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SECONDARY MACERALS OF COALS AS AN INDICATOR OF TRANSFORMATION OF GELIFIED MATTER

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Abstract. Coal is a valuable fossil raw material for the power, metallurgy, and chemical industries. The intended usage of a particular coal is determined by several reasons, including its petrographic composition. The petrographic composition of coals as a set of macerals is formed in diagenesis and is unchangeable according to normative documents. In this paper, based on the long-term practice of the petrographic study of coals, we show the macerals referred to a certain group by their petrographic features (shape, color, brightness, relief). Still, by genesis, they have nothing to do with this group. They are the product of the gelified matter transformation, i.e. "primary" macerals of the vitrinite group, thus being "secondary macerals". In such cases, errors may occur in evaluating the properties of coals as raw materials for a particular industry.

The paper aims to give examples for the identification of "secondary macerals" of coals depending on their genesis and to give recommendations for their consideration. Genetic prerequisites for the occurrence of "secondary macerals" are described. Basically, it is a transformed gelified substance as a result of stress thermal impact by gas fluid, solidification of liquid phase of fluid on vitrinite of interlayer surfaces, dynamometamorphism in fractured zones, bacteriological processing. The authors carried out research with the help of video-optical complex: MBI-11, NV200, software Scope photo. The genesis of "secondary" macerals can be varied: stress thermal transformation of gelified substance by gas fluid, solidification of liquid phase of fluid on vitrinite of interlayer surfaces, dynamic metamorphism in fractured zones, bacteriological processing of the gelified matter, chemical reactions between liptinite and vitrinite during regional metamorphism. It is recommended, when diagnosing maceral as a "secondary" one, to exclude it from the calculation of petrographic composition in order not to make a mistake in estimating the properties of coal, for the reason that their properties are not studied at this stage. Since the counting is carried out according to the principle of "chessboard" the excluding the fact of meeting of such maceral will not significantly affect the result.

Keywords: coal, petrographic composition, optical microscopy, macerals, gelified matter.

1. Introduction

Despite the global shift towards renewable energy sources (renewable energy is energy obtained from natural sources that are replenished at a rate greater than the rate at which it is consumed. Examples of such constantly replenished sources are sunlight and wind. In contrast, fossil fuels – coal, oil, and gas – are non-renewable resources that take hundreds of millions of years to form. When fossil fuels are burned to produce energy, harmful greenhouse gases such as carbon dioxide are emitted), and the world still remains dependent on fossil fuels. Moreover, in 2023, global primary energy consumption reached historical highs – about 81% of the world's energy balance comes from fossil fuels, while renewable energy sources account for only 15%. Although this is the highest figure in human history, it certainly does not allow us to completely abandon primary energy. Oil, coal, and gas remain the most popular ways of obtaining energy in the world. Coal is considered the cheapest and dirtiest fossil fuel. In 2023, humanity used 1.5% more fossil fuels than in 2022. The reason for the new records was a surge in energy demand, more than half of which comes from the Global South, where demand is growing twice as fast as the global rate. India consumes more coal than all the countries in Europe and North America taken together.

In addition to energy, coal is intensively used in metallurgy and the chemical industry.

Where and what coals to use is determined by the properties of these coals. The properties of coals from a petrological viewpoint depend on three factors: metamorphism, recoverability, and petrographic composition of coals [1].

As the outstanding Ukrainian scientist Uziyuk V.I. notes: “The philosophical doctrine that the composition of a substance determines its quality is unquestionable” [2].

The transformation of dead plants into coal occurs as a result of a continuous process, in which it is common to distinguish two main phases: 1) humification - the transformation of dead plants into peat and 2) coalification - the transformation of peat successively into brown, hard coal and anthracite. Coalification is divided, in turn, into two parts: 1) diagenesis of coal, during which, under the influence of predominantly biochemical transformations due to the vital activity of microorganisms, peat is transformed into brown coal, and 2) metamorphism, during which brown coal, under the influence of physical factors - increased temperature and pressure of rocks - is transformed into hard coal and anthracite.

