

## Research of Surface Physical and Chemical Properties of Coal in the Process of its Hydrotransportation

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### Abstract

In the article, in the laboratory and field experiments an evolution of surface physical and chemical properties of coal was traced in the process of its hydrotransportation. Laboratory imitation of the coal hydrotransportation process was effected by mixing water-and-coal hydraulic mixture  $F : L = 1:1$  in the turbulent mode during 48 hours that is equivalent to pulp movement in the coal pipeline at the rate of 2 m/s for the distance of 350 km. High-volatile coal with a grain size of 0-2 and 0-0.1 mm  $A^d=8.6\%$ ,  $V^{daf} = 37.5\%$ ,  $R_o = 0.70\%$  was used as the initial material. Disperse medium is distilled water, 0.2% solution  $Ca(OH)_2$  and 0.1% solution  $Na_2SiO_3$ . Before and after hydrotransportation, coal was researched by IR and UV spectroscopy methods. Oxidative-hydrolytic processes associated with addition of water to hydrophilic groups (carboxyl, carbonyl, etc.) and leaching of individual components into the liquid phase have been established. The reactions are intensified with the change in pH of the environment, that is, with the introduction of corrosion inhibitors of the coal pipeline.

Field modelling of the hydrotransportation process of coking coal was performed on a hydrodynamic torus, which was a ring 3 m in diameter made of a steel pipe with an inner diameter of 202 mm, installed vertically. The ring rotated with a direct current motor. The ring coal pipe was filled with hydrocarbon pulp to 2/3 of the volume. The initial material is a balanced coal charge for coking from the Donetsk basin ( $A^d= 9.0\%$ ,  $V^{daf} = 30.6 \%$ ,  $S_a^t= 2.2\%$ ,  $R_o = 0.21 \%$ , grain size 6 - 0 mm). The distance of landfill transportation of the charge is 2.000 km. The initial charge was investigated by IR spectroscopy before and after hydrotransportation for 300, 1000, 2000 km. The surface  $\zeta$ -potential of the coal material was determined for the charge before and after hydrotransportation. According to the evolution of IR spectra of the samples, the oxidation of the coal phase was established, which is most contrasted when comparing the spectra of the original coal and after hydrotransportation for 1000-2000 km. Changes in the spectra increase with the increase in the range of hydrotransportation. Significant changes in the shape of grains as a result of their rolling in the pipeline are already observed at the point of 450 km of hydrotransportation. Hydraulic transport leads to a significant increase in the  $\zeta$ -potential of the coal surface. This confirms its hydrophilic behaviour in the process of hydrotransportation.

**Keywords:** *Coal; Surface properties; Hydraulic transport; Molecular spectroscopy; Laboratory experiment; Field experiment.*

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## 1. Introduction

One of the main problems of hydrotransportation of coal, especially over long distances, is a change in the physical, chemical, and, therefore, technological characteristics of the organic mass during transportation. In particular, in the process of hydrotransportation, grinding of

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the organic mass of coal, its strengthening, wetting of clays, and changes in the shape of coal grains are observed.

Svitlyi established [1] that coal is crushed in pumps and pipelines during hydrotransportation. The change in the total degree of coal grinding reaches 1.4; 1.5; 1.8 at transportation distances of 500 km, 1000 km and 1500 km, respectively. According to Yu.G. Svitlyi's nomograms of coal crushing ability with the length of the coal pipeline  $L = 500$  km and material of uniform grain size passes into smaller classes: grains 3 mm - 68%, 1 mm - 47%, 0.5 mm - 39%, 0.3 mm - 33%, 0.06 mm - 23%. At the same time, rolling of coal grains in the pipeline of the main hydraulic transport system (MGTS) takes place, as well as the enveloping of coal particles with wet clay [2]. Erdman recorded deterioration of dehydration of hydraulically transported coal [3], and Yu. Get - deterioration of coking [4], which the authors associated, in particular, with its oxidation and other transformations in the process of hydrotransportation.

