UDC 550.3:622.02:552.57:552.08 DOI: https://doi.org/10.15407/geotm2024.170.156

GEOMECHANICS AS A FACTOR OF CARBONIFICATION TRANSFORMATIONS AT VARIOUS HIERARCHICAL AND SCALE LEVELS

Bulat A., Krukovskyi O., Bulich Yu., Bezruchko K., Burchak O.

M.S. Poliakov Institute of Geotechnical Mechanics of the National Academy of Sciences of Ukraine

Abstract. The purpose of the initiated research for studying geomechanical factor of carbonization transformations is to assess the energy state of coal substance for further conscious control of structural transformations occurred in the atomic-molecular structure of coal due to the manifestations of the accumulated elastic energy. Elastic properties of coal were determined by the static method based on compressive resistance. The level of energy that can be accumulated in the molecular structure of coal under laboratory conditions was assessed at the stage of unloading the sample with arbitrary adjustment of its parameters by moving the meter after preliminary loading. The primary analysis of the results suggests that at the first stage, both plastic and elastic deformations occur simultaneously in heterogeneous, multicomponent, amorphous coal substance. At the second stage (reloading), only elastic deformations are observed. The energy of structural stresses at the molecular level is a parameter critically necessary for understanding the nature of carbonization transformations. Thus, it is for the first time when the possibility to assess experimentally the elastic energy, which can be accumulated in the molecular structure of a heterogeneous, multicomponent, amorphous substance of coal under force load, is proven. Taking into account the fact that the rate of free radical transformations in metastable carbonized organic substance is determined by the processes of relaxation of elastic energy accumulated in the substance at the atomic-molecular level, the understanding of the ability of the molecular structure of coal substance for the stress accumulation and manifestations makes it possible to adequately assess the properties of coal in order to develop the advanced technologies for the use of solid hydrocarbons (destruction to a gaseous state and, subsequently, nanosynthesis). The obtained results form the basis for further research involving measurements with different grades of coal and different particle classes, state and properties. Understanding the changes in the state of coal substance under the influence of the geomechanical factor will make it possible to establish the interdependence of elastic properties and gas generation capacity of carbonized organic substance.

Keywords: geomechanics, coal substance, structural stresses, energy.

1. Introduction

Geomechanics is the science of mechanical state of the earth's crust and the processes, which occur in it as a result of the action of natural physical factors, including mechanical and thermal factors. It explains and predicts changes in the stress-strain state of sections of the earth’s crust, provides conditions for studying natural phenomena in the material world at different hierarchical and scale levels, which contributes to the comprehensive solution of the most important problems in the fields of the following natural and technical sciences defined by the National Academy of Sciences of Ukraine: "Earth Sciences", "Mechanics", "Physical and Technical Problems of Energy" "Physics and Astronomy" [1]. The comprehensive approach of geomechanics to multi-level and multi-disciplinary scientific problems is based on the fundamental laws of nature, which ensures the connection between different research directions concerning the structure and existence of the material world.

Under current conditions, hydrocarbon formations remain a powerful potential source of natural resources. Therefore, the study of geomechanical processes caused by natural factors and technological actions at the macro level and the study of the influence of force loads on the atomic-molecular structure, state and properties of rocks, as well as on processes occurred at all hierarchical and scale levels of the natural geological environment are highly relevant.

In Ukraine, the leading position in the study of geomechanical processes in the geological environment is occupied by the Institute of Geotechnical Mechanics of the National Academy of Sciences of Ukraine (IGTM of the NASU). In recent years, a number of promising scientific developments were made by the IGTM in this area. In particular, for the macro level, a hypothesis was proposed regarding the formation of a secondary fractured-porous structure in rocks in exploited gas fields as a result of a gas pressure reduction and the development of compression deformations in the productive reservoir, which cause tensile deformations, contributing to the water and gas redistribution and forming a technogenic reservoir in it [2]. A mathematical model was developed to study the processes occurred during gas field development, and numerical simulation of changes in geomechanical and filtration parameters of the test model was performed [3].

Within the framework of solving the problem of searching for and forecasting coalbed methane deposits, the parameters of the properties of the coalbed massif that contribute to the release, migration, accumulation and preservation of gas accumulations were substantiated - the ranges of changes in the filtration and capacitive properties of coal-bearing deposits that characterize fractured rock-reservoirs, the boundary between the reservoir and the screen, and screens (covers) were determined. A new model of the formation of free methane accumulations was developed using the fundamental principles of the theory of fractured-porous media under the influence of a complex of geological and geomechanical factors on the state of the gas-saturated massif [4].

