Development and substantiation of technological solutions for the transformation of organic matter of fine-dispersed waste coal

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Abstract. A plant for liquefying organic matter has been create to solve the problems of using low-grade coals and waste coal utilization. The results of the performed studies are presented and the prospects for the solution of existing problem are shown, initial data are obtained for creating a pilot sample of the technological complex.

1. Introduction
Currently, the use of substandard coals and coal sludges is difficult. For example, a filter cake with a particle size of 0-1000 µm with a content of a class of 0-100 µm up to 90%, a moisture content of 30-45% and an ash content of 23-62% cannot be added to a commercial product, is not used as an independent product and is sent to dumps together with large rocks. As a result, the environment is significantly polluted and a significant share of the processed coal is lost (up to 10-12%). In addition, the circulating water is saturated with residual flocculants, which disrupt the technological process of coal preparation [1-5]. To solve the problems of using low-grade coals and waste coal utilization from coal enrichment, a plant for liquefying organic matter has been created. The paper presents the results of the research performed and shows the prospects for solving the existing problem. Thus, the purpose of the work was to obtain initial data for creating a pilot technological complex.

2. Characteristics of raw materials
To study the possibility of preparing suspension coal-water fuel based on fine-dispersed waste coal from the processing plants of mines “Komsomolets” and “N.a. S.M. Kirov” samples of filter cake weighing 2000 kg each were delivered. The delivered samples were analyzed in the coal chemical laboratory. The qualitative characteristics of the investigated samples are given in table 1.

Analysis of the data in table 1 showed that the moisture content of the filter cake samples submitted for research was consistently high $W_t = 35.2-40.8\%$. Unlike moisture, the ash content of the filter cake depends on the quality of the feedstock and changes simultaneously with it and can vary both in a narrow range of values ($A_d = 30.7\%; 26.8\%$), and in a wide range of deviations — up to 15.6\%
(\(A^d=32.8\%, 48.4\%\)) filter cake with PF w. “Named after S.M. Kirov”. Granulometric composition of filter cake from the processing plant of “Mine n.a. S.M. Kirov” included size classes up to 3.0 mm.

Table 1. Characteristics of samples.

| Index                                      | “PP named after S.M. Kirov” |
|--------------------------------------------|-----------------------------|
|                                            | sample No. 3 | sample No. 4 |
| Total moisture, %                          | 40.3          | 40.8          |
| Ash content (dry fuel), %                  | 32.8          | 48.4          |
| Volatile matter release (for dry ash-free state of fuel), % | 42.4          | 41.4          |
| Total sulfur (dry fuel), %                 | 0.15          | 0.13          |
| Higher calorific value (dry fuel), MJ      | 33.08         | 33.00         |
| Net calorific value of working fuel, MJ    | 11.8          | 8.7           |
| Granulometric composition, mm              |               |               |
| 0.250 – 3.0                                | 4.5           | 6.3           |
| 0.071 – 0.250                              | 26.8          | 17.3          |
| 0.071 – 0.071                              | 68.7          | 76.4          |
| Total                                      | 100.0         | 100.0         |

To confirm the feasibility of the proposed method for the destruction of the organic mass of coal, the original brown coal grade B2 was also used from the Chulym-Ugol open pit (Krasnoyarsk Territory) with the following characteristics: moisture content – 44.2%, ash content per dry mass (\(A^d\)) – 11.3%, volatile yield substances – 44.8%.

3. Research methodology for the preparation of suspension coal fuel

The resulting filter cake is actually a semi-finished product for obtaining coal-water suspensions with characteristics that allow for its efficient transportation through pipelines.

In the process of research, laboratory samples of suspension coal fuel were prepared from a mixture of filter cake and an aqueous solution of a reagent-plasticizer. Taking into account the initial size of the filter cake, a mixing or grinding chamber of periodic action of a universal shaker was used to prepare CWS samples. Dosing of the starting components was carried out in manual mode.

CWF samples were analyzed for the mass fraction of the solid phase, particle size distribution, and viscosity. Static stability was determined by the presence of sediment and water separation during storage of the sample under static conditions. The mass fraction of the solid phase was determined by the standard drying method in accordance with GOST 27314-91, the granulometric composition was determined by wet sieving on sieves 0.355 mm; 0.250 mm and 0.071 mm according to GOST 2093-82, ash content – according to GOST 11022-95.

Viscosity measurements were carried out on a RHEOTEST rotary viscometer in the shear rate range from 1.0 to 437.4 s\(^{-1}\) with a standard S2 cylinder system. The measurement temperature was 20±5 °C [5-10].

