CONVERTING COAL MINING INTO UNDERGROUND PRODUCTION OF ARTIFICIAL OIL

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ABSTRACT. A method for direct liquefaction of coal is presented. After separating the coal from waste rock gravitationally in an aqueous salt solution, drying and grinding, the purified separated coal is mixed with a paste-forming reagent. The paste is ground until a colloidal particle size and hydrogenated for 2 hours at temperature of 405°C and 11 MPa hydrogen pressure. Artificial oil was obtained with over 95 % yield with respect to the initial amount of purified coal when the contact of excavated coal with air was avoided. Based on findings, a technology is proposed for underground coal liquefaction with significantly reduced energy consumption of the process, removed environmental hazards and higher efficiency with respect to existing technologies.

(Key words: coal liquefaction, synthetic oil, underground

Introduction

Humankind needs oil - for industry development, transport and trade, and for its normal everyday life.

In refineries different parts of the crude oil are separated into useable petroleum products, such as gasoline, distillates (diesel fuel and heating oil), jet fuel, petrochemical feedstocks, waxes, lubricating oils, and asphalt - https://www.eia.gov/energyexplained/oil-and-petroleum-products/.

So, oil is needed to generate electricity to light our homes, run factories and machines, as a raw material to produce fertilisers to increase food production, etc.

For example, in 2020 the USA petroleum accounted for 34.7% of energy consumption and was used mainly in the transportation, manufacturing, and electricity sectors (USEIA, 2022).

Another example, in 2021, the total gasoline consumption (based on energy content) accounted for about 58% of transportation sector total energy consumption and 16% of U.S. total energy consumption, and (based on volume) 45% of total petroleum consumption - https:// www. eia. gov/energyexplained/gasoline/use-of-gasoline.php.

However, the oil price is steadily increasing - Figure 1.



Fig. 1. Oil price development in 35 years. Source: https://tradingeconomics.com/commodity/crude-oil, accessed 29.06.2022

The problem with oil prices and shortage is not a recent one. It was posed more than a century ago. The scientists and engineers searched for engineering means for using scientific knowledge for applied purposes, namely to produce synthetic oil (Burke et al, 2001; Mitchell, 2008; Vasireddy et al., 2011; Towler, 2014).

As early as 1913, more than 100 years ago, a great German chemist Friedrich Bergius was the first in the world to obtain artificial liquid fuel from coal in laboratory conditions (Schulz, 1999). The idea of the method consisted in catalytic hydrogenation of coal under the action of hydrogen at high temperature and high pressure in the presence of a solvent. The solvent facilitates coal extraction and the addition of hydrogen. The solubilised products, comprising mainly of aromatic compounds, are further upgraded by conventional petroleum refining techniques such as hydrotreating to meet final liquid product specifications (Burke et al, 2001). In 1931, Bergius was awarded the Nobel Prize in chemistry for his "merits in the introduction and development of a high-pressure method in chemistry".

During World War II, 18 plants were built in Germany and worked for coal hydrogenation (with 95 000 employees), liquefying annually 50 million tons of lignite and 10 million tons of coal. 92% of aviation petrol consumed by Luftwaffe were of synthetic origin (Schulz, 1999).

Indeed, such artificial fuel was about 10 times more expensive than natural liquid fuel obtained from natural oil.

In the modern epoch, the attention to the problem of coal liquefaction was attracted again in 1973, when Egypt and Syria declared during the October war at the Middle East that they would not supply oil at former prices to the countries that supported Israel in this conflict. This made Western governments activate researches in the field of coal processing into artificial liquid fuel.

On the other hand, nuclear tragedy of Chernobyl and Fukushima disaster led to an obvious apathy with respect to atomic power engineering all over the world.

Note that only 2% of integral coal resources have been exhausted (for comparison, 85% of total oil resources in the world have been already exhausted) (Sharovar, 1989).

Therefore, it is no wonder that efforts aimed at the search for more perfect methods of coal liquefaction are activated again.

The greatest recent achievement in this direction is the largest, on a global scale, plant producing artificial liquid fuel

constructed in China (Shenhua Ordos DCL Project, Inner Mongolia) (Plaza and Pevida, 2019).

