

ANALYTICAL REVIEW OF ELECTRICAL PHENOMENA RESEARCH IN COALIFICATION IN VIEW OF THE SCALE HIERARCHY

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Abstract. Electrical processes in carboniferous rocks saturated with organic matter have to be considered at three scale levels: the macrolevel of the bed, the mesolevel of the sample, and the microlevel of the point. The electrical resistivity of coal is affected by rank, humidity, salinity, temperature, and other parameters. Temperature changes in the electrical conductivity of coals strongly depend on the thermal history of the bed and have a complex multi-stage nature as the sample heats up, due to multidirectional processes. Thermal activation of coals leads to an increase in the number of charge carriers in a local continuous microregion. At the same time, the process of hollow crack formation prevents the growth of electrical conductivity on a macroscale. During underground fires of coal beds, the electrical resistivity decreases significantly at the source of the fire, and an abnormally high value of electrical self-potential is recorded on the surface of the earth. The specific electrical conductivity of a moisture-saturated coal bed can increase significantly due to the ionic conductivity of pore electrolytic solutions. The dielectric properties of coals are highly dependent on rank, temperature, and humidity. The relative dielectric constant increases with increasing temperature. The ultra-high-frequency broadband spectrum has a complex shape with many local maxima. Dielectric losses tend to rise with increasing frequency and differ significantly for coals of different ranks. Coals subjected to uniaxial compression and shear loading can produce ultra-low-frequency electromagnetic radiation signals that correlate well with applied stress and acoustic emission. The physical nature of the electromagnetic emission of a coal bed, caused by the movement of charges on the surface of pores and cracks, can be explained by the large-scale effect of the microsurface self-potential of coal. Metamorphism can affect the surface potential of coals through the evolution of polar functional groups, distribution of surface microstructures, micromorphology, internal connectivity, and heterogeneity.

The purpose of the work is to review, analyze, and systematize published experimental research on the electrical properties of coals depending on the metastable structure of their carbon matrix and external factors at different scale levels.

Keywords: coal, scale hierarchy, electrical conductivity, dielectric constant, self-potential, electromagnetic emission.

1. Introduction

By now, a large number of studies have been published devoted to the electrical, magnetic, and electromagnetic processes in the carbonized matter of fossil fuels of organic origin. This group of fossil fuels includes such solid combustible materials as brown and hard coals, anthracites, and oil shale with varying proportions of ash components and the degree of chemical maturity of kerogen. They exhibit a wide variety of electrical properties, which are determined by the atomic-molecular and supramolecular structure of the matter and significantly depend on the impact of external factors.

It is necessary to distinguish between static electrical phenomena caused by slowly changing fields, as well as fast effects associated with high-frequency and microwave dynamics. In addition, both unexpected phenomena that arise spontaneously under the impact of external factors of a non-electromagnetic nature, and the reaction of a substance to electromagnetic effects are possible. The first includes the phenomena of electromagnetic emission, electric self-potential, superparamagnetism, ferromagnetism, piezomagnetism, the emergence of piezo-, pyro- and triboelectricity, etc. The second includes various types of relaxation of the substance to electromagnetic

radiation such as photoelectric effect, luminescence, absorption, reflection, re-emission of electromagnetic waves, etc.

Theoretically, in a pure substance with an ideal structure, the electrical parameters at the macro level should correspond to the number and mobility of charge carriers at the micro level. However, in real natural substrates, such large-scale invariance is often not observed. The translation of electrical properties can be interrupted by numerous macro- and microstructural defects of coal, foreign inclusions, developed crack-pore space, heterogeneity of the substance, etc. Therefore, electrical and magnetic processes in carboniferous rocks saturated with organic matter must be considered at three scale levels: the macro level of the bed, the mesolevel of the sample, and the micro level of the point (the atomic molecular structure of the matter).

At the same time, it is necessary to take into account the ambient temperature, since the temperature dependence of the electrical resistivity in almost all non-metals is due to the thermal activation of the electric charge. Charge carriers can be ions, ionic vacancies, electrons, or quasiparticles. Every microparticle is held in its place with a certain force and must receive enough energy to overcome the energy barrier and become a mobile charge carrier. Temperature rise increases the energy of particles, which causes more localized particles to overcome their energy barrier and become mobile.