In subsequent studies, an attempt was made to separate the effect on coal of the liquid phase and mechanical influences. Thus, the results of our research with the help of IR spectroscopy and microscopy show the evolution of the surface characteristics of coal during its long-term storage, in particular in conditions of watered sludge storage tanks. As a result, there is a change in the hydrophobic-hydrophilic balance towards the hydrophilization of the organic surface. Long-term contact of coal with water causes a significant increase in the  $\zeta$ -potential, and, as a result, leads to an increase in the degree of hydration of the coal surface [5].

In another our study, new oxygen-containing groups were found in the peripheral chains of the coal macromolecule in the coal lying in watered sludge accumulators by the method of IR spectroscopy, which indicates an increase in the hydrophilic behaviour of the coal surface. At the same time, microscopic analysis on the surface of unused coal revealed an increase in the thickness of the oxy-film in proportion to the duration of the coal's stay in the sludge accumulator. In 60 days, the thickness of the oxy-film increases from 0-0.2 to 1.3-1.6 microns. Thus, the data of IR spectroscopy and microscopic analysis correspond [6].

In [7], it was established that the uniaxial compressive strength and modulus of elasticity of the organic matter of coal decrease with increasing water content in coal, and these two parameters have an exponential relationship with water content. At the same time, plasticity of hydrated coal increases. In water-saturated coal, uniaxial compressive strength and modulus of elasticity decrease by 48.3% and 37.6% compared to the initial dry state, respectively. Based on scanning electron microscopy data, the authors of [7], conclude that the number and size of microcracks in hydrated coal will increase with an increase in water content, and the structural integrity of the coal will deteriorate. Hydration of clay minerals causes a decrease in local coal cohesion.

The purpose of this article is to study the evolution of the surface physical and chemical properties of coal during its hydrotransportation using molecular spectroscopy methods. To achieve this purpose, the following research tasks are set: 1. study of changes in the surface physical and chemical properties of coal during its laboratory simulated hydrotransportation; 2. study of changes in the surface physical and chemical properties of coal during the field process of its hydrotransportation.

## 2. Experimental (Methods and materials)

The laboratory study was carried out by simulating the hydraulic transport of coal by mixing the water-coal-water mixture  $F : L = 1 : 1$  in a turbulent mode for 48 hours, which is equivalent to the movement of pulp along a coal pipeline at a speed of 2 m/s over the distance of 350 km. After mixing, the pulp was subjected to vacuum filtration. Carbonic matter and the dry residue of the filtrate were studied by IR and EPR spectroscopy according to the methods described in [8] using PerkinElmer -180 and PE - 1306 spectrometers. UV spectra were obtained on a Specord device. Coal with a grain size of 0 - 2 and 0 - 0.1 mm with initial quality characteristics  $A^d=8.6\%$   $V^{daf} = 37.5\%$ ,  $R_o = 0.70\%$ . was studied. The dispersion medium is distilled water, 0.2%  $Ca(OH)_2$  solution and 0.1%  $Na_2SiO_3$  solution.

Field modelling of the hydraulic transportation process of coking coal was performed on a hydrodynamic torus, which was a ring 3 m in diameter made of a steel pipe with an inner

diameter of 202 mm, installed vertically. The ring is rotated with a direct current motor. It was filled with hydrocoal suspension to 2/3 of the volume, which according to [9] ensures maximum approximation of the model conditions to a real linear coal pipeline. As an object of research, a balanced coal charge for coking of the Donetsk basin was taken ( $A^d = 9.0\%$ ,  $V^{daf} = 30.6\%$ ,  $S = 2.2\%$ ,  $R_o = 0.21\%$ , grain size 6-0 mm). Spectra were obtained by means of PerkinElmer-180 spectrophotometer. Coal preparation method [8].