The analysis of prime cost of finished product of this plant shows that about a half of all expenses in the total structure of its production costs falls on coal purchase and transportation to the liquefaction spot.

All this suggests a conclusion that new ways of cardinal reduction of underground mining price are needed, coupled with a complete denial of drawing coal (produced at a much lower price) to the ground surface and usage of highly efficient and compact methods of its underground direct processing into synthetic oil in the immediate vicinity of the mining face.

In contrast to silica sand or other oxygen-containing rock formations, coal is not a chemically inert material and its contact with oxygen irreversibly leads to gradual processes of endogenous oxidation, resulting in loss of not only calorific value, but also in decrease of synthetic oil yield during liquefaction of the solid fuel.

Therefore, the main object of the study was not only finding new, non-energy-intensive ways of underground coal mining, but also development of compact and precise methods for ultra-clean coal concentrates production directly in underground conditions, and identifying the correlation between the contact of freshly mined coal with oxygen in air and the effectiveness of its liquefaction.

Here, we propose such a method of coal mining and precise beneficiating with its subsequent underground liquefaction.

Materials and methods

Coal processing was carried out in the laboratory by the following methodology. Freshly mined from the working face regular raw coal (ash content 8.9%, volatile matter content of 45.2%, and sulphur content of 0.8%) was immediately immersed in a jar with an aqueous solution of Lewis acids, which also act as catalyst of the hydrogenisation process described below. The coal sample delivered to the laboratory, isolated from oxygen in the described manner was loaded into a steel mortar filled with a Lewis acid of density of 1.417 g/cm³. Under such conditions the cleanest coal particles in the raw material, which are not contaminated by gangue, remained afloat of such liquid while the rest sank to the bottom of the vessel. The material that sank was crushed down manually into smaller pieces using a steel pestle. During this operation, through the layer of such aqueous solution, a continuous bubbling of methane, which is insoluble in such environment, occurred. Also, under the impact of the crushing pestle, the pure combustible minerals were automatically released from their aggregates with gangue and floated to the surface of the liquid, thus avoiding additional irrational energy costs for their further destruction. Accordingly, at the bottom of the mortar less and less solid material remained with each time, which continuously reduced the efforts invested into the crushing.

When combustible minerals with density less than 1.417 g/cm³ no longer floated on the liquid surface, the contents of mortar was poured into a container with stirrer, from which the stirred-up suspension was passed through a hydrocyclone by means of a centrifugal pump, to make more intensive separation of the coal from the gangue. The emerging from the conical part of the hydrocyclone heavy product of the first step of crushing, which is a mixture of gangue minerals, was dewatered on a Nutsche filter. The light product suspension of

the first stage of crushing collected into the bucket at the outlet of the cylindrical part of the hydrocyclone, was heated to 100°C and fed into insulated ball mill equipped with a methane collector and a mechanical seal, flooded with hot (100°C) Lewis acid solution. In such manner, in the second stage of wet grinding, even cleaner coal extraction from aggregates with waste rocks occurred, accompanied also by methane release. That is because with decreasing density of the liquid phase, only practically clean from mineral impurities pure coal will float to the surface of the heavy liquid medium in the thermally insulated mill.

The hot suspension of the ground coal discharged from the thermally insulated ball mill, was separated in a high-speed (4000 rpm) laboratory centrifuge. The cake, pressed by centrifugal forces into the surface of the inner rotating rotor, was discharged from the centrifuge by a rotating screw, cooled, and returned to the steel mortar for mixing with the next batch of the raw mined mass. The centrifuge concentrate, which is a suspension of deeply purified from mineral impurities coal, was separated using a filtration centrifuge.

Without waiting for the wet cake pressed on the filtration centrifuge to cool down, 108 grams of the material was collected, which corresponded to a net of 100 grams of dry, demineralised solid carbonic material, and mixed with 200 ml of hot (160 °C) paste-forming agent, which is a mixture of (90: 10 by volume) tetralin (1,2,3,4-tetrahydronaphthalene— $C_{10}H_{12}$) with anthracene oil (a fraction of coal-tar resin boiling within the range 270-360 °C). In addition, during the mixing process almost complete removal of water vapour from the prepared paste occurred and was evacuated with a vacuum pump.