The petrographic composition of coal as a set of macerals is formed once during humification and the initial stages of diagenesis and then does not change, while the properties of the macerals themselves change during metamorphism, as a rule, converging with each other as the degree of coalification increases.

The nomenclature of coal macerals is regulated by the International Committee of Coal Petrology (ICCP) [3] and the Society of Organic Petrology (TSOP) [4].

Several dozen macerals are distinguished [5]. Such diversity is due to several reasons:

- botanical diversity of the original plant material (trees, bushes, moss, grass, etc.);
- anatomical diversity (root, trunk, branches, bark, sporulation organs, etc.);
- autochthonous and allochthonous conditions of accumulation of plant material;
- processes of transformation of biomaterial (gelification, fusinization, etc.).

Such a fractional division of macerals is justified from a scientific viewpoint. In practice, it usually comes down to dividing macerals into three groups: vitrinite, inertinite, and liptinite. Vitrinite is lignin-cellulose plant tissues buried in anaerobic conditions; inertinite has an initial organic matter similar to vitrinite, but, presumably, the burial environment is aerobic; liptinite is integumentary tissues and resinous substances buried in various environments [6].

Macerals differ under a microscope by characteristic petrographic features: color, reflectivity, relief, morphology, and size. These features are smoothed out with the growth of metamorphism, so, for example, the color and relief of liptinite become identical to vitrinite, the reflectivity of which in anthracites becomes equal to the reflectivity of inertinite.

The need to classify coal macerals by their genesis often leads to controversial conclusions, such as the origin of micrinit and mixtinite.

Currently, there is no unambiguous classification of all macerals, including their genetic origin.

Coal petrographic methods for researching organic matter (OM) have long gone beyond "coal" and are used for different types of OM, and therefore the term "organic petrology" is used much more often today about microscopic methods for studying rocks containing organic inclusions.

Over the past 50 years, the world has gained a great experience in microscopic OM research, thanks to the study of oil source rocks at different stages of rock transformation, from which "shale gas or shale oil" is extracted, as well as any sedimentary rocks with carbonaceous residues. American and European researchers are in the lead here [7,8].

Thus, macerals formed during diagenesis as a result of the transformation of the original plant material during gelification and fusification should be considered "primary". New (secondary) macerals are a product of the transformation not of the original plant material but of the already formed "primary" macerals under the continuing impact of regional metamorphism factors (temperature, pressure, and the associated process of gas generation), as well as individual factors: local stress pressure (dynamometamorphism), local increase in temperature during the movement of intraformational fluids, bacterial activity, chemical interaction of individual macerals.

Many years of experience in coal petrographic research at the IGTM of NAS of Ukraine has allowed us to gather extensive material on macerals assigned to a certain group by coal petrographic features. They are shape, color, brightness, and relief. But, they do not have any relation to this group by genesis and are the product of the transformation of the gelified matter, i.e. "primary" macerals of the vitrinite group, thus being "secondary macerals".

In such cases, errors may arise in evaluating the properties of coals as raw materials for a certain type of industry. At the same time, such cases provide material for studying the processes occurring in coals under various geological conditions and help to explain some complex gas-dynamic phenomena.

This research includes examples of identifying "secondary macerals" of coals depending on their genesis. Basically, this is a gelified substance transformed under various external impacts and as a result of internal processes.

The purpose and methods of the work are to give examples of "secondary macerals", to provide signs of their identification and recommendations for their accounting.

2. Methods

The authors applied the method of petrographic analysis of coal samples according to the produced coal petrographic preparations (cut sections with polishing of two planes – bedding and section). The research was carried out using optical microscopy (video optical complex: MBI-11, NV200, software - Scope photo.

3. Results and discussion

Secondary macerals formed by the transformation of the gelified matter with a local increase in temperature.

Various authors have named the gelified matter differently over more than a century of practice in describing coal macerals. We consider the names of gelified matter, groundmass, and vitrinite group macerals to be identical.