Further, the process of liquefaction of the organic mass of coal in an organic solvent was carried out by exposing the initial mixture (coal + organic solvent) to extreme physical influences: hydrodynamic cavitation, electromagnetic radiation, ultrasound, etc. Processes are necessary for deep processing of coal.

The purpose of the work is to reduce production costs, increase the efficiency of the process of destruction of organic compounds of coal raw materials and the mechanical reliability of the equipment used in the technological line.

To achieve this goal, coal raw materials were preliminarily subjected to wet grinding to grade 0-1 (3) mm, followed by demineralization of coal-water suspension by oil granulation to obtain coal concentrate and hydrogen suspension, while an organic coal suspension (OCS) prepared on the basis of the obtained coal concentrate. In addition, the technological line was additionally equipped with a block for oil granulation of raw materials, including units: preliminary wet grinding to class 0-1(3).
mm, mixing the obtained coal-water suspension with an oil agent and separating the coal-oil granulate from mineral impurities (rock) and water, an installation for producing an organic coal suspension based on coal-oil granulate, a unit for separating rock from water returned to the process and an additional ultrasonic unit cavitation.

Preliminary demineralization of the initial coal raw material ensured effective removal of the mineral part of the coal raw material to obtain demineralized coal concentrate or coal-oil granulate, which is directed to the preparation of an organic coal suspension, and mineral sediment that is removed from the process, which reduces production costs and increases the mechanical reliability of the equipment used (cavitators, pumps and etc.).

Moreover, the use of predominantly the original organic solvent as an oil agent leads to an increase in the efficiency of the destruction of organic compounds and a decrease in production costs.

The return of water obtained during dehydration of a hydrogen suspension, rocks, to the beginning of the process ensures cost savings and environmental friendliness of the process, since an organic solvent may be contained in the circulating liquid phase. As a result, its consumption is reduced and the discharge of contaminated water into external sedimentation tanks is excluded.

Figure 1 shows the technological line that implements the proposed method for the destruction of organic compounds of coal raw materials in an organic solvent by using extreme physical effects.

![Figure 1. Destruction of organic compounds of coal raw materials in an organic solvent.](image)

The technological line contains: a unit for preliminary wet grinding of raw coal raw materials to class 0-1(3) mm, unit for mixing the coal-water suspension with the oil agent, unit for separating the formed coal-oil granules and separating the hydrogen suspension; unit for dehydration of hydrogen
suspension; an installation for producing an organic coal suspension based on coal concentrate – coal of oil granulate and an organic solvent, which is mainly used as an oil granulation agent; node of extreme physical impacts, including zones of hydrodynamic and ultrasonic cavitation, high-frequency electromagnetic effects and temperatures.

Method and technological line for its implementation work as follows.

The initial coal is subjected to preliminary wet grinding to class 0-1(3) mm. The resulting coal-water slurry enters the mixer, where it is mixed with an oil agent. As a result of mixing, due to the fixation of drops of the oil agent on the lyophobic surface of clean coal particles, coal-oil granules–coal-oil granules are formed, the density of particles of which is less than the density of the liquid phase – water. As a result, particles of coal-oil granulate float to the surface, and rock particles, mainly lyophilic, settle to the bottom of the container. Thus, the separation of the coal-oil granulate from the rock is carried out.

Screening of the resulting mixture can also be performed, and since the formed coal-oil granules are larger in size of rock particles, the latter pass with water through the screen of the screen, and the top product of screening is removed from the screen. Hydrogen suspension is subjected to dehydration, for example by a centrifugal method, and the separated water is sent to the head of the process, and the rock is removed from the process.

The obtained coal-oil granulate in the form of coal-oil granules with a size up to 3-5 mm together with an organic solvent, which is mainly used as an oil agent, are directed to the plant for preparing an organic coal suspension. Vibrating mills, drum mills or high-intensity mixer-homogenizers are used as the main technological equipment for the preparation of OCS. The resulting OCS is pumped to the node of extreme physical influences. To implement hydrodynamic cavitation, cavitator pumps are used, the rotational speed of the working wheel(s) of which and the number of gaps correspond to the resonant frequency of oscillations of the molecules of the destructured organic compound. Achievement of frequency resonance is determined empirically in the process of commissioning when obtaining the maximum yield of light fractions in the finished product. Simultaneously or sequentially, the OCS is subjected to ultrasonic cavitation when using ultrasonic devices and to high-frequency electromagnetic action by using a generator of high-frequency electromagnetic discharges (figure 2).

