associated with three neighboring carbon atoms. Such grains are interconnected by covalent and metallic electronic bonds. Among the graphite-like nodes, we can note CH-radicals that connect neighboring graphite nodes. These hydrocarbon groups are a fringe, at the expense of which radicals CH_2, CH, H are formed, giving methane and other derivatives. The process is a solid-phase homogeneous reaction.

9. More than 90 % of sudden emissions occur in areas of tectonic disturbances. With this in mind, the ratio of aromatic and aliphatic components in coal of different brands was determined by NMR (nuclear magnetic resonance), which indicates an increase of aliphatic in hazardous coal (Fig. 4).

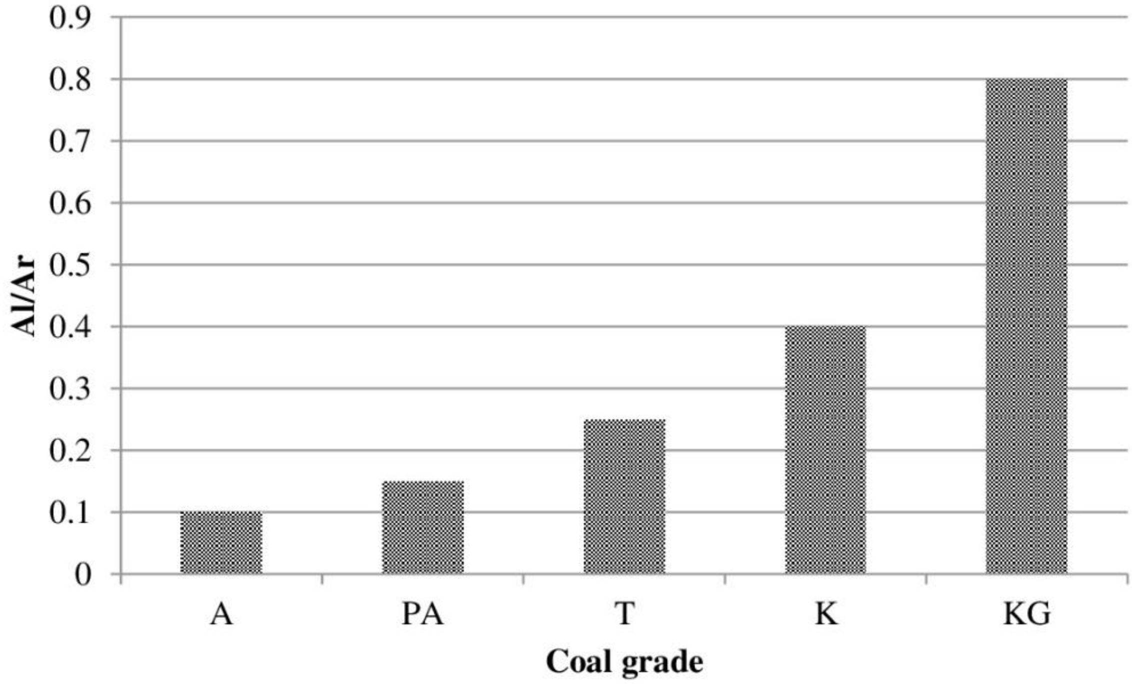

Figure 4 – The ratio of aliphatic and aromatic groups (Al/Ar) in coal of different grades (Ukrainian classification) according to NMR data

The very fact of increasing the content of aliphatic groups in coal indicates in favor of increasing the emission risk of the coal seam. The development of the process itself will be due to the influence of thermodynamic factors and the rate of flow. In this case, the denser the fringe, the less stable it is. As the resistance of graphite grows, the fringe disintegrates, and the more intense, the more concentrated the hydrogen areas.

10. At GDP coal substance is exposed to a shock wave of enormous force. To detect possible, in this case, chemical transformations in coal substance on coal samples and the method of electric-pulse shock modeled sudden emission. Then EPR spectroscopy (electron paramagnetic resonance spectroscopy) of the formed micro particles was performed. It is established that after the shock-wave destruction of coal matter in its EPR spectrum, in addition to a marked increase in the intensity of the main absorption line, an additional line of intensive absorption appears. The probability of this line is confirmed by a series of similar experiments conducted on coal grades DG, G and K. That is, in the process of shock dispersion of coal in its chemical formula there are significant changes, such as the formation of free radicals with high activation capacity.