Fluids circulating in coal beds (liquid and gaseous mobile components of magma or gas-saturated solutions circulating in the depths of the earth) can be formed as a result of gas generation [9] and as a result of deep degassing of the earth's depths [10].

In both cases, this is an exothermic process, but with a difference in the composition and temperature of the fluids.

Migration of fluids (gas, water, oil) from the place of generation or accumulation due to increased pressures is always directed toward unloading along the shortest path – vertically into fault and crack zones, but primarily along the bedding upward along the rise of the beds [11].

Our research confirms these observations the results of this migration and the effect of fluids on coal matter we see on the walls of cracks and interlayer surfaces.

Let us consider both options:

I. Transformation of gelified coal matter under the temperature impact of gases moving along cracks.

In Figure 1, on the surface of the bedding of grade " Lean " coal in bed l_3 at the "Ilovaiska" mine, a crack is observed in the gelified matter (vt), the walls of which are zonally transformed by the temperature of gases moving along the crack.

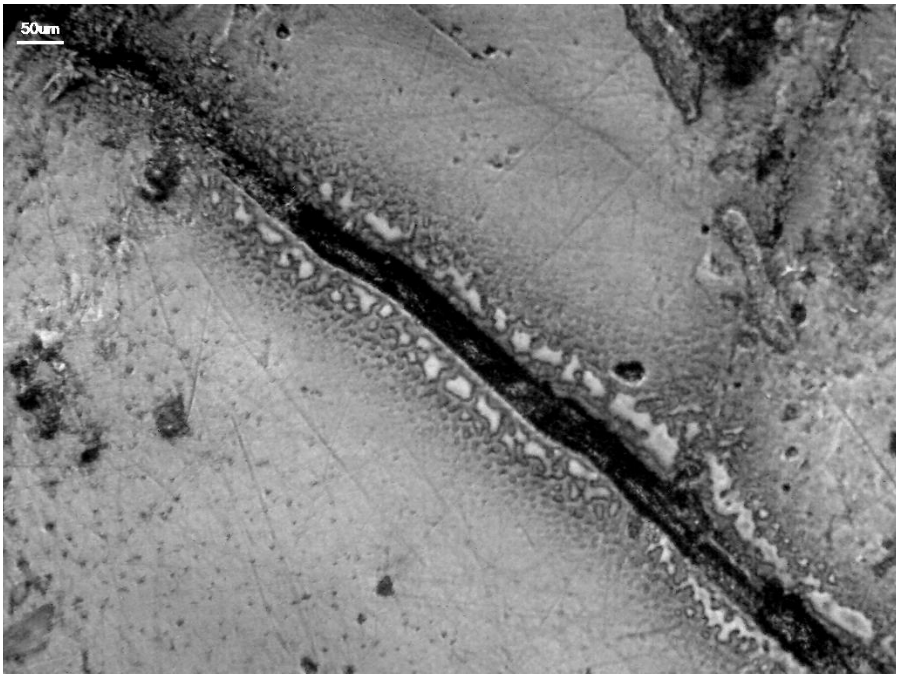

Figure 1 – Transformation of the gelified matter along crack walls

Two zones with a thickness of 30–50 μm were formed from the crack center outward. The first is represented by lighter-than-vitrinite lumps of isometric shape, 1-20 μm in size, and the larger ones were obviously formed by merging smaller ones. Here the temperature was the highest, the gelified matter was plasticized, liquefied, mixed, and degassed. The second zone, located further from the center of the crack, was under lesser temperature impact and is represented by darker than vitrinite lumps, 1–2 μm in size. This is probably one of the initial stages in the transformation of the gelified matter that occurred in the first zone.

Coal grade “Lean” is formed at a temperature of 250–300 °C and the ease with which the gaseous fluid transformed the already formed maceral with the formation of "secondary" maceral – light lumps of the first zone, indicates a fluid temperature of over 400 °C.

Measurements of the gray shades’ intensity (analogous to the reflectivity by the method of the IGTM of NAS of Ukraine [12]) in the micrograph (Figure 1) showed that the values of this indicator for the light lumps of the first zone exceed similar values for vitrinite and even semifusinite (the lower right corner in the Figure) and are equal to the values of inertinite or vitrinite of the anthracite stage.