At the microlevel, coal-petrographic methods revealed the presence of traces of gas generation in the coalbeds across the entire carbonization series of Donbas. Their quantity, shape and size were analyzed, and it was concluded that solid coal substance turns into a gas-liquid substance (fluid) under external geomechanical influence [5]. It was experimentally proven by electron diffraction methods that under the influence of weak heat flows, structural stresses arise in coal, which are associated with the heterogeneity and defects of the structure of the substance. In the conditions of a rock massif, such stresses are formed during sudden outbursts and are accompanied by the release of fluids and formation of a fractured-porous structure in coal [6].

Based on the results of studies of processes in the coal massif, the Institute of Geotechnical Mechanics of the National Academy of Sciences of Ukraine developed a tectono-geochemical hypothesis of ongoing gas generation in real geological conditions, which is in full accordance with the basic laws of nature, is based on the principles of thermodynamics, does not contradict known empirical laws and is confirmed by practical experience [7, 8]. To support the proposed hypothesis, the Institute of Geotechnical Mechanics of the National Academy of Sciences of Ukraine experimentally proved that the transformations, which occurred in organic substance during the carbonization process, are identical to those which are activated in the layers under external influence in our time [9, 10]. It was also proven that the properties of carbonized organic substance, both concentrated and dispersed in any host rocks, are the same [11]. Accordingly, purely natural processes leading to methane generation can be activated in carbonized organic substance, regardless of its form, providing the possibility to control such processes and create the advanced technologies for coal mining and processing.

Based on the analysis of the results of theoretical and experimental studies, a physicochemical model of the formation of gas content in the coal-bearing massif was also developed [8]. The essence of this model is that under the action of geomechanical factors in coal, simultaneously with structural transformations at the molecular level with formation of free radicals, energy accumulation also occurs in the form of structural stresses. This additional energy is released by the system during the relaxation through the free radical mechanism resulting in the release of gaseous products of solid phase destruction, which, in turn, affect the state of substance and, accordingly, the kinetics of transformations [12–14].

Carbonization transformations of fossil organic substance, which occur under the influence of external geomechanical factors, are the natural system reaction, which involves the redistribution and accumulation of energy in the molecular structure of coal followed by relaxation of structural stresses through mechanochemical free radical reactions. The peculiarities of the coal reaction to the action of geomechanical factors are determined by the behavior of the coal macrostructure, mechanical properties of the substance and its microcomponents. The deformation energy depends on the structure of the substance and interatomic interaction in the molecular structure, which, in turn, is formed in the process of physicochemical transformations. However, at this time, there are no adequate experimental studies of the mechanical properties of coal substance, particularly regarding the quantitative assessment of the elastic energy accumulated in the atomic-molecular structure of coal, due to which carbonization transformations occur.

For developing technologies, the main task today is the advancement of theoretical concepts and interpretation of experimental data on the mechanisms of processes in substance and their quantitative assessment for conscious control of structural transformations at the nanoscale level [15]. The properties and state of substance at the atomic-molecular level are studied by molecular mechanics, which is based on the geometry (conformation and torsion angles) and energy characteristics of molecular particles using empirical potential functions, the form of which is taken from classical mechanics with accounting the Van der Waals forces and electrostatic interaction.

Coal is a copolymer, a heterogeneous, multicomponent substance that exists in a metastable state [12, 16, 17]. At the same time, coal represents a single genetic series, that is, buried remains of vegetation, which are almost identical in their primary state throughout the coal seam, and now are at different stages of carbonization. The degree of these transformations depends on the geomechanical conditions under which the buried organic substance was preserved over geological time. Transformations occur under the condition that the carbon atoms of the coal substance have the ability to switch bonds in the macromolecule. The formation of a particular structure is determined by the degree of excitation and hybridization of carbon atoms, while the speed and degree of structural transformations depend on the geomechanical conditions [18]. The process of carbonization has two manifestations: the removal of the coal substance of heteroatoms and hydrogen in the form of stable low-molecular compounds, and the structuring of the carbon-enriched residue. Both manifestations are entropically favorable and lead to a decrease in the energy of the system - that is, they are real in the conditions of a coal-bearing massif [8, 10].