2. Methods

Review, analysis, and systematization of published experimental research on the electrical properties of coals in terms of their dependence on the metastability of the carbon matrix structure and external factors at different scale levels.

3. Theoretical part

1. Electrical conductivity. On the one hand, a thermally activated increase in the number of charge carriers leads to a decrease in the electrical resistivity of coal and accompanying rocks symbatically with temperature. On the other hand, thermal fracturing prevents the growth of electrical conductivity. Various minerals of carboniferous rocks will demonstrate thermodynamic differences at high temperatures, such as anisotropy and heterogeneity under thermal expansion. Deformations in some directions cannot occur, as a result of which thermal stress appears in the structure of the matter. When the stress exceeds the strength limit of the rock, microcracks appear, they connect with each other and turn into macrocracks. The emergence of cracks leads to the growth of a fissure-porous structure, which increases the electrical resistivity of the rock.

At the low-temperature stage, up to 150 °C, the number of small cracks in minerals with an uneven distribution of components gradually increases. Meanwhile, the mineral expands because of high temperature and can change the shape and size of the primary pores in the rock. As a result, the fluid permeability of the matter changes slightly and even decreases. Thus, at this stage, an increase in the number of charge carriers is the dominant factor leading to a decrease in resistivity.

When the temperature rises to 200 °C (the threshold temperature for thermal destruction), many small cracks connect with each other. As a result, large cracks form, and the fluid permeability of the rock increases sharply. At this stage, the thermal rupture is strongest and the permeability changes most dramatically. Thermal fracture becomes the dominant factor leading to an increase in resistivity.

Between 300 °C and 400 °C, fluid permeability begins to decrease due to the activation of physicochemical processes in the molecular structure of the matter. At this stage, an increase in the number of charge carriers becomes the main factor, which again leads to a decrease in resistivity. After 400 °C, although the permeability begins to increase again, the increase in resistance caused by it can no longer compete with the process of decrease in resistance associated with an increase in charge carriers. Therefore, the resistivity of carboniferous rocks continues to decrease with temperature until the end of the heating process [1].

An increase in the permeability of carboniferous rocks with increasing temperature during metamorphism improves the ingress of underground salt water to the coal bed. At the same time, due to the dehydration process, a system of cleavage cracks is formed in coal, available for filling the pore space with aqueous electrolytes. These processes lead to an increase in the ionic electrical conductivity of the moisture-saturated coal bed.

Electrical conductivity is further influenced by the thermal history of the coal. During underground fires of coal beds, a significant decrease in resistivity in the combustion zone is observed due to the pyrolysis of coal at high temperatures. This, in turn, leads to a higher carbon content in the semicoke.

The electrical resistivity of coal increases with the transition from brown coal to hard one and reaches a maximum at the middle stage of coalification, and then decreases with an increasing degree of metamorphism. However, the electrical resistance of coal for a particular grade of coal cannot be determined accurately, since it depends on the maceral, chemical and mineral composition, as well as on humidity and temperature. Wet coal in its original state has a resistivity of approximately 100 to 500 Ohm·m measured at a frequency of 1.0 kHz. During pyrolysis in laboratory experiments at temperatures above ~ 650 °C, coal becomes a good conductor with a resistivity of ~ 1 Ohm·m. At temperatures > 800 °C, coal can have a conductivity 10⁵ times greater than that of the original water-saturated sample, with the parameter varying from 1000 Ohm·m to 0.01 Ohm·m [2].

Unlike carboniferous rocks, the electrical resistivity of bituminous coals practically does not change up to a moisture content of 0.6% by weight; further moisture results in a sharp decrease. The temperature dependence is nonlinear; in the temperature range of 0.0–80 °C, the electrical resistivity gradually decreases, and at temperatures of 80–180 °C, it increases sharply; further heating leads to a smooth decline. The electrical conductivity of coal also significantly depends on the presence of mineral matter. In this case, the resistivity is greater for mineral components than for coal matters [3].