When preparing a preparation for quantitative petrographic research (polished section briquette), the coal presented by the polished section in Figure 1 will be crushed to a size of less than 1 mm, and the light lumps of transformed the gelified matter along the crack will fall out of the context of the mutual arrangement of macerals and as separate objects, according to all coal-petrographic features, will be identified as macerals of the inertinite group, namely micrinite. However, the above facts show that they have nothing to do with the fusinization process, but are transformed vitrinite, i.e., "secondary" maceral of this group.

As for the lumps of the second zone (more distant from the crack), which are darker than the parent vitrinite, then in polished sections, vitrinite fragments with them as heterogeneous or contaminated will most likely be excluded when measuring, for example, reflectivity, which significantly reduces the risk of obtaining questionable results when using "secondary" macerals.

The results of this process are observed only on interlayer surfaces and are absent in polished sections made across the strike of the bed or tangentially.

II. The second variant of interaction between fluid and coal matter occurs in the case of gas-liquid fluid and consists of solidification of the liquid phase mainly on the interlayer surfaces of coals (Fig. 2).

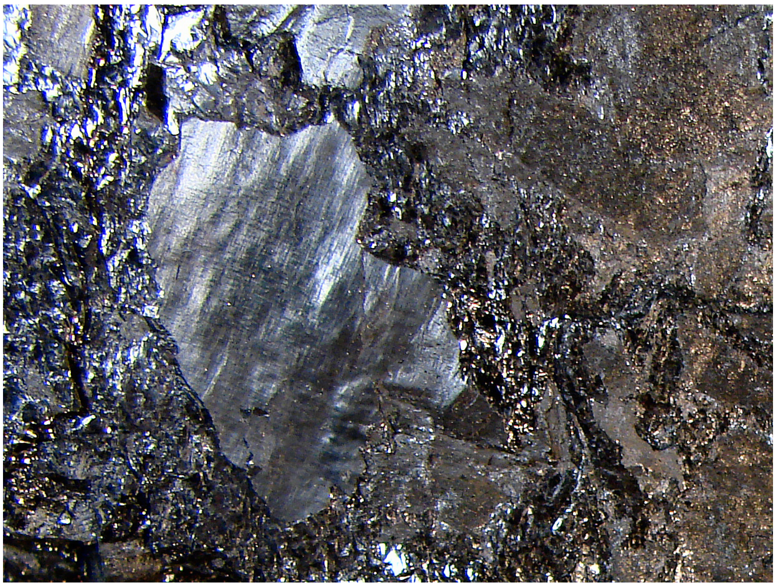

Figure 2 – The liquid phase of fluid solidified on the interlayer surface of coal

Similar films on interlayer surfaces in textbooks of past years are mistakenly interpreted as "slip mirrors", i.e. as a result of friction of two surfaces. The morphology of these surfaces would not allow this to be done – this is confirmed by the absence of terrigenous material and our research of the petrographic composition of these films, which showed their absolute homogeneity, in contrast to the gelified matter, which either cements at least a small number of macerals of other groups or preserves the structure of the original plant material.

In organic petrology, liquid hydrocarbon compounds released from macerals during their transformation and present in the rock in the form of films, deposits, or clots are called bitumen.

It is impossible to determine exactly at the coal petrographic level which bitumens are formed from which macerals, although most researchers consider alginite and liptinite to be the source material for them.

In our case, the film of the solidified liquid phase of fluid was formed from gelified coal matter in the process of gas generation by coal itself, it has nothing to do with alginite, although liptinite products may participate here.

In organic petrology, they have taken the path of proposing new names for macerals of organic matter in inorganic rocks. However, the same names in different branches of research can have different meanings, for example, in organic geochemistry the term "bitumen" is used to describe naphthoids.

In the present research, to avoid confusion in terminology, we do not propose new names for macerals but use examples to show the features by which "secondary" macerals can be identified.

