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### **Chemical, phase composition and properties of products of thermal decomposition of coal of Tavantolgoi deposit of Mongolia**

The results of investigation of chemical and phase composition of products of thermal decomposition of hard coal of the Tavantolgoi deposit in Mongolia were presented, namely, the solid residue of thermal decomposition of coal, tar fraction, liquid condensate and organic fraction separated from the condensate. Their compositions, temperatures of formation and phase transformations were determined. Relicts of aromatic hydrocarbons, ester groups and phenols were preserved in a small amount in the solid residue. The boiling condensate is an aqueous solution with a small admixture (~1 %) of the organic component that is the extract and traces of the resinous fraction. Condensate extract mainly contains phenol and its derivatives. The resinous fraction contains low-boiling oils and a phase that is thermally similar to asphaltenes, including up to 70 organic substances, with predominance (~40 % relative) of saturated hydrocarbons (cozanes), the presence of various groups of aromatic and polycyclic hydrocarbons, namely benzene derivatives, naphthalenes, phenanthrenes, hopenes, traces of phenols and cholestenes.

*Keywords:* phase decomposition, solid precipitate, resinous fraction, condensate, sublimates, organic fraction of condensate.

#### *Introduction*

The situation concerning the possibility and prospects for the industrial use of coal from the Tavantolgoi deposit in Mongolia that exists until recently was presented in some detail in the study of the phase decomposition of coal from this deposit [1]. It has been noted that the hard coals of Mongolia, in particular the Tavantolgoi deposit, are valuable raw materials for metallurgy, coke chemistry and the chemical industry [2, 3]. The explored reserves of the field, the technical characteristics of these coals, as well as a number of other fields in Central Asia (in particular, Tuva) provide the possibility of their integrated use [4–10].

However, the mined coals of these deposits are mainly used as a fuel for heat and power plants and the private sector due to the remoteness from industrial centers and the lack of necessary transport communications [10–12]. At the same time, dispersed soot and other products of incomplete combustion are formed in the furnaces of CHP plants and domestic furnace units due to the incomplete combustion of coal and the formation of cakes, which leads to the release of environmentally hazardous substances into the environment. The mountainous landscape and continental climate of Central Asia predetermine the formation of stagnant zones, smog in the confined spaces of mountain valleys, which leads to pollution of the atmosphere of urban and other settlements of these regions with products of flue gases with high concentrations of toxic substances, including carcinogens [13].

Despite numerous studies on the modification of materials of this type, the environmental problems of flue gases have not been resolved yet. In this regard, research on the dynamics of decomposition of coals from a number of fields in Tuva and Mongolia during their roasting, for example [1, 14], has been carried out as

part of the research program for creating technology for the efficient and environmentally friendly processing of mineral raw materials. In particular, when studying the thermolysis of coal from the Tavantolgoi deposit in Mongolia, the following products were obtained, namely, a solid residue, a resinous fraction (coal tar), a condensate of boiling liquid fraction and an organic extract of this fraction [1]. This article is devoted to the results of further studies of the chemical, phase composition and properties of the products formed during the thermolysis of coal from this field.

### Experimental

The composition of thermolysis products was studied by instrumental analysis methods. The structural characteristics of the solid products of thermolysis were determined by IR spectroscopy. Reference manual was used when interpreting the spectrograms obtained [15]. Liquid sublimate condensate and the resinous fraction were investigated by thermal analysis (DTGA) using an MOM-1000 model derivatograph (Paulik, Paulik-Erdey, Hungary). Samples of the organic component of the liquid sublimate condensate were analyzed by gas chromatography-mass spectrometry. The preparation consisted in threefold extraction of organic components with methylene chloride ( $\text{CH}_2\text{Cl}_2$ ). The compounds were identified by comparing with known substances from the library of mass spectra of the BIST 08 Library. Chromato-mass spectrometric studies (CMS) of the resins formed during thermolysis were performed using a system that included a gas chromatograph equipped with an interface with a highly efficient mass-selective detector. Mass chromatograms were obtained by total ion current (TIC). Compounds were identified using a computer search in the National Institute of Standards NIST-05 library, according to literature data and by reconstructing structures according to the nature of ion fragmentation during electron impact [16, 17].

Figure 1 shows the obtained IR spectra of the average sample of coal (curve 1) and the solid product of its thermolysis (curve 2) in order to assess the result of thermolysis.