Such processes are possible only if carbonization transformations occur by relaxation, spontaneously, due to the energy accumulated in the substance itself, in the molecular structure in the form of various types of stresses. The hierarchy of transformations is determined by the law of minimum energy, according to which those reactions take place that have the lowest energy barrier. In solid carbonized organic substance, these are mechanochemical free radical reactions, or, in other words, reactions with an active complex. The rate of such reactions is proportional to the number of active complexes (free radicals) and depends on the energy accumulated in the molecular structure in the form of stresses [19]. By assessing the energy accumulated in the substance as a form of structural stresses, it is possible to calculate the energy of the external influence, which will activate natural relaxation and free radical processes.

The purpose of the initiated work on the study of the geomechanical factor of carbonization transformations is to assess the energy state of coal substance for further conscious control of structural transformations that occur due to the manifestations of the accumulated elastic energy in the atomic-molecular structure of coal.

To achieve this goal, it is necessary to make a model of natural conditions – comprehensive compression - during the experiment. During the research, the ability of heterogeneous, multicomponent and amorphous coal substance to accumulate elastic energy at the molecular level was tested. At the same time, the possibility to assess experimentally the accumulated elastic energy was examined.

Under mechanical pressure, coal, the basis of which is high-molecular formations, undergoes both plastic and elastic deformations. Plastic deformation involves the displacement of elements of the supramolecular structure relative to each other with a change and rupture of intermolecular bonds and the destruction of the macrostructure of the solid.

Instead, when the system operates in the elastic region, deformation of macromolecules occurs causing changes in interatomic distances and torsion angles with the formation of conformational defects, due to which energy accumulates in the metastable coal substance in the form of structural stresses [14, 20, 21].

2. Methods

The coal elastic properties can be determined by the static method based on the compressive resistance. The level of energy that can be accumulated in the molecular structure of coal under laboratory conditions was assessed at the stage of unloading the sample and arbitrary adjustment of its parameters by moving the meter after preliminary loading. The experiments were conducted using a manual fifteen-ton press of the Specac company (Great Britain) using coal of the "G" grade from Yuvileyna mine. The grain size class was 1.0–0.2 mm and < 0.05 mm. The amount of coal powder was determined by the volume of a special cuvette, the weights of samples were recorded after the experiment. All studies were conducted at constant temperature and humidity.

In natural conditions, the molecular structure of coal is very dense, that is, the spatial structure of one chain is well aligned with the features of the structure of neighboring chains. The intermolecular space, in this case, is minimized. In order to eliminate the gaps between the coal powder particles, a force of 10.0 kN was initially applied to the sample in the press mold until the force stabilized at a new, lower level. This state of the sample was taken as “zero”, at which the space between the coal particles was minimized.

At the first stage, the data from the meters and manometer were recorded from the moment when the force reached value of 100.0 kN. Under conditions of volumetric compression with a similar force, the axial stresses are $735 \cdot 10^6$ Pa, which is equivalent to the pressure at a depth of ≈ 29.4 km. This significantly exceeds the values characteristic for the existing coal mining depths, but is in good agreement with the stress values that arise in coal seams during mining operations. The use of a compressive force comparable to those experienced during mining operations brings us closer to understanding the conditions for activation of a sudden coal and gas outbursts, which is an extreme manifestation of the carbonization process [9, 11].

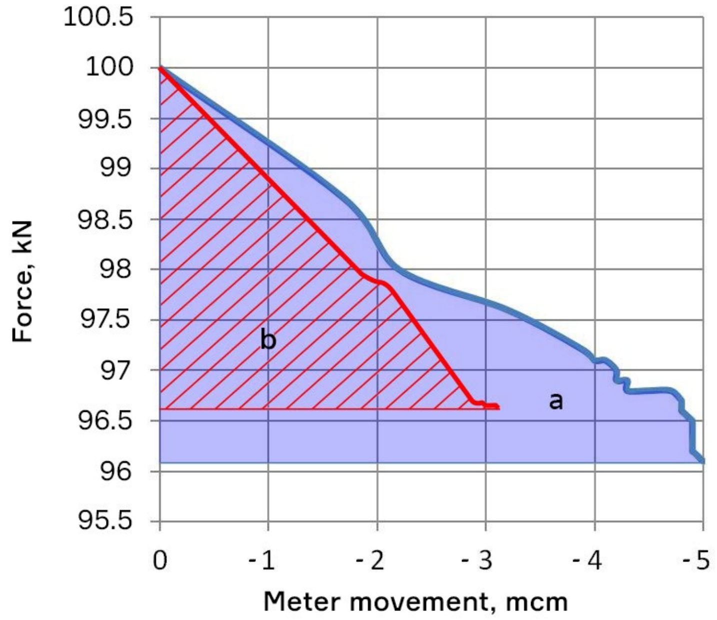
After stabilization of the process (600 s), the force was reset to zero, the system was not dismantled, and the sample was not removed. Almost immediately after reducing the force and restarting the recording system, the force (100.0 kN) was applied again (the beginning of the second stage). The second stage (repeated measurements of displacements and forces) also lasted 600 s.