11. It is known that chemical processes proceed much faster in the presence of catalysts - impurities of various metals. In Fig. 5 shows the study’s results of the paramagnetic centers concentration in coal of different brands. Coal was extracted in various mines. The diagram shows that the maximum number of paramagnetic centers is observed in coal, which is prone to sudden emissions. This is further evidence that one of GDP mechanisms is chemical processes in coal.

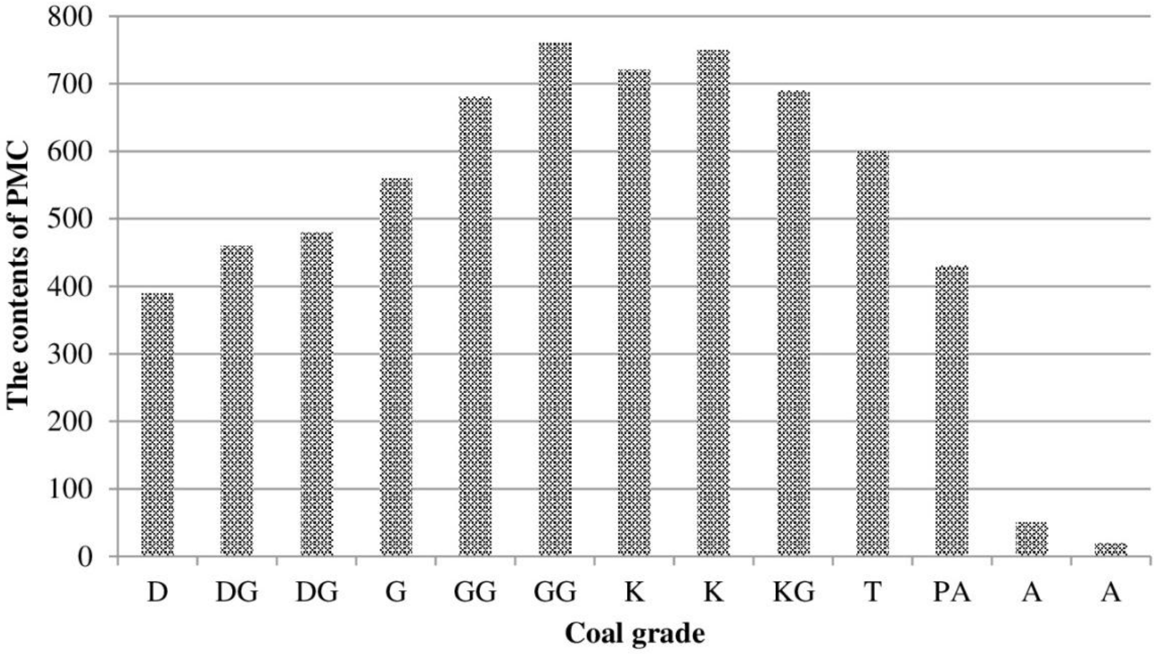

Figure 5 – The content of paramagnetic centers (PMC) in coal of different grades (Ukrainian classification), which is selected at different mines in Donbas

Based on the above provisions, the process of methane abnormal formation in the destruction of coal can occur as follows.

During the destruction of coal matter as a result of the consumption of hydrocarbon fringe, gaseous molecular methane is formed. In this case, the entropy increases rapidly and the free energy decreases. The process proceeds spontaneously, naturally, provided that the temperature is above 30 ^{0}C. The higher the temperature, the faster the fringe growth processes (solid-phase process) and its costs (heterogeneous process). The latter is accompanied by an increase in volume by 1700 times. In the absence of the output of the formed methane there is a local rapid increase in pressure. This is because the deeper the coal seam, the higher its temperature and the faster both processes. In addition, with increasing depth, methane removal conditions deteriorate. With difficult filtration process, sorption of generated methane leads to an additional increase in reservoir temperature. This has been repeatedly recorded in the working face. The formation of such a man-made focus of gas-dynamic danger is characterized by elevated temperatures and stress concentration zones in the field of generating additional amounts of methane from coal during mechanics-chemical processes.