Secondary macerals formed by the transformation of the gelified matter under local pressure increase.

Laboratory experiments on uniaxial compression of coal petrographic preparations directly on the microscope stage with video recording of the results conducted at the IGTM of NAS of Ukraine allowed us to record the wave-like nature of stress transmission through coal matter before crack formation.

After crack formation, the result of this impact is detected in polished sections during micro petrographic research in the form of thin (0.3-1 μm) light stripes that exactly repeat the crack contour and are parallel to each other (Fig. 3). The stripes are located only on one side of the cracks, along the entire length of the cracks, their number can reach ten.

Light stripes are the transformed gelified matter. Their reflectivity can exceed the reflectivity of the parent gelified matter by 2–4 grades, i.e. these stripes in coal of grade "Fat" can have values characteristic of anthracites.

When crushing such a section of coal in the process of making a coal petrographic preparation, we will obtain fragments of these stripes, which will be diagnosed as inertinite but, in fact, are transformed the gelified matter of a higher degree of carbonization, i.e. "secondary maceral".

When light stripes are formed, i.e. increasing the coalification of the gelified matter in this area, a certain volume of gas will be generated. The stripes (as noted above) are very thin, but their number is determined by the number of cracks during coal crushing. If crushing occurs spontaneously, such as during a sudden outburst of coal and gas, the increasing number of cracks will be accompanied by an increasing amount of the transformed gelified matter and an increasing amount of generated gas, and the process will become avalanche-like.

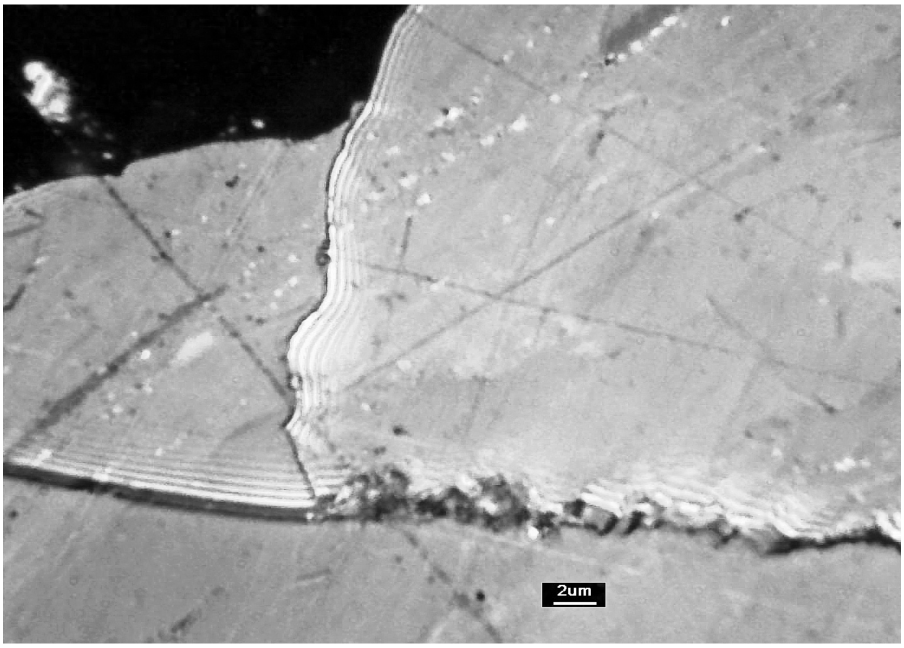

Figure 3 – Light stripes of transformed gelified matter as a result of the wave-like passage of stress from external pressure, the gelified matter

The formation of such stripes along cracks is most often observed in medium-grade coals, slightly less in highly metamorphosed coals and isolated cases of grades "Long Flame-Gas". They are not characteristic of the entire coal bed, but are found locally and can be as a sign of the tendency of coals to manifest gas-dynamic phenomena.

Secondary macerals formed by the transformation of the gelified matter under bacterial action.

Along with the role of aerobic bacteria in the humification of the original plant material, the issue of the activity of anaerobic bacteria in coals is widely discussed in the scientific literature.