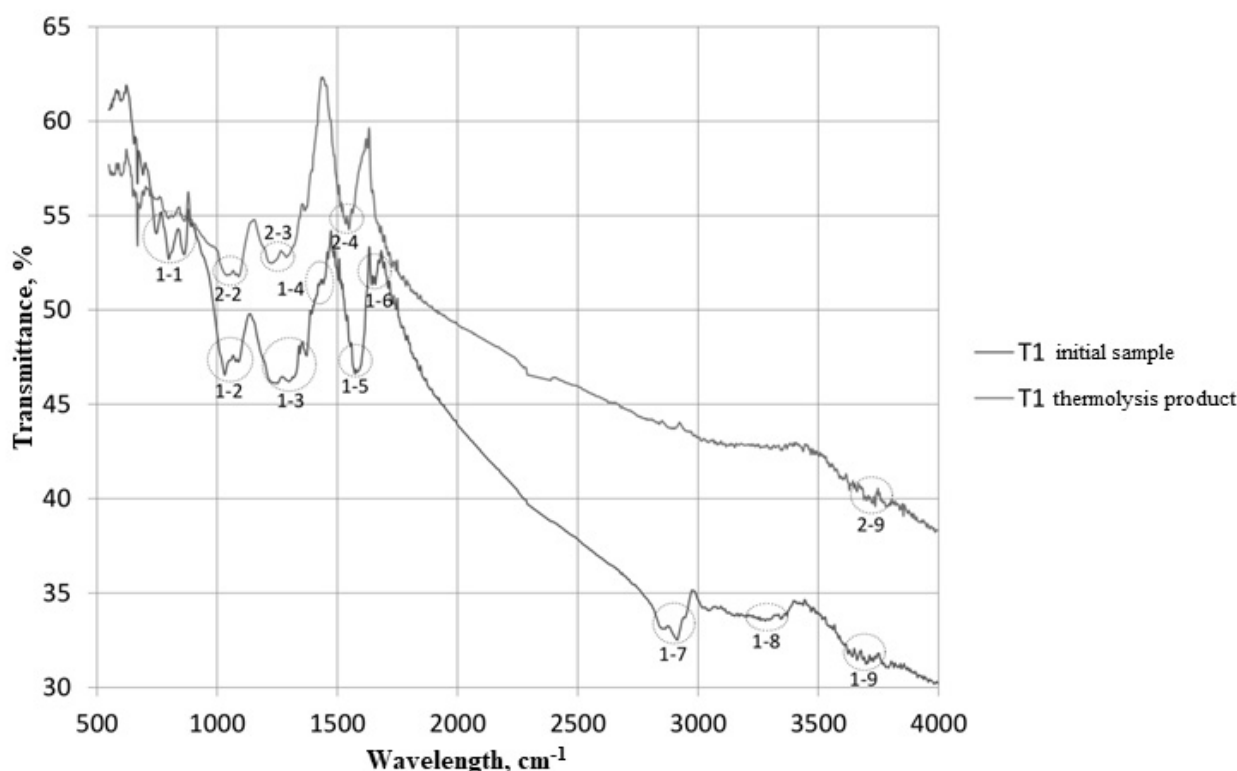


Figure 1. IR spectrograms of the average initial sample of Tavantolgoi coal (1) and its thermolysis product (2)

The infrared spectrum of the original coal sample (curve 1) shows absorption bands (peak 1) corresponding to the stretching vibrations of C–O–C and bending vibrations of CH, as well as the absorption bands (peaks 2 and 4) related to the vibrations of ether groups. Peak 3 corresponds to C–O vibrations of phenolic groups; C=O vibrations of quinolide groups are peak 5, and peak 6 is due to vibrations of intensive absorption of carbonyl groups. Peak 7 refers to vibrations of CH-aliphatic groups, OH-phenolic groups and carboxylic acids,

and peak 8 refers to weak vibrations of OH-phenols and carboxylic acids associated with the presence of a hydrogen bond. Peak 9 is due to vibrations of peripheral phenolic groups.

The IR spectra of solid thermolysis product (curve 2, Fig. 1) are significantly different from the IR spectrum of the starting material. Weak (relic) absorption bands of ether, phenol, and quinol groups are noted. There are no bands related to the vibrations of the CH-aliphatic, OH-phenolic groups and carboxylic acids.

*Thermal analysis of sublimates of liquid condensate samples.* DTGA samples of liquid condensate from sublimates obtained during thermolysis of an average sample of a batch and a sample prepared from a lumpy, heterogeneous in size starting material (~30 mm diagonally) turned out to be identical (Fig. 2).