### 3. Results and discussion

The laboratory and field process of coal hydrotransportation was carried out. The evolution of its surface physical and chemical properties in the process of hydraulic transportation was studied by molecular spectroscopy.

#### 3.1. Laboratory research

Analysis of the obtained spectra (Fig. 1.) shows that long-term contact of distilled water with coal (Fig. 1b, c) leads to a decrease in the intensity of the peaks at  $380\text{ cm}^{-1}$ , which correspond to valence vibrations of C-O groups in phenols [10]. The transition of phenol-containing compounds to water ("salting out") is probably taking place. The content of active groups belonging to various alcohols and ethers (range  $1120 - 1030, 1010\text{ cm}^{-1}$ ) [11-14], as well as minerals such as kaolinite, changes somewhat weaker, but similarly. A weak peak at  $1550\text{ cm}^{-1}$  present in the initial coal, which corresponds to the deformation vibrations of NH-groups in secondary amines and amides [15-16], disappears in the coal substance after filtration, which gives reason to assert about the "salting out" of these active groups.

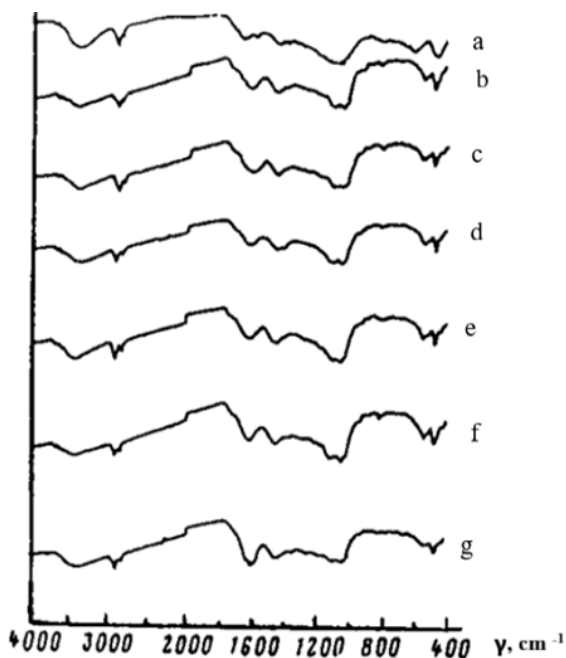


Figure 1. Infrared spectrum of coal: a – initial; b-g – after hydrotransportation, b, c – in water; d, e – aqueous solution of  $\text{Ca}(\text{OH})_2$ ; f, g – aqueous solution of  $\text{Na}_2\text{SiO}_3$ ; b, d, f – fraction 0 - 0.1 mm; c, e, f – fraction 0 - 2.0 mm.

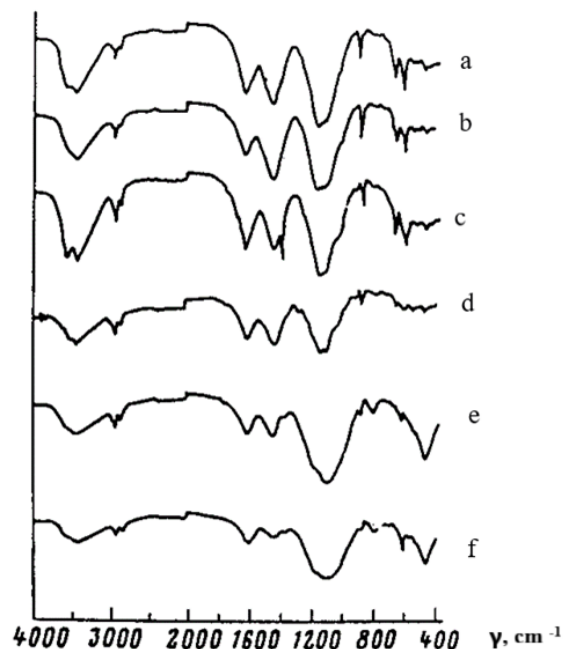


Figure 2. Infrared spectra of dry filtrate residues: a, b – in water; c, d – aqueous solution of  $\text{Ca}(\text{OH})_2$ ; e, f – aqueous solution of  $\text{Na}_2\text{SiO}_3$ ; a, c, f – fraction 0 - 0.1 mm; b, d, e – fraction 0 - 2.0 mm.