Experimental research of the electrical resistivity of coals and rocks have shown that sediments, minerals and coals have different electrical resistivities (Fig. 1).

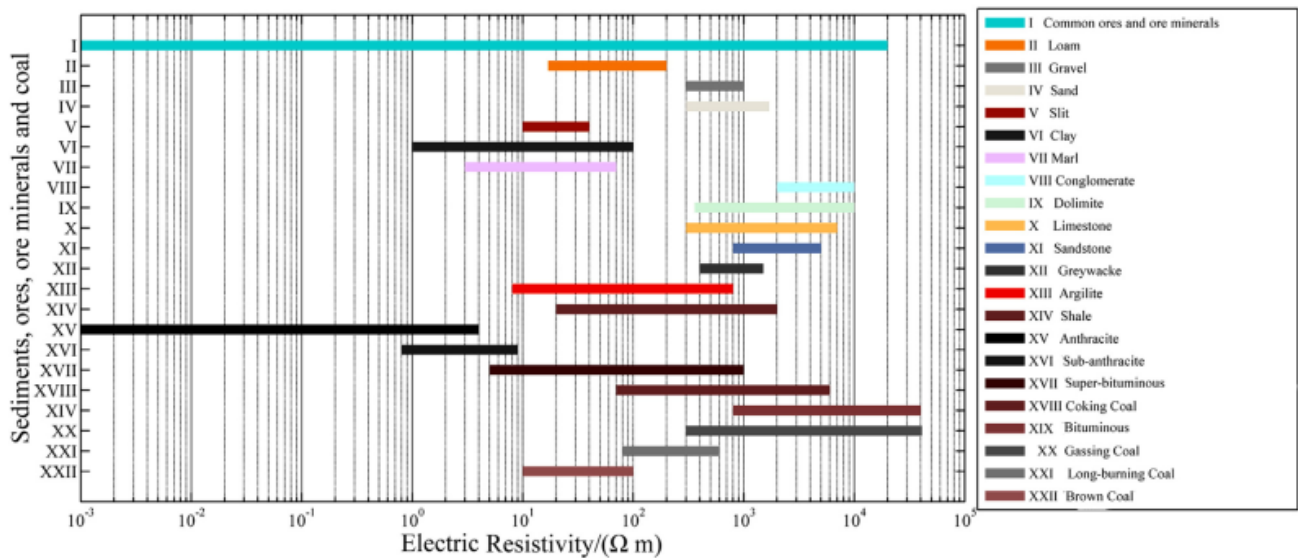


Figure 1 – Electrical resistivity ranges of coals, cemented and loose sediments, ores and ore minerals [4]

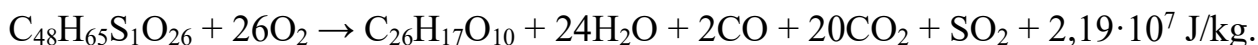
Thus, the electrical resistivity of coal is influenced by the grade of coal, the development of the cracked-porous structure, humidity, mineralization, temperature and other parameters of both the coal itself and the enclosing rocks. Electrical resistivity in the metamorphism series has two trends:

- a) growth with increasing quality of coal from brown to bituminous coal due to a decrease in moisture content and humic acid ions;
- b) decrease from bituminous coal to anthracite due to an increase in carbon content with delocalized electrons [4].

2. Self-potential. When coal beds are strongly heated on the surface of the earth, an abnormally high value of electrical self-potential is recorded. The geophysical self-potential method is used to detect coal fires. However, the origin of the thermally stimulated self-potential is still unclear. There are three known possible sources of self-potential anomalies associated with coal fires: thermoelectric potential, oxidation-reduction potential, and flow potential. The flow potential can be neglected since there is no pressure of water or other fluids in the coal fire area.

The temperature around the combustion source usually reaches a thousand degrees Celsius. However, due to the high heat capacity and low thermal conductivity of sedimentary rocks, heat transfer to the Earth's surface is difficult. Consequently, the temperature on the daytime surface is much lower than in the burning center. Charge carriers in rocks diffuse predominantly from a hot region to a relatively cold one. Thus, thermoelectric potential accumulates in a cold region near the Earth's surface, and this charge separation creates a potential difference. The corresponding current density is the input member in Maxwell's equations, which generate electric and magnetic field disturbances.