Thus, at great depths fringe accumulates, which is prone to spontaneous consumption in the case of unloading part of the array. The term fringe consumption refers to the chemical generation of methane by the release of active excess hydrogen and its compounds with carbon atoms, that is the reaction of methane synthesis.

After the formation of unloaded rock mass section, another parallel fractal process begins to develop in the coal. Initially, the high gas pressure due to the accumulated potential energy of intermolecular repulsion of gas molecules breaks the interatomic bonds in the weakest link in the structure of the coal substance. A microcrack is formed, the germination of which is accompanied by mechanic-electric phenomena, the emission of electrons with energy above the activation energy, subsequent generation of methane and its additional desorption, increasing pressure, and so on. That is, the process becomes fractal.

At the stage of deformation of the coal seam, a tree-like hierarchical network of formed transport channels can be formed in it. In the process, transport channels grow in the surface gaps, which can be considered as fractal cracks. Under certain geomechanical conditions, a situation is possible when cracks will develop in the mode of "self-sustaining". That is, to grow only due to methane, which passes on the shores of cracks from the bound state to the free. The main conditions for this are sufficient unloading of the formation, high methane content of coal and low value of the specific energy of free surface formation. In practice, these conditions are confirmed by the dynamic destruction of coal in the event of coal and gas sudden emissions.

Calculations using the equation of methane mechanic-chemical generation, which has the form: $C_{58}H_{40}O_6N \rightarrow C_{56}H_{32}O_6N + 2CH_4$. From this, one ton of coal can generate more than 50 m^3 of methane. This will consume only a quarter of the fringe. That is, 0.9 % of unstable hydrogen in the fringe of coal is enough for the reaction in coal at an activation energy of 35 kcal to give a sufficiently powerful release of methane from the subsoil.

As for the speed of the process, it will depend on the total number of carbon atoms in the fringe, capable of generating methane. The effect of formation temperature at the appropriate depth is significant. At greater depths, where the temperature is higher, the power of the methane "boiler" chemical formation increases.

Conclusions. Based on the performed studies, a model of methane anomalous manifestations in the destruction of coal during GDP is proposed. The model takes into account physical, chemical and mechanic-chemical processes. First of all, the release of free methane, the mechanic-chemical reaction of methane synthesis (methane-generation), the desorption of physically bound methane and the amplification of this component by mechanic-electric processes. As well as the decomposition of solid carbon-methane solution and the release of the gas-hydrate component of the gas. The scheme of the model is shown in Fig. 6. The model assumes three phases of the gas-dynamic phenomenon.

The first phase (before the sudden release) includes the formation of the geological environment and the influence of technological factors. This phase is the main in assessing the susceptibility of the coal seam to GDP. The rating of natural and technological factors and processes by their contribution to the dynamic phenomena occurring in the mines are given in Table. 1. The validity of the first phase is unlimited and is determined by years and even millennia.

The second phase is sudden release the methane. It occurs as a result of the destruction of coal and includes all possible physical, chemical and mechanic-chemical processes. From the point of gas formation view and process speed it is, first of all, release of free methane, mechanic-chemical reaction of methane synthesis (methane-generation), desorption of physically bound methane and strengthening of this component by mechanic-electric processes, decomposition of solid carbon gas-hydrate solution. The duration of this phase is from 3-5 s to 10-15 s.