Bacterial theory is one of the most common theories of spontaneous combustion of coals, along with the pyrite theory of the "coal-oxygen" complex and phenolic one.

Theoretically, it is possible that, along with methane of thermogenic generation, biogenic methane, which is a product of the vital activity of anaerobic bacteria, may be present among the combustible gases of coal basins [13].

We have discovered new formations on the interlayer surfaces of coals that look like colonies of microorganisms or products of their vital activity (Fig. 4).

Colonies of these microorganisms, in addition to the oval shape, can also be rectangular.

Their reflectivity is mainly lower than that of vitrinite, although it varies widely.

Bacteria are located on interlayer surfaces represented by the gelified matter; they are not noticed on either liptinite or inertinite. Probably, inert macerals and macerals formed from fats, waxes, and suberin are of no value to them as a nutrient medium.

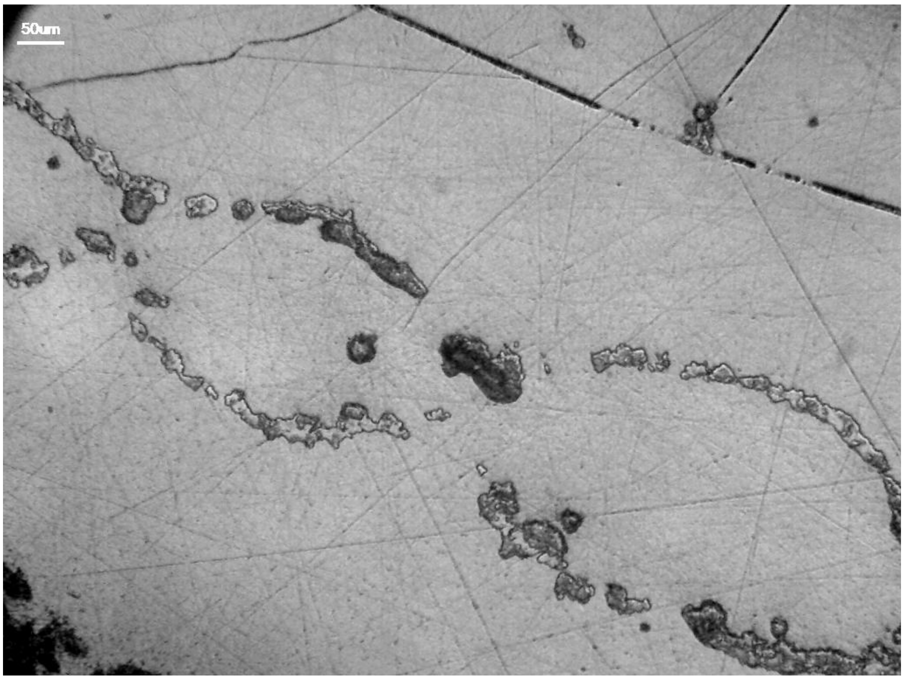

Figure 4 – Traces of bacterial activity on the gelified coal matter

On interlayer surfaces, they are located above the system of cleavage cracks and are not connected with them in any way. Consequently, their appearance is not connected with the replenishment of fluids of various genesis. They were formed during the inversion period of the basin development.

In coal-petrographic calculations of the maceral composition, fragments with these new formations will be erroneously (based on external features, mainly reflectivity) attributed to macerals of the liptinite group, although they are not such based on their genesis and physical properties (hardness). They should also be considered "secondary" macerals.

Being attributed to liptinite, these "secondary" macerals, especially at low stages of metamorphism, will erroneously overestimate the quality of coals, for example, in terms of sintering, i.e. use in the charge in the coke-chemical industry.

4. Conclusions

1. Coal macerals are formed during diagenesis.

2. During coalification, new formations of various genesis appear, which should be considered "secondary".

3. In regulatory documents on the identification of macerals, only their external coal petrographic features are considered without regard to genesis. This can lead to the classification of "secondary" macerals of the transformed gelified matter as liptinite and inertinite groups, which they are not, which will result in either an overestimate or underestimate of coal as a raw material for certain industries.