Coal combustion involves a violent chemical reaction between combustible components and an oxidizing agent, usually atmospheric oxygen, releasing heat, light and reaction products. Coal oxidizes to form H_2O , CO_2 , CO and other products:



During the exothermic chemical process, the coal loses electrons, which will migrate through the rocks to the surface, where they are recombined by a terminal electron acceptor. In this process, coal is the electron donor on the anode, and gaseous oxygen acts as the final electron acceptor on the cathode [5].

The self-potential measured at the surface increases with increasing temperature of the heat source and sharply decreases with increasing buriedness. In addition, the oxidation-reduction potential reduces the total values of self-potentials on the surface. Consequently, the self-potential values measured in the depth of the bed should be significantly higher than those recorded on the earth's surface. However, both described mechanisms do not fully explain the abnormally large self-potential.

3. Dielectric constant. Measurements of the dielectric constant of dry coals without mineral impurities in the high-frequency range of 0.6–2.2 GHz showed that it decreases with increasing grade of coal. The presence of moisture and minerals can significantly increase the dielectric constant of bulk coal. The real part of the relative dielectric constant of coal is higher than that of most mineral components, with the exception of pyrite. The dielectric properties of coals depend strongly on temperature and weakly on microwave frequency. A significant decrease in the dielectric constant of coal and the loss factor occurs between 80 °C and 180 °C and is associated with the removal of moisture, which is in good agreement with the behavior of electrical conductivity in this temperature range (see p. 1). Natural moisture coals of both high and low grades have the highest values of dielectric constant and loss factor. For sub-bituminous coals, high rates are due to the presence of water dipoles, which are strongly connected with the molecular structure [6].

More recent research measured the relative dielectric constant and dielectric loss of three coal samples (at $T = 0\text{--}80$ °C and $f = 500\text{--}1000$ MHz) by ultra-wideband dielectric spectrometry. The impact of ultra-wideband frequency and temperature on the dielectric properties of coals of different degrees of metamorphism is analyzed. The frequency spectrum of the relative dielectric constant of coals has a complex shape (Fig. 2).

The relative dielectric constant of coals first decreases with increasing frequency to a local minimum and then increases to values close to the initial ones, after which a significant decrease follows. Analysis of the given graphs shows that the dielectric constant of a medium is connected with its polarization. Each polarization mode has its own relaxation time. When the frequency of the electric field is high, slow polarization processes may not keep up with the change in the electric field, so the relative dielectric constant decreases with increasing frequency. The local maximum is associated with the electronic conductivity of certain coal structures. At low frequencies, the electrons in carbon are in a bound state with functional groups, and the microwave energy is not enough to excite them to overcome the barrier. As the frequency increases, the probability of electron delocalization grows.

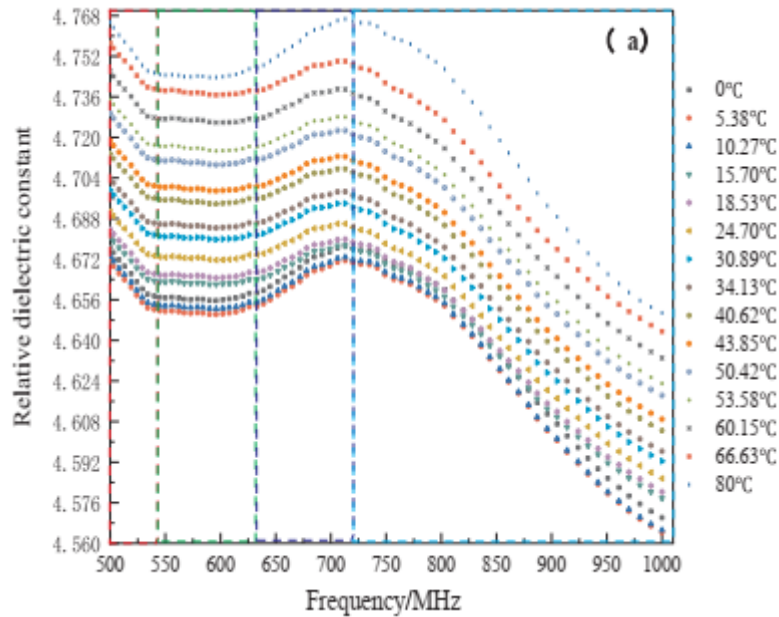


Figure 2 – Family of dielectric spectra of lean coal at different temperatures [7]