Mineral impurities (region  $700 - 500\text{ cm}^{-1}$ ) of coal matter also react with coal. At the same time, individual elements of pyrite inclusions and carbonaceous argillites, according to [10], act as active hydrophilic centres of coal. The effect of water on them catalyses oxidation-reduction reactions in which metal compounds participate. These processes may be the reason for the disappearance of medium and strong peaks at  $640$  and  $540\text{ cm}^{-1}$  in the spectra of water-

treated coal. In the spectra of Fig. 1-g, peaks of weak intensity appear at  $1200\text{ cm}^{-1}$ , which indicate the presence of groups  $=\text{C}-\text{O}-\text{O}$  [13-14] interacting with an aqueous solution of  $\text{Ca}(\text{OH})_2$  in coal. In the presence of  $\text{Na}_2\text{SiO}_3$  in coal, a peak at  $2100\text{ cm}^{-1}$  appears due to valence vibrations of groups in amino acids [13-14], a peak at  $1630\text{ cm}^{-1}$  indicating valence vibrations of  $\text{C}=\text{O}$  groups in quinoids [10]. All these chemical changes cause an increase in the hydration of the coal surface.

Analysis of the IR spectra of the dry residue of the filtrate (Fig. 2 a-c) shows the presence in them of compounds containing the groups  $-\text{OH}$ ,  $-\text{CO}$ ,  $-\text{COH}$ ,  $-\text{NH}$  (strong peaks in the region  $3400-3360$ ,  $2920$ ,  $2860$ ,  $1460-1462$ ,  $1170-1090$ ,  $875$ ,  $800-750$ ,  $670-650$ ,  $600-620\text{ cm}^{-1}$ ). This confirms the conclusion about the "salting out" of oxygen- and nitrogen-containing compounds into the aqueous phase, which is intensified in the presence of inhibitors.

UV spectra of filtrates (Fig. 3) are close to the spectrum of humic acids. This is especially evident for filtrates of aqueous solutions.

EPR spectroscopy shows the stability of the concentration of paramagnetic centres in the coal substance (regardless of the influence of hydrotransport factors) at the level of  $(1.0 - 1.3) \cdot 10^{18}\text{ spin/g}$ . Obviously, this is due to the absence of reactive free radicals on the coal surface.

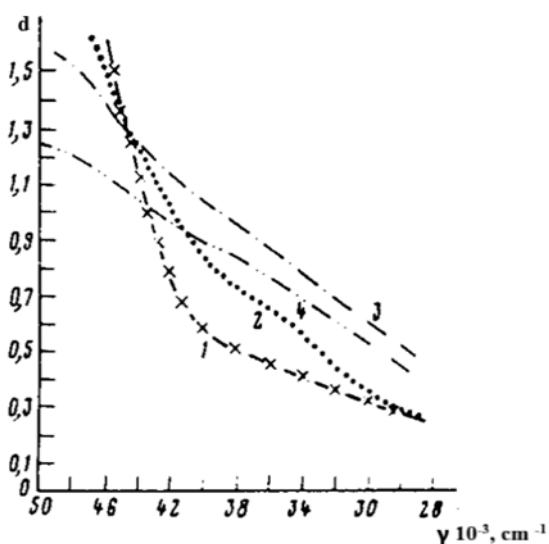


Figure 3. UV spectra of coal filtrates and humic acid salts: 1 - coal filtrate with water; 2, 3 - with aqueous solutions of  $\text{Ca}(\text{OH})_2$ ;  $\text{Na}_2\text{SiO}_3$ ; 4 - salts of humic acids.