3. Results and discussion

Figure 1 shows the experimental curves of structural stresses of the coal substance during two stages within one experiment. Similar trends were observed for all samples used in the research. The preliminary analysis of the results suggests that during the first stage both plastic and elastic deformations occur simultaneously in the heterogeneous, multicomponent, amorphous coal substance. At the second stage (reloading) only elastic deformations are observed - a parameter critically necessary for understanding the nature of carbonization transformations [14]. It should be noted that when performing the calculations, the influence of friction of the sample against the walls of the press mold was neglected.

It should be noted that diameter of the resulting pellet always exceeds the diameter of the press mold hole, which convincingly proves the existence of elastic deformations in the atomic-molecular structure of the coal substance. Analysis of the data given in Table 1 also confirmed that at the second stage (reloading of the sample) the substance absorbs approximately one-third of the energy that was absorbed at the first stage. Accordingly, it can be stated that experimental confirmation is obtained for the ability of the coal substance to accumulate elastic energy in the molecular structure of coal.

Coal is a copolymer or geopolymer, whose macromolecule mass can reach 10^5–10^7 Da. Based on the calculation of the average statistical structural unit per 10,000 carbon atoms, a mole of coal is 148–14,800 kg. Given that the activation energy of free radical reactions is 5.0–25 kJ/mol, and the average value of the specific elastic energy for three samples (Table 1) is 0.022 J/g, a mole of coal can accumulate from 3.256 kJ to 325.6 kJ, which is sufficient to activate free radical reactions in fossil organic substance. When processing the data, the accumulated energy was calculated by the area of the triangle, not of the trapezoid, and even with this approach, the level of energy released is sufficient to trigger structural transformations in the coal substance similar to carbonization.



a) first stage – recording of work in elastic and plastic region,
b) second stage – recording of work in the elastic region

Figure 1 – Experimental curves of structural stresses of coal substance

Table 1 – Parameters of coal samples based on the assessment of structural stress

Parameter	Sample No 1	Sample No 2	Sample No 3
Grain size class, mm	1.0–0.2	1.0–0.2	<0.05
Sample weight, g	0.175	0.188	0.229
Pellet diameter after leaving the press mold, mm	13.22	13.23	13.08
Pellet height, mm	1.23	1.28	1.56
First stage work, J	0.011	0.008	0.01
First stage. Specific deformation energy, J/g	0.060	0.043	0.063
Second stage. Specific elastic energy, J/g	0.017	0.018	0.031

The assessment of the energy, which can be accumulated in the molecular structure of coal, will provide further scientific justification for the technology of artificial control over the properties of the coal-bearing massif, taking into account geological indicators and the stress-strain state of rocks and structural transformations under external mechanical influence, and will also give impetus to promote the degassing of mining operations, environmental protection, energy resource extraction, and further integrated processing of natural hydrocarbons with the extraction of trace elements (liquid and rare-earth elements).

The results obtained are the basis for further research including measurements with various grades of coal and different particle classes, state and properties. Understanding the change in the state of coal substance under the influence of geomechanical factors will make it possible to establish the interdependence between the elastic properties and gas-generation capacity of carbonized organic substance.

4. Conclusions

The relationship between the manifestations of geomechanical processes at different scale levels was studied. This approach is based on the fundamental laws of nature (thermodynamics), provides a connection between different scale levels of the study of the state of the coal-bearing massif and substantiates the hierarchy of processes, which occur in it due to the action of natural physical factors (thermal, mechanical), i.e. geomechanical processes occurred at the macro level form processes at the micro level and vice versa.

It is for the first time when the possibility to assess experimentally the elastic energy, which can be accumulated in the molecular structure of a heterogeneous, multicomponent, amorphous substance of coal under force load is proven. Taking into account the fact that the rate of free radical transformations in metastable carbonized organic substance is determined by the processes of relaxation of elastic energy accumulated in the substance at the atomic-molecular level, understanding the ability of the molecular structure of carbonaceous substance for the stress accumulation and manifestations allows for an adequate assessment of the coal properties in order to develop the advanced technologies for the use of solid hydrocarbons (destruction to the gaseous state and, subsequently, nanosynthesis).