Characteristics of the obtained granulate: moisture – 68.0%; ash content (A_d) – 1.8%. The resulting granulate was mixed with an organic solvent (fuel oil) in a ratio of 60:40 (by weight). The resulting coal-oil suspension was accumulated in a container and then passed sequentially through a RPA-type cavitator pump, an IZAP-1/22-OP ultrasonic apparatus and a generator of high-frequency electromagnetic discharges. The treatment of the coal-oil slurry was carried out in this way for 20 minutes. After this treatment, the resulting liquid was analyzed for the yield of liquid fractions. After distillation of samples of the obtained product on a Soxhlet apparatus, the total yield of liquid fractions, boiling up to 350 °C, was more than 90% in all samples obtained. In addition, under these conditions, more than 95% of the organic part of the coal was destroyed. The mineral residue was 1.5%.

4. Results and discussion

The liquefaction product obtained as a result of processing was distilled at a temperature of 380 °C and transferred to the Testing Laboratory of the West Siberian Testing Center (ZSITS) for fractional analysis. Table 2 shows the fractional analysis of the liquefaction product.
Table 2. Fractional composition.

| Name                                           | Mass fraction in the sample, % | Name                        | Result |
|------------------------------------------------|-------------------------------|-----------------------------|--------|
| Petroleum products                             | 43.54                         | NK, °C                       | 104    |
| Solid residue (over 380 °C), including:         |                               | 170–364, °C                 | 70%    |
| Ash as part of coal                             | 34.57                         | Total distillation          | 77%    |
| High boiling fractions                          |                               | The remainder in the flask  | 21%    |
| Water                                          | 12.44                         |                             |        |
| Gas losses                                      | 9.45                          |                             |        |

Thus, according to the results of the analysis of the ZSIC, it can be seen that part of the solid residue of the sample obtained as a result of distillation is a high-temperature hydrocarbon fraction. Therefore, the “High-boiling fractions” output can be summed up with the “Oil products” output.

Table 3 shows the result of liquefying the starting product obtained by mixing the components of the raw materials in the above proportions.

Table 3. Result of liquefaction.

| The name of the raw material components in the obtained sample | Mass fraction, % |
|--------------------------------------------------------------|------------------|
| Petroleum products, including:                               | 61.15            |
| Petroleum product, (170-364 °C)                              | 33.53            |
| Oil product, (over 364 °C)                                   | 25.63            |
| Petroleum product lost in the form of gas during the analysis by the ZSIC | 2.00             |
| Ash from coal                                                | 18.09            |
| Water                                                        | 12.44            |
| Losses, gas during sample distillation                       | 9.45             |

The following calculation was carried out to check whether 61.5% of oil products were actually obtained as a result of the liquefaction of coal, and not as a result of the influence of the “Waste oil” component and the transfer of a part of the mass of coal during its heating to 380 °C into the liquid phase. It is known [3] that in the process of thermal decomposition of coal there is a loss of mass of coal depending on temperature (table 4).

Table 4. Loss of mass of coal for different temperature ranges.

| Temperature range, °C | Weight loss, % | Weight loss by 1°C, % |
|-----------------------|----------------|-----------------------|
| 110–230               | 13             | 0.11                  |
| 230–380               | 4              | 0.03                  |
| 380–485               | 41             | 0.39                  |
| 485–850               | 42             | 0.12                  |

Based on the results of table 4, the obtained results of the fractional analysis were analyzed in terms of the temperatures used. Table 5 shows the results of calculating the amount of oil products obtained as a result of processing the original sample.
Table 5. Results of calculating the amount of oil products after refining.

| Component name                                                                 | Mass fraction, % |
|---------------------------------------------------------------------------------|------------------|
| Petroleum products, incl.:                                                      | 46.15            |
| Petroleum product, (170-364 °C) from mining                                      | 13.62            |
| Petroleum product, (over 364 °C) from mining                                     | 29.96            |
| Petroleum product, (170-364 °C) from coal sludge                               | 2.46             |
| Petroleum product, (over 364 °C) from coal sludge                               | 0.11             |

5. Conclusion

Comparing the results of tables 4 and 5, we find that if there was no liquefaction of the WMD of the original product, then only 46.15% of the raw material components would be obtained. The results of the experiments confirmed the technological feasibility of the proposed method for the destruction of the organic mass of coal in an organic solvent with a minimum content of the mineral part in the finished product. Thus, the organic part of the solid mass of the prepared coal-oil suspension turned into a relatively heavy organic liquid, which can be used as a boiler or motor fuel, as well as as a feedstock for the production of various hydrocarbon liquids.

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