The relative dielectric constant increases with increasing temperature. Dielectric losses increase with increasing frequency and differ significantly for different coals. At the same frequency and temperature, the dielectric losses of lean coal are greater than those of long-flame coal. Dielectric losses of all coals increase at temperatures $> 60\text{ }^{\circ}\text{C}$ [7].

In coals and solid residues during pyrolysis, the density of charge carriers is low and there is practically no charge transport. Charge mobility inside a macromolecule can only occur if the macromolecule contains systems of conjugated links. The application of an external electric field leads to polarization of the conjugated system of double links in the macromolecule chain and the appearance of quasiparticles induced by external solitons, polarons, etc. These quasiparticles can be generated in coals and their carbonized residues during thermal degradation under the impact of an applied electric field. Which leads to a decrease in the electrical resistivity of solid residues for sintered coals under the impact of a larger electric field.

Various types of impurities are of no small importance in increasing the electrical conductivity of high-molecular dielectrics. In the case of hard coals, such impurities can be low-molecular destruction products and substances such as plasticizers located in the intermolecular space. The removal of these products from intermolecular nanopores at the moment of re-solidification of the plastic mass of sintered coals (at $T = 450\text{--}520\text{ }^{\circ}\text{C}$) or their insufficient amount during the restructuring of the supramolecular structure of non-sintered coals (at $T = 380\text{--}620\text{ }^{\circ}\text{C}$) leads to the emergence of a local maximum in the temperature curve of electrical resistivity.

The electrical resistivity of solid residues of both sintered and non-sintered coals is determined by the dielectric properties of the disordered phase. The increases in temperature of low-frequency dielectric constant ($f = 0.1\text{ MHz}$) for solid residues of non-sintered coals occur due to the loosening of their structure. The temperature

curve of the dielectric constant for solid residues of sintered coals has a characteristic minimum in the region of the plastic state, which is determined by electronic saturation. This phenomenon manifests itself in anomalies in the electrical and dielectric properties of molecules and anisotropy of optical properties [8].

4. Electromagnetic emissions. In practice, it has been observed that deformation and fracture of loaded rock can cause electromagnetic radiation (EMR). Marble, granite, clays, sandstones, and other hard rocks are capable of generating it. This phenomenon is explained by the piezoelectric effect, friction electrification, flow potential, migration of electrons, holes, and polarization caused by force stresses, mainly at the macro- and mesoscale. From this, it follows that with the help of EMR and acoustic emission, it is possible to control the stressed state of a coal bed [9].

When the mechanical stress increases, the outer electrons are excited, creating an instantaneous dipole moment, which emits electromagnetic waves. The mechanism of rock radiation is due to high-speed movement and collision of external electrons. Some types of rocks in the process of destruction can emit an electron; when the external force reaches a certain threshold, the number of electrons increases rapidly with a wide distribution of them across energy levels. The signal amplitude is greatest in the crack. The frequency of the electrical and magnetic signals is not synchronized, and the time and amplitude of the electrical signal is small.

Experiments show that coals subjected to uniaxial compression and shear loading can produce ultra-low frequency (300 Hz–3 kHz) EMR signals that correlate well with applied stress and acoustic emission. Ultralow-frequency signals after applying voltage are caused, firstly, by changes in the induction field due to the movement of charges, and secondly, by the piezomagnetic effect arising from the presence of certain iron-containing minerals such as pyrite in coals [10].