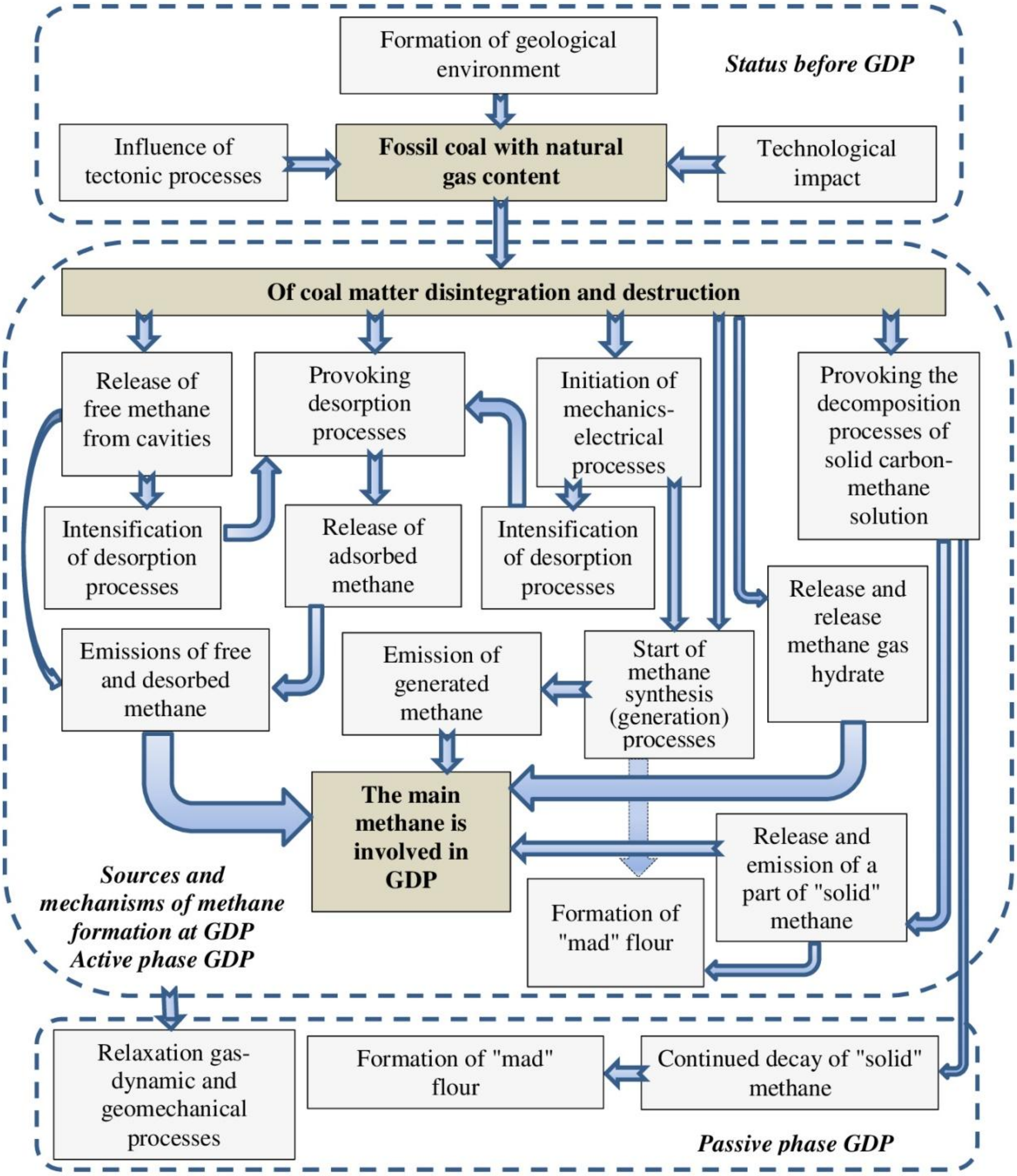

Figure 6 – Model of methane anomalous manifestations in coal destruction substances as a result of coal and gas sudden release

In the third phase, the process of solid carbon-methane solution decomposition continues for several hours and relaxation gas-dynamic and geomechanical processes take place.

Table 1 – Rating of factors and processes by their contribution to the GDP in mines

Rating no.	Gas-dynamic phenomenon in a coal mine
Rating of natural factors by their contribution to the formation of the dynamic phenomenon focus	
1	Concentration of natural methane in the array
2	The ability of natural methane components to release rapidly
3	Depth of mining (rock pressure)
4	The presence of tectonic stresses
5	Structural features of the rock mass
6	Humidity of the rock mass
7	The presence of catalysts for methane synthesis
8	The presence of mechanic-electric properties of rocks
Rating of man-caused factors in the mechanism of the dynamic phenomenon occurrence	
1	Speed of the working face advance
2	The degree of rock mass grinding
3	The shape of the tunnel face surface
4	The presence of sparking focus
5	Seismic action of an explosive wave
Rating of secondary physic-chemical processes that increase the scale of the dynamic phenomenon	
1	Occurrence of thermodynamic conditions for methane synthesis
2	Methane synthesis reaction
3	The presence of exposure areas of the working face surface – concentrators of mechanical stresses
4	Friction of crushed rock mass
5	Emission of charged particles