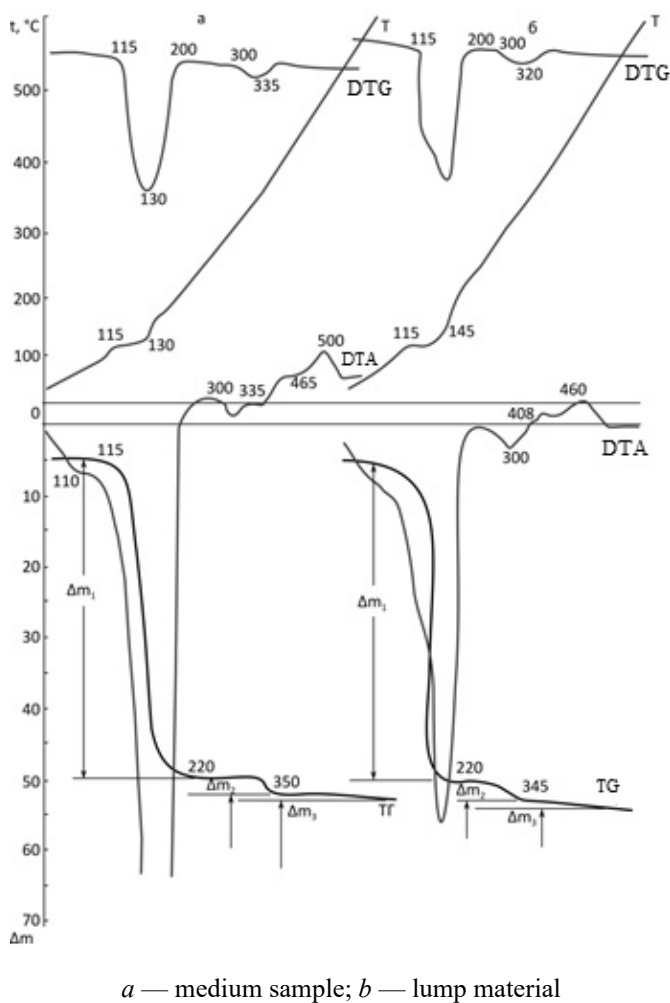


Figure 2. DTGA samples of condensate extract sublimates thermolysis

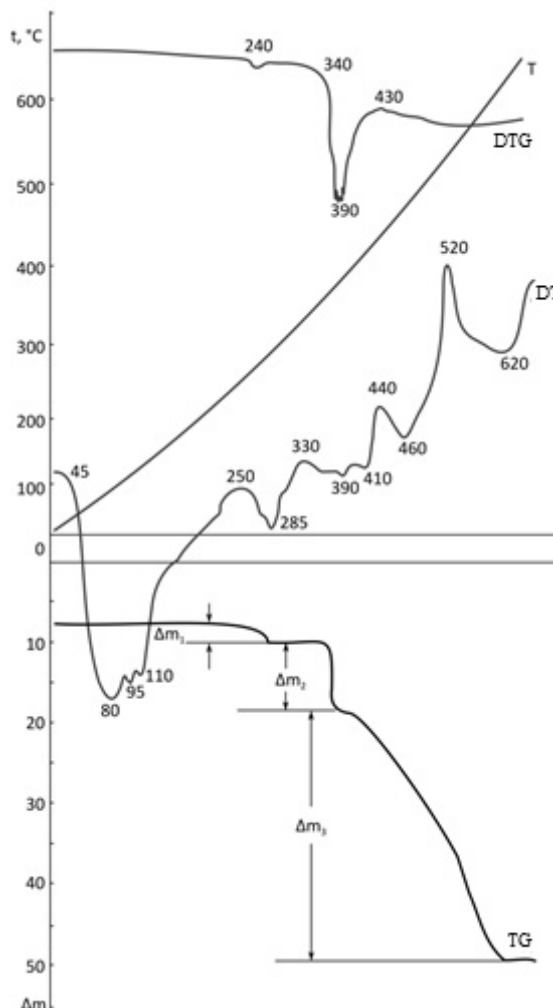


Figure 3. DTGA of tar fraction of coal

There is a deep endothermic effect of evaporation of an aqueous solution at 115–130 °C and with further heating a number of minor endo-effects are fixed at 300–340, ~ 400.460–500 °C. This is due to the fact that a small amount of the resinous fraction is formed during thermolysis of samples of this coal, in addition to liquid sublimates. And, apparently, all thermal effects and changes in the mass of the material observed at temperatures above 300 °C are due to the presence of an admixture of the resinous component in the condensate mechanically trapped or partially dissolved in the condensate. Accompanied thermal effects, the mass loss of samples of liquid condensate of this type of coal occur sequentially in three stages, namely it is 92 % at 115–130 °C, 5.6 % at 300–350 °C and then (up to 600 °C) it has the monotonous nature of the decline and it is about 2 %.