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About the authors

Bulat Anatolii, Academician of the National Academy of Science of Ukraine, Doctor of Technical Sciences (D. Sc), Professor, Director of the Institute, M.S. Poliakov Institute of Geotechnical Mechanics of the National Academy of Sciences of Ukraine (IGTM of the NAS of Ukraine), Dnipro, Ukraine, igtmnanu@ukr.net ORCID ***0000-0002-6541-2140***

Krukovskyi Oleksandr, Corresponding Member of NAS of Ukraine, Doctor of Technical Sciences (D.Sc), Deputy Director of the Institute, M.S. Poliakov Institute of Geotechnical Mechanics of the National Academy of Sciences of Ukraine (IGTM of the NAS of Ukraine), Dnipro, Ukraine, igtm@ukr.net ORCID ***0000-0002-2659-5095***

Bulich Yurii, Master of Science, Researcher in Department of Rock Mechanics, M.S. Poliakov Institute of Geotechnical Mechanics of the National Academy of Sciences of Ukraine (IGTM of the NAS of Ukraine), Dnipro, Ukraine, bulicurij@gmail.com ORCID ***0000-0003-3442-9203***

Bezruchko Kostiantyn*,* Doctor of Geology Science (D.Sc), Senior Researcher, Head of Department of Geology of Coal Beds at Great Depths, M.S. Poliakov Institute of Geotechnical Mechanics of National Academy of Sciences of Ukraine (IGTM NAS of Ukraine), Dnipro, Ukraine, gvrvg@meta.ua ORCID ***0000-0002-3818-5624***

Burchak Oleksandr, Doctor of Technical Science (D.Sc), Senior Researcher, Senior Research in Department of Geology of Coal Beds at Great Depths, M.S. Poliakov Institute of Geotechnical Mechanics of National Academy of Sciences of Ukraine (IGTM NAS of Ukraine), Head of Department of Geology of Coal Beds at Great Depths, Dnipro, Ukraine, gvrvg@meta.ua ORCID ***0000-0001-9114-8585***

ГЕОМЕХАНІКА ЯК ЧИННИК ВУГЛЕФІКАЦІЙНИХ ПЕРЕТВОРЕНЬ НА РІЗНИХ ІЄРАРХІЧНИХ І МАСШТАБНИХ РІВНЯХ

Булат А., Круковський О., Буліч Ю., Безручко К., Бурчак О.

Анотація. Мета започаткованих робіт з дослідження геомеханічного чинника вуглефікаційних перетворень – оцінка енергетичного стану вугільної речовини для подальшого свідомого керування структурними трансформаціями, що відбуваються за рахунок реалізації накопиченої пружної енергії в атомно-молекулярній структурі вугілля. Пружні властивості вугілля визначалися статичним методом за опором стиску. Рівень енергії, що може бути накопичена в лабораторних умовах в молекулярній структурі вугілля оцінювався на етапі розвантаження зразка та самодовільного встановлення його параметрів за переміщенням вимірювача після попереднього навантаження. Первинний аналіз результатів дозволяє стверджувати, що на першому етапі в гетерогенній, полікомпонентній, аморфній вугільній речовині одночасно відбуваються як пластичні, так і пружні деформації. На другому етапі (повторне навантаження) спостерігаються лише пружні деформації. Енергія структурних напружень на молекулярному рівні – параметр критично необхідний для розуміння природи вуглефікаційних перетворень. Таким чином, вперше доведена можливість експериментальної оцінки пружної енергії, що може бути накопичена в молекулярній структурі полікомпонентної, гетерогенної, аморфної речовини кам'яного вугілля під силовим навантаженням. З урахуванням того, що швидкість вільно-радикальних перетворень у метастабільній вуглефікованій органіці визначається процесами релаксації пружної енергії накопиченої в речовині на атомно-молекулярному рівні, розуміння здатності молекулярної структури вугільної речовини до накопичення та реалізації силових напружень, дозволяє адекватно оцінити властивості вугілля з метою розробки новітніх технологій використання твердих вуглеводнів (деструкції до газоподібного стану та, в подальшому, наносинтезу). Отримані результати є підґрунтям для розвитку досліджень з проведенням вимірів на різних марках вугілля, з різними класами частинок, станом та властивостями. Розуміння зміни стану вугільної речовини під впливом геомеханічного чинника уможливить встановлення взаємозалежності пружних властивостей та газогенераційної здатності вуглефікованої органіки.

Ключові слова: геомеханіка, вугільна речовина, структурні напруження, енергія.