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About authors

Skipochka Serhii Ivanovych, Doctor of Technical Sciences (D.Sc.), Professor, Head of Laboratory of Physics and Geomechanical Monitoring of Rocks Mass, Institute of Geotechnical Mechanics named by N. Poljakov of National Academy of Sciences of Ukraine (IGTM NAS of Ukraine), Dnipro, Ukraine, skipochka@ukr.net

Krukovskyi Oleksandr Petrovych, Corresponding Member of the National Academy of Sciences of Ukraine, Doctor of Technical Sciences (D.Sc.), Deputy Director, Institute of Geotechnical Mechanics named by N. Poljakov of National Academy of Sciences of Ukraine (IGTM NAS of Ukraine), Dnipro, Ukraine, igtm@ukr.net

Palamarchuk Tetiana Andriivna, Doctor of Technical Sciences (D.Sc.), Leading Researcher in Laboratory of Physics and Geomechanical Monitoring of Rocks Mass, Institute of Geotechnical Mechanics named by N. Poljakov of National Academy of Sciences of Ukraine (IGTM NAS of Ukraine), Dnipro, Ukraine, tp208_2008@ukr.net

Prokhorets Liliia Victorivna, Candidate of Technical Sciences (Ph.D.), Senior Researcher in Laboratory of Physics and Geomechanical Monitoring of Rocks Mass, Institute of Geotechnical Mechanics named by N. Poljakov of National Academy of Sciences of Ukraine (IGTM NAS of Ukraine), Dnipro, Ukraine, prohoreclv@gmail.com

МЕХАНОХІМІЧНА МОДЕЛЬ МЕТАНОГЕНЕРАЦІЇ ПРИ ДЕСТРУКЦІЇ ВУГІЛЬНОЇ РЕЧОВИНИ

Скіпочка С.І., Круковський О.П., Паламарчук Т.А., Прохорець Л.В.

Анотація. Предмет досліджень, викладених в статті, це аномальні виділення метану з вугільних пластів під час їх розробки. Метою розробки є оцінка максимально можливих об'ємів основних фазових складових метану, що вміщує вугільній пласт, обгрунтування та розробка моделі генерації метану при деструкції вугільної речовини. В роботі використано аналіз та узагальнення результатів власних і сторонніх експериментельних та теоретичних досліджень, виконаних методами рентгеноструктурного аналізу, ядерного магнітного та електронного пара-магнітного резонансу, растрової мікроскопії, інфрачервоної спектроскопії, фізичного та математичного моделювання. Встановлено, що роль твердого вуглеметанового розчину, яка домінує в теорії газодинамічних явищ, і його частка в загальному об'ємі метану суттєво перебільшена. Основні невідповідності цієї теорії стосуються трьох положень. Невідповідності обсягів метану в твердому розчині об'ємам, що зареєстровані під час газодинамічного явища. Неспівставності швидкостей дифузійно-фільтраційних процесів при розпаді твердого розчину та протікання газодинамічного явища. Невідповідності черговості процесів викиду і утворення скаженої муки. Розроблена модель аномальних проявів метану при деструкції вугільної речовини, як один з основних, враховує процес генерації метану. Модель передбачає три фази газодинамічного явища. Довготривалу, що включає формування геологічного середовища і вплив технологічних факторів. Основну – тривалістю 5-15 с, що відбувається в результаті деструкції вугільної речовини і включає всі можливі фізичні, хімічні і механохімічні процеси, насамперед, вивільнення вільного метану, механохімічна реакція синтезу метану (метаногенерація), десорбція фізично зв'язаного метану та підсилення цієї компоненти механоелектричними процесами, розпад твердого вуглеметанового розчину та вивільнення газогідратної складової газу. Фазу згасання процесу протягом декількох годин, під час якої продовжується процес розпаду твердого вуглеметанового розчину та відбуваються релаксаційні газодинамічні і геомеханічні процеси. Робота спрямована на підвищення безпеки праці шахтарів за рахунок уточнення теорії газодинамічних явищ та вдосконаленні на цій основі методів та засобів їх запобігання.

Ключові слова: вугільна шахта, газодинамічні явища, теорії явищ, механо-хімічні процеси, метаногенерація, модель.

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