*Thermal analysis of the coal tar fraction.* The results of DTGA of the coal tar fraction are presented in Figure 3. The course of the thermogram curves shows that a series of combined softening processes ( $\geq 45$  °C) and sequential melting of the resin components (80–95–110 °C) occur in the temperature range of 45–110 °C,

while the mass of the material remains unchanged. At 230–240 °C, the beginning of the boiling of the liquid is noted, which is fully completed at 285–300 °C, and a dry porous product is formed at 310–320 °C. Starting at temperatures of 330–350 °C, dry decomposition of the sample material is observed. By the heating temperature of 600 °C, the sample mass is reduced to 0.5 % of the initial sample, and the entire sample is completely sublimated upon subsequent short exposure (5–10 min). From this it follows that the resin material is represented by low-boiling (about 230–250 °C) mineral oils and a phase that is thermally close to asphaltenes, with solid phase decomposition at 330 to 340 °C, into components, with higher temperatures (up to 600–650 °C) dry sublimation.

*The material composition of the extract sublimes liquid condensate.* The analysis showed that the condensate extract of thermolysis of lump material contained phenol (45.70 %) and its derivatives (in total, by weight, equal to 50.44 %), as well as impurities, namely cyclopentanone, cyclohexanone, diethyl phthalate, dimethyl derivative cyclopentene and 2-methylquinoline. In total, the mass of phenol and its derivatives was 96.14 % of the total mass of the extract.

The content of phenol is somewhat lower (37.54 %) than in the extract of the lump sample in the liquid extract of sublimation of the initial average sample of this coal, but the content of the derivatives of phenol is approximately the same. The total content of phenols is slightly less and equals to 92.02 %. The content of diethyl phthalate in the material is comparable to that in the lump sample extract. Also there is a small content of cyclohexane in the sample. Unlike lump sample extract, this extract contains a whole group of quinoline derivatives (total of 7.04 %). Thus, the main components of the condensate extract from sublimes of coal from the Tavantolgoy deposit, as well as from brown coal from the Baganur coal [18], are phenol and its derivatives. The extracts of brown and black coal differ in the content of the fractions — small in the mass of components.