Then, to the paste obtained in the above manner and cooled to 80°C, 100 ml of isopropyl alcohol was added, and a dilution of the mixture was carried out in a high-speed mixer.

The diluted with isopropyl alcohol paste was then sent to a centrifugal disperser in which additional grinding of the solid phase of the system was carried out until a colloidal particle size was achieved. After this, the content of the disperser was reloaded into a steel autoclave, placed in a muffle furnace with a tangential inlet of compressed hydrogen (for a more efficient mixing), carrying out hydrogenisation of coal in the medium. For this purpose, the mixture in the autoclave was gradually heated from 80°C and initial hydrogen pressure of 2 MPa to the temperature of 405°C and 11 MPa hydrogen pressure.

After 2 hours of hydrogenisation, the autoclave was removed from the muffle furnace, cooled, unsealed, and the formed synthetic oil was separated from solid particles using the filter centrifuge. The extracted solid precipitate from such liquid was washed from the residues of synthetic oil with petroleum ether and dried with hot air, then sufficiently washed with warm distilled water, dried, and weighed. This material was directed to mixing with the cake received from pressing of the heavy product of the first stage of grinding on the Nutsche filter. The obtained tailing mixture of solid minerals was washed with hot fresh water using the Nutsche filter and removed from the process as waste. The formed washing water was mixed with the discharge of washing of the solid residue separated from the synthetic oil and evaporated to adjust the density of the salt-water solution to its initial value of 1.417 g/cm³.

Figure 2 presents a scheme of the procedures carried out. Similar experiments were carried out exactly with the same procedure and for the same coal, but already delivered to its destination by a railway from the place of mining.

The produced oil was characterised using the spectrometry method. All analyses for the content of the main five elements were performed using an optical emission spectrometer with a rotating disk electrode SpectrOil 100 (RDE-OES).

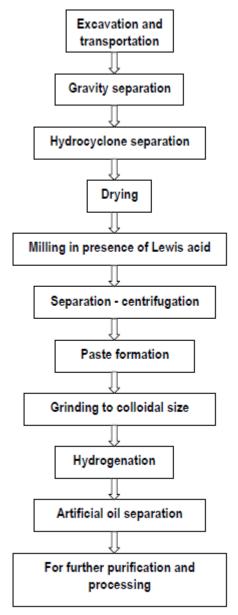


Fig. 2. Scheme of the experimental procedures carried out

Results and discussion

The dry residue from the autoclave weighed 2.74 grams. Since the autoclave was loaded with paste prepared according to the procedure described above, containing 100 grams of pure feedstock, the degree of liquefaction of the material which entered to hydrogenisation was 97.26%.

The degree of liquefaction obtained in the experiments carried out for the same coal, but already delivered to its destination by a railway, was equal to 82.48 - 83.56%, that is 13 - 15% lower than of coal freshly extracted in the mining face.

The elemental composition of the obtained synthetic oil is characterised by the following data (%): C - 79.2, H - 8.3 and (O+N+S) - 12,5. The most valuable part of the obtained artificial oil is the fraction of neutral oils. It makes up at least

half of the product obtained and is significantly enriched in hydrogen compared to the original artificial oil and it has less ballast (O+N+S). Its composition, in % is C - 83.5, H - 9.0 and (O+N+S) - 7.5.

Based on the analyses the non-refined produced oil can be classified as paraffin-naphthenic type oil. The following microelements were found in the composition of artificial oil: Cr; Sb; Ni; Sc; Zn; Co; Fe; Hf; Re.

At the same time, the content of these micro-impurities is so insignificant that they are not able to have a harmful effect on the processing of such raw materials.