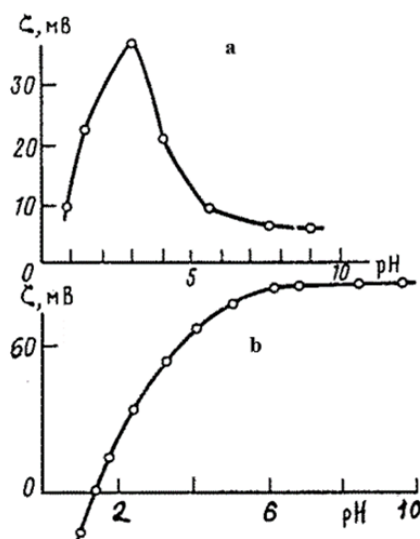


Figure 4. Dependence of the electrokinetic  $\zeta$ -potential on pH factor for a suspension of coal charge: a - initial; b - after hydrotransportation.

### 3.2. Field modelling

The analysis of the IR spectra of the charge (Fig. 5) shows that with the increase of the hydrotransmission range up to 2000 km, the changes in the spectra increase. At the same time, rolling of coal grains occurs (Fig. 6). At the same time, significant changes in the shape of coal grains are observed already when reaching 450 km of hydrotransportation.

There is a decrease in the intensity of the absorption bands at  $3050$ ,  $2910$ ,  $1450\text{ cm}^{-1}$ , which correspond to the valence vibrations of the  $\text{C}-\text{H}$  bond in the  $\text{CH}_2$  and  $\text{CH}_3$  groups [17-18], peaks at  $1380$  and  $1030\text{ cm}^{-1}$ , associated with the valence vibrations of the  $-\text{CO}$  and  $-\text{COH}$  groups [11-12, 19], as well as absorption bands at  $870 - 860$ ,  $750 - 700\text{ cm}^{-1}$ , caused by various vibrations of the  $\text{C}-\text{H}$  bond in the  $-\text{HC}=\text{CH}-$  and  $\text{C}=\text{C}$  [17-18]. The similarity of the changes in the spectra in the sections  $0 - 300$ ,  $300 - 1000$  and  $1000 - 2000\text{ km}$  of the transportation path indicates, obviously, the similarity of the ongoing processes, which are most clearly manifested at  $L_g = 1000 - 2000\text{ km}$ . These processes, in particular, include oxidation of the coal phase, which causes a change in absorption intensity in the regions corresponding to valence and deformation vibrations of polar groups ( $2100 - 2000$ ,  $1380$ ,  $1030\text{ cm}^{-1}$ ). Changes in other

areas of the spectra are probably caused by mechanical destruction, which causes changes in surface groupings.