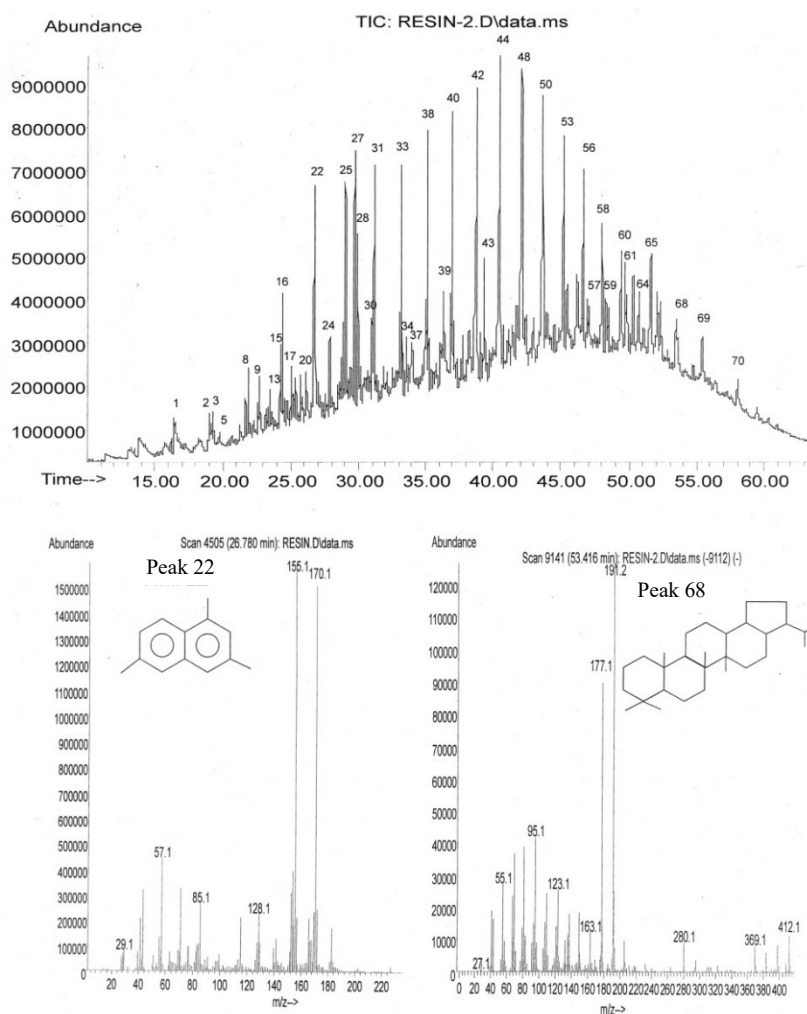


Figure 4. Chromato-mass spectrogram of the resinous coal fraction

*The material composition of the resinous fraction of coal.* The data obtained by chromatography-mass spectroscopy of the resinous fraction of Tavantolgoi coal (Fig. 4) show a complex composition.

This product consisting of 69 individual compounds belonging to different classes and groups of organic compounds are:

- aromatic hydrocarbons (0.765 % phenols; 15.842 % benzene group);
- polynuclear aromatic hydrocarbons (19.96 % naphthalenes; 11.61 % phenanthrenes);
- polynuclear hydrocarbons (10.50 % hopene-hopane; 0.97 % sterile steroids);
- saturated hydrocarbons (40.32 % cozanes).

#### Results and Discussion

As a result of thermolysis, Tavantolgoi coal decomposes into solid residue, the resinous fraction (coal tar) and sublimes including the gaseous component and liquid boiling condensate. The process of thermal decomposition of coal from the Tavantolgoi deposit can be represented by the following scheme (Fig. 5).

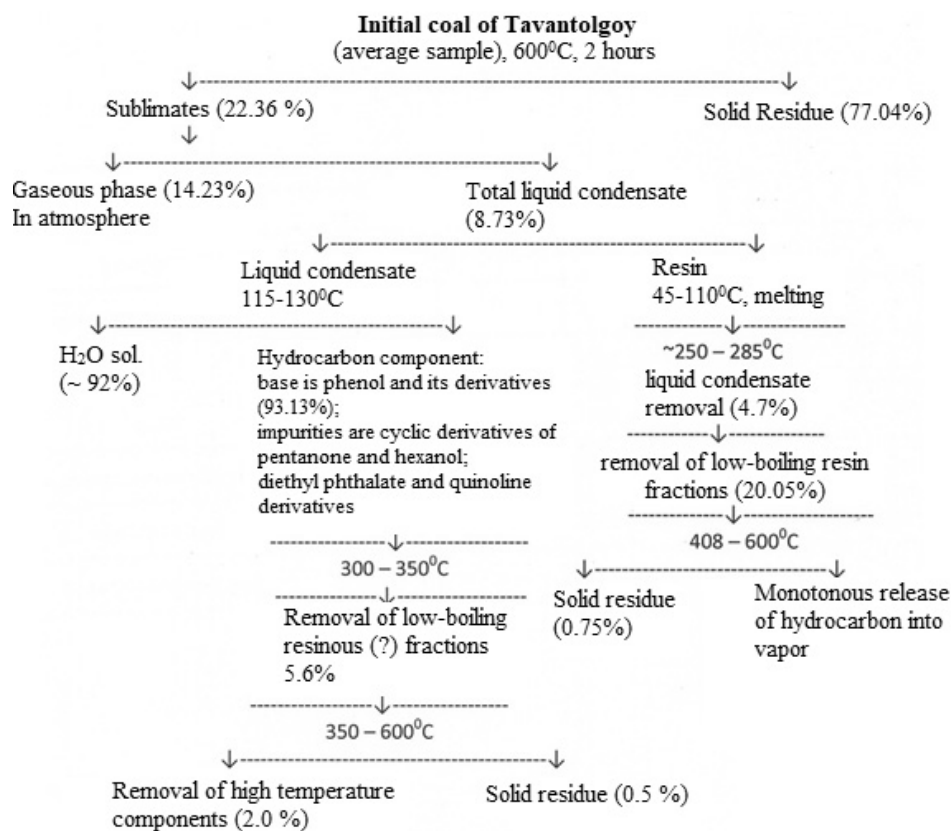


Figure 5. Diagram of the sequence of decomposition of coal during its thermolysis

The hydrocarbon components of the original coal are preserved in small amounts, namely, aromatic hydrocarbons, ether groups, and phenols. Low-boiling condensate, which is an aqueous solution, contains insignificant amounts of the organic component, the extract (up to ~ 1 %), and an impurity of the resinous fraction. This is confirmed by the data of DTGA condensate, where thermal effects in the temperature range of 115–130 °C indicate boiling of the aqueous solution, and above 300 °C, the removal of resinous impurities. Sublimate condensate extract mainly contains phenol and its derivatives. Samples obtained from lumpy or averaged material differ somewhat in the ratio of the contents of phenol and its derivatives, as well as in the presence of impurities in them, in particular quinoline and its derivatives.