Previously, the Institute of Geotechnical Mechanics of the National Academy of Sciences of Ukraine showed that the surfaces of coal burstings can have a stable spontaneous potential at a constant temperature [11]. In another innovative research, the microsurface potential of coals of different thermal maturity ($R_o = 0.90\% \sim 2.47\%$) was measured using atomic-force microscopy at five scanning scales [12]. The electrical self-potential of the coal microsurface depends on the degree of metamorphism and has a significant scale effect. The coal microsurface potential is heterogeneous and has a Gaussian distribution. The spontaneous charge is mostly positive and can reach more than 800 mV (Fig. 3).

The average surface potential varies from -0.73 V to +0.87 V. The potential difference between different points, measured in a certain scanning area, can exceed 100 mV. The average potential decreases with increasing scan scale, while the dispersion increases (Fig. 4).

The scale effect of coal microsurface potential is mainly due to its heterogeneity. Metamorphism can affect the surface potential through the evolution of polar functional groups, distribution of surface microstructures, micromorphology, internal connectivity and heterogeneity. Accordingly, the existence of the microsurface potential of coal, especially its large-scale effect, provides a physical basis for EMR generation when charges move on the surface of pores and cracks.

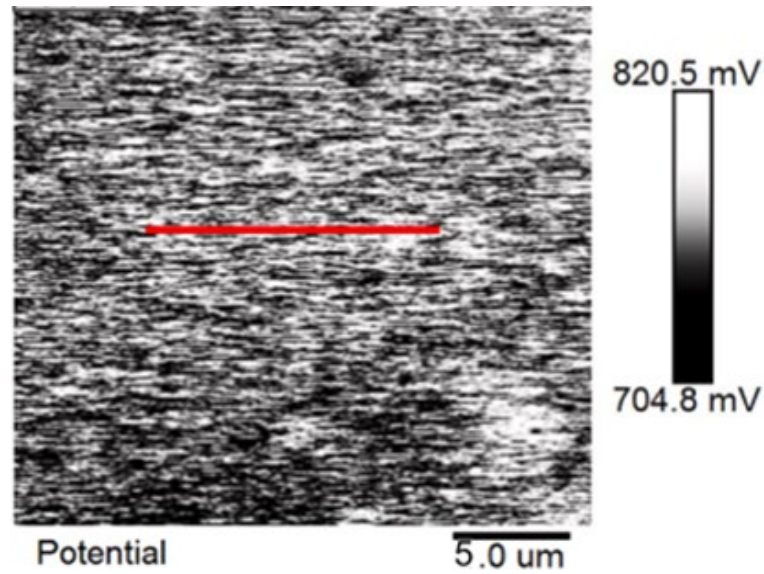


Figure 3 – The example of heterogeneity in the spatial distribution of microsurface self-potential of coal [12]

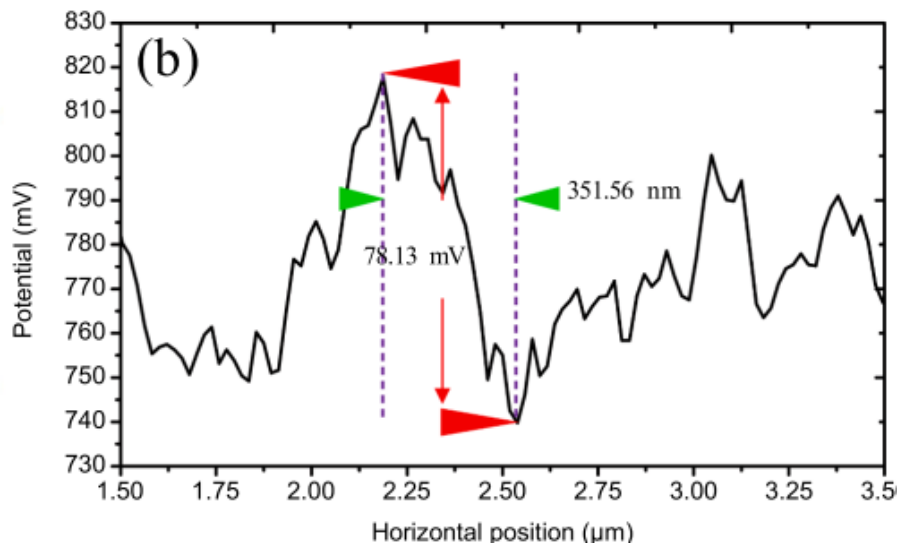


Figure 4 – The example of the impact of local potential inhomogeneity [12]