4. In percentage terms, the share of "secondary" macerals is small (except for

stripes of transformed vitrinite along tectonic cracks, especially in zones of crushed coal), but they should not be ignored.

5. The genesis of "secondary" macerals can be varied: stress thermal transformation of gelified substance by gas fluid, solidification of liquid phase of fluid on vitrinite of interlayer surfaces, dynamic metamorphism in fractured zones, bacteriological processing of the gelified matter, chemical reactions between liptinite and vitrinite during regional metamorphism.

6. In this paper, we present the features of "secondary" macerals of various genesis for their identification, but do not propose new names to maintain a balance between new coal petrographic facts for the classification of macerals and the overload of terminology.

7. In coal petrographic studies, in addition to the GOST preparation - polished section - briquette, it is desirable to have a polished section - sample for a preliminary evaluation presence of "secondary" macerals.

8. The fact that all "secondary" macerals have external signs of liptinite or inertinite and have sizes from 1 to 100 microns facilitates their diagnostics.

Recommendations:

1. When diagnosing a maceral as a "secondary" one, it should be excluded from the calculation of the petrographic composition, so as not to make a mistake in evaluating the properties of coal, since at this stage their properties have not been studied. Since the calculation is carried out according to the "chessboard" principle, excluding the fact of finding such a maceral will not significantly affect the result;

2. In connection with the development of research technologies, it is necessary to study the properties of "secondary" macerals by various methods, in particular, by FTIR-spectroscopy.

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ВТОРИННІ МАЦЕРАЛИ ВУГІЛЛЯ ЯК ІНДИКАТОР ПЕРЕТВОРЕННЯ ГЕЛІФІКОВАНОЇ РЕЧОВИНИ

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Анотація. Вугілля - цінна викопна сировина для енергетики, металургії, хімічної промисловості. Напрямок використання конкретного вугілля визначається низкою причин, у тому числі його петрографічним складом. Петрографічний склад вугілля як сукупність мацералів формується в діагенезі і за нормативними документами є незмінним. У даній роботі на основі багаторічної практики петрографічного вивчення вугілля показано мацерали, які за вуглепетрографічними ознаками (форма, колір, яскравість, рельєф) віднесено до певної групи, але за генезом не мають до цієї групи жодного стосунку, а є продуктом перетворення геліфікованої речовини, тобто "первинних" мацералів групи вітриніту, які, таким чином, є "вторинними мацералами". У таких випадках можуть виникнути помилки в оцінці властивостей вугілля як сировини для певної галузі.

Мета роботи - навести приклади ідентифікації "вторинних мацералів" вугілля залежно від їхньої генетики та дати рекомендації щодо їхнього обліку. Описано генетичні передумови виникнення "вторинних мацералів". Здебільшого це перетворена геліфікована речовина внаслідок стресового термічного впливу газовим флюїдом, застигання рідкої фази флюїду на вітриніті міжшарових поверхонь, динамометаморфізму в тріщинуватих зонах, бактеріологічного перероблення, а також досліджень, що проводились за допомогою відеооптичного комплексу: МБІ-11, НВ200, програмне забезпечення Scope photo. Генезис «вторинних» мацералів може бути різним: стресове термічне перетворення гелеподібної речовини газовим флюїдом, затвердіння рідкої фази флюїду на вітриніті міжшарових поверхонь, динамічний метаморфізм в тріщинуватих зонах, бактеріологічна обробка гелеподібної речовини, хімічні реакції між ліптинітом та вітринітом при регіональному метаморфізмі. Рекомендується при діагностуванні мацералу як "вторинного", виключити його з підрахунку петрографічного складу, щоб не помилитися в оцінці властивостей вугілля, оскільки на даному етапі їхні властивості не вивчені. Оскільки підрахунок ведеться за принципом "шахової дошки", виключення факту зустрічі такого мацералу істотно не позначиться на результаті;

Ключові слова: вугілля, петрографічний склад, мацерали, геліфікована речовина.