The resinous fraction obtained during thermolysis (coal tar) contains low-boiling (at 230–250 °C) mineral oils; a phase similar in properties to asphaltenes, subjected to solid phase decomposition at 330–340 °C, and components with higher temperatures (up to  $\geq 600$  °C) of dry sublimation. It has a complex organo-mineral composition of 69 compounds, mainly saturated hydrocarbons — cozanes (~ 40 %) and compounds of various

groups of aromatic and polycyclic hydrocarbons, namely benzene derivatives (~ 16 %), naphthalenes (~ 20 %), phenanthrenes (~ 11 %), hopene (~ 10 %). There are impurities of phenols and cholestenes (<1 % each).

The results of the work confirm the conclusions made on the basis of the results of previous studies on the effect of the aggregate state of coal samples on the material and the effect on it during the storage of the environment on the magnitude of the coal gas during thermolysis and on the impurity composition of the sublimate condensate extract.

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Н.И. КОПЫЛОВ

## Монғолиядағы Таван-Толгой кен орны тас көмірінің термиялық ыдырау өнімдерінің химиялық, фазалық құрамы мен қасиеттері

Мақалада Монғолиядағы Таван-Толгой кен орны тас көмірінің термиялық ыдырау нәтижесінде түзілген өнімдерінің — көмірдің термиялық ыдырау қатты қалдығы, шайырлы фракциясы, сұйық конденсат және конденсаттан бөлінген органикалық фракциясының химиялық, фазалық құрамдарын зерттеу нәтижелері келтірілген. Олардың құрамдары, түзілу және фазалық түрленуінің температуралары анықталды. Қатты қалдықта аздаған мөлшерде ароматты көмірсутектер, эфирлі топтар мен фенолдар

қалдықтары қалады. Жеңіл қайнайтын конденсат құрамында органикалық компоненттен (~1 %) тұратын сулы ерітіндіні — экстракт пен шайырлы фракция қалдықтары құрайды. Конденсат экстракты фенол және оның туындыларынан тұрады. Шайырлы фракциясы жеңілқайнайтын майлардан және фазадан тұрады, ол термиялық қасиеттері бойынша асфальтендерге жақын, оның құрамы 70-ке жуық органикалық заттан тұрады, соның ішінде қаныққан көмірсутектер (~40 % жуық), ароматты және көпядролы көмірсутектер: бензол, нафталин, фенантрен, гопен туындылары мен фенол және холестен қалдықтарынан тұратындығы анықталды.

*Кілт сөздер:* фазалық ыдырау, қатты тұнба, шайырлы фракция, конденсат, возгондар, конденсаттың органикалық фазасы.

Н.И. КОПЫЛОВ

## Химический, фазовый состав и свойства продуктов термического разложения каменного угля Таван-Толгойского месторождения Монголии

Приведены результаты исследования химического и фазового состава продуктов термического разложения каменного угля Таван-Толгойского месторождения Монголии: твёрдого остатка термического разложения угля, смолистой фракции, жидкого конденсата и выделенной из конденсата органической фракции. Определены их составы, температуры образования и фазовых преобразований. В твёрдом остатке в небольшом количестве сохраняются реликты ароматических углеводородов, эфирные группы и фенолы. Легкокипящий конденсат представляет водный раствор с небольшой примесью (~1 %) органического компонента — экстракта и следы смолистой фракции. Экстракт конденсата в основном содержит фенол и его производные. Смолистая фракция содержит легкокипящие масла и фазу, по термическим свойствам близкую асфальтенам, включающей до 70 органических веществ, с преобладанием (~40 % отн.) предельных углеводородов, наличия различных групп ароматических и многоядерных углеводородов: производных бензола, нафталинов, фенантронов, гопенов, следы фенолов и холестенов.

*Ключевые слова:* фазовое разложение, твёрдый осадок, смолистая фракция, конденсат, возгоны, органическая фракция конденсата.

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