3. Results and its discussion

An analysis of published works has shown that most of the research is of an episodic applied nature, on the basis of which it is difficult to form a single holistic system of hierarchy for observed electrical phenomena with a cause-and-effect relationship between factors, processes, and properties for matters with a solid hydrocarbon matrix. One factor can generate many processes for a variety of carbon matrices at different scale levels, which accordingly exhibit different properties. In this case, it is necessary to look for analogies and rely on fundamental knowledge borrowed from more developed scientific fields, where the objects of research are pure substances with a known atomic-molecular, micro- and macro-structure.

4. Conclusions

The wide range of electrical properties of rocks saturated with organic matter is associated with the degree of maturity of kerogen and the degree of external factors that impact the energy state of coal, which in turn forms the electrical and magnetic properties of the matter. This gives rise to complex temperature dependencies and multidirectional processes from polarization of dielectrics to electronic or ionic conductivity. Their scale hierarchy strongly depends on the type and proportion of geopolymer, its electrical properties, as well as the filling of the pore space with liquid electrolytes.

Prospects for studying the electrical properties of coal are associated with modeling the corresponding processes in carbon structures with a known structure and properties.

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АНАЛІТИЧНИЙ ОГЛЯД ДОСЛІДЖЕНЬ ЕЛЕКТРИЧНИХ ЯВИЩ У ВУГЛЕФІКОВАНІЙ РЕЧОВИНІ З УРАХУВАННЯМ МАСШТАБНОЇ ІЄРАРХІЇ

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Анотація. Електричні процеси у вуглецевмісних породах насичених органічною речовиною необхідно розглядати на трьох масштабних рівнях: макрорівень пласта, мезорівень зразка, мікрорівень точки. На питомий електричний опір вугілля впливає ранг, вологість, мінералізація, температура та інші параметри. Температурні зміни електропровідності вугілля сильно залежать від термічної історії пласта і мають складний багатоетапний характер у міру нагрівання зразка зумовлені різноспрямованими процесами. Термічна активація вугілля призводить до збільшення кількості носіїв заряду в локальній безперервній мікрообласті. У той же час процес утворення порожнистих тріщин перешкоджає зростанню електропровідності в макромасштабі. При підземних пожежах вугільних пластів питомий електричний опір значно зменшується в осередку займання, але в поверхні землі реєструється аномально високе значення електричного самопотенціала. Питома електропровідність вологонасиченого вугільного пласта може значно зростати за рахунок іонної провідності порових розчинів електролітів. Діелектричні властивості вугілля сильно залежать від рангу, температури та вологості. Відносна діелектрична проникність збільшується з підвищенням температури. Надвисокочастотний широкосмуговий спектр має складну форму з безліччю локальних максимумів. Діелектричні втрати мають тенденцію зростати зі збільшенням частоти і значно відрізняються для вугілля різних рангів. Вугілля, піддане одновісному стисненню та зсувному навантаженню, можуть давати ультранизькочастотні сигнали електромагнітного випромінювання, які добре корелюють з прикладеною напругою, і акустичною емісією. Фізичну природу електромагнітної емісії вугільного пласта, обумовлену рухом зарядів на поверхні пор і тріщин, можна пояснити масштабним ефектом мікроповерхневого самопотенціалу вугілля. Метаморфізм може впливати на поверхневий потенціал вугілля за рахунок еволюції полярних функціональних груп, розподілу поверхневих мікроструктур, мікроморфології, внутрішньої зв'язності та гетерогенності.

Мета роботи – провести огляд, аналіз та систематизацію опублікованих експериментальних досліджень електричних властивостей вугілля залежно від метастабільної структури їх вуглецевої матриці та зовнішніх факторів на різних масштабних рівнях.

Ключові слова: вугілля, масштабна ієрархія, електропровідність, діелектрична проникність, власний потенціал, електромагнітна емісія.