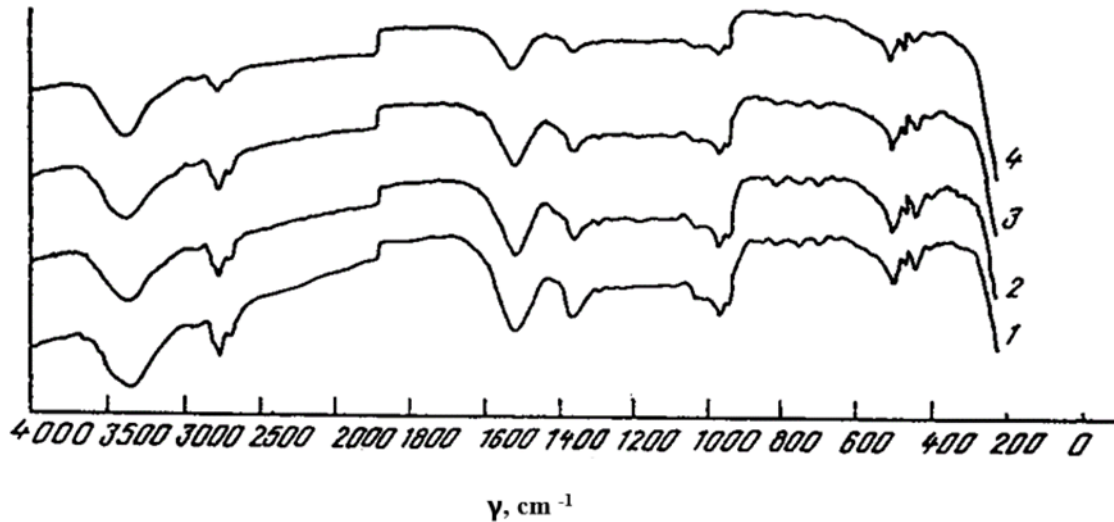


Figure 5. IR spectra of the balanced coal charge from the coal of the Donetsk Basin: 1 – initial charge, 2-4 – after hydrotransportation for 300, 1000, 2000 km.

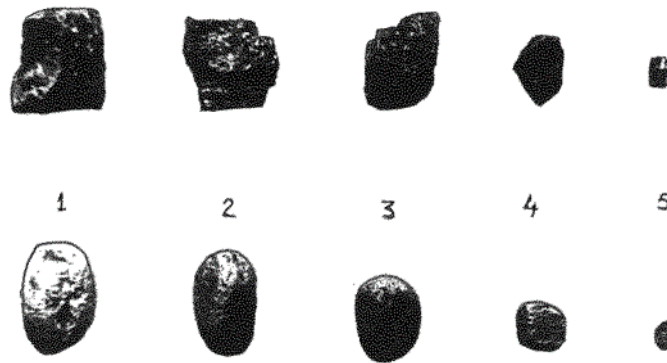


Figure 6. Change in the shape of coal grains during its hydrotransportation over a distance of 450 km.

The electroosmosis method was used to measure the  $\zeta$ -potential of the charge before and after hydrotransport on the device of the original design. The obtained results (Fig.4) show that the dependences of  $\zeta(\text{pH})$  for the charge to and after hydrotransport are sharply different. At the same time, hydrotransport leads to a significant increase in the absolute values of the  $\zeta$ -potential. This confirms the hydrophilic nature of the coal surface in the process of hydrotransport.

#### 4. Conclusions

In the laboratory simulation of the hydrotransport of highly volatile coal, oxidative-hydrolytic processes occur, associated with the addition of water to hydrophilic groups (carboxyl, carbonyl, etc.) and the leaching of individual components into the liquid phase. The reactions intensify with a change in the pH of the medium, that is, with the introduction of inhibitors corrosion of the coal pipeline.

During the polygonal physical modelling of the hydraulic transport of coal charge for a distance of up to 2000 km, oxidation of the coal phase occurs, which is illustrated by the evolution of the IR spectra of coal. At the same time, the most contrasting is the evolution of the IR spectra when comparing the spectra of the original coal and after hydrotransportation



for 1000-2000 km. Changes in the spectra increase with the increase in the range of hydro-transportation. Significant changes in the shape of grains as a result of their rolling in the pipeline are already observed at the point of 450 km of hydrotransportation.

### Symbols

$F : L$	<i>firm : liquid;</i>
$IR$	<i>Infrared;</i>
$UV$	<i>Ultraviolet;</i>
$EPR$	<i>Electron Paramagnetic Resonance;</i>
$MGTS$	<i>main hydraulic transport system;</i>
$A^d$	<i>Ash dry basis, %;</i>
$V^{daf}$	<i>Volatile matter dry ash free, %;</i>
$R_o$	<i>Vitrinite Reflectance, %;</i>
$Ca(OH)_2$	<i>calcium hydroxide;</i>
$Na_2SiO_3$	<i>sodium silicate</i>

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