The methane release can be gathered thus contributing to the multipurpose use of the coal, and liquefaction becomes more complete (due to the supply for hydrogenation of superhigh purity coal concentrate thoroughly impregnated, besides, by a hydrogenation catalyst). This essentially decreases the specific consumption of the initial raw material per each ton of the obtained liquid product

Based on the results from above-described and other experiments a technology has been proposed for underground liquefaction of combustible material (Brodt, 2016) - schematically presented in Figure 3.

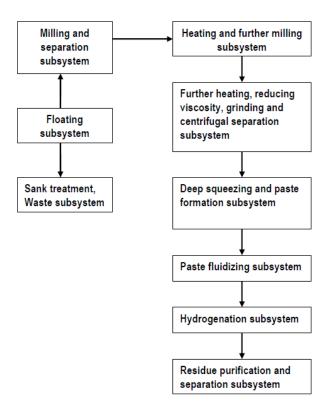


Fig. 3. High level schematic illustration of subsystems in the combustible material processing system

It is proposed the liquefaction process to be carried out underground using directly excavated coal and other combustible materials.

At that, harmful effect of underground mining and artificial liquid fuel production of the natural environment considerably weakens, since all final tailings of coal beneficiation and solid waste of artificial liquid fuel production remain in the underground worked-out space.

Besides, the denial of drawing crude solid fuel from coal mines to the ground surface and shipping the ready product in the form of artificial oil obtained in underground conditions, make it possible not only to abolish from the mine economy

such powerful electric power consumers as rope-skip hoists (the power of modern mine hoists reaches up to 15,000 kW) and coal-beneficiation factories (total electric power of the drives of all kinds of basic technological equipment installed at such ground-based production units can reach up to 10,000 kW), but also to wholly reject the services of railway transport. The mechanical losses of solid fuel due to the coal dust blowing by wind from the wagons is also avoided.

In this case, it is much more profitable to deliver the production to its destination by pipeline transport, which is about three times cheaper than coal transport by railway (not to mention mechanical losses of solid fuel due to coal dust blowing out of railway cars by wind).

Such proposal is of special interest for developing deep and extra-deep deposits of energy minerals. For instance, today the record depths reached by modern coal-mining industry are close to 2000 meters. Coal liquefaction at such extreme depths automatically leads to the working pressure growth in the zone of its hydrogenation up to 21 \div 22 MPa (without bringing in any power inputs from outside). This significantly intensifies such underground technological process and drastically facilitates the conversion of even the most coalified kinds of combustible minerals into artificial oil.

However, according to today's geological data, coal series cover much greater depths. This fact opens prospects of developing these huge supplies of energy minerals for converting mining from coal (solid fuel) production to ecologically clean production of synthetic liquid fuel instead.

With deepening level of underground mining, a steady growth of the bearing strata temperature allows a considerable increase in the share of free heat of the interior of the Earth in the total energy balance of such technological process.

We cannot ignore the fact that the saturation of coal and bearing strata with methane with growing coal seam depth also drastically increases. In this connection, the possibility of complex usage of the energy potential of the produced raw mineral by simultaneous methane extraction assured by the proposed method, also advantageously distinguishes said method from known technologies of solid fuel processing into alternative energy carriers.

A significant contribution to the profitability of underground coal liquefaction in comparison with ground-based production of artificial liquid fuel is due to a reduced hydrogen demand, since energy minerals freshly produced from the mining face are immediately isolated from the mine atmosphere by liquid medium. Thus, since the very beginning of coal separation from the mining face, air oxygen automatically loses contact with the coaly component of the combustible mineral and does not trigger the mechanism of its irreversible endogenic oxidation.

Meanwhile, the conventional approach to the production of artificial liquid fuel with coal liquefaction system located on the ground surface, far from the coal supplier, and deepening of underground mining, on the contrary, leads to an irreversible growth of the prime cost of the obtained artificial liquid fuel and to the growth of harmful effect of such human industrial activity on the natural environment.

Conclusion

The use of the proposed method in the coal industry as energy-saving and environmentally friendly, using underground, and not above ground production of synthetic

liquid fuel from such carbonaceous raw materials, promotes significant improvement of technical and economic efficiency of processing of such fossil fuels, particularly characterised by high content of non-combustible mineral impurities in it.

At the same time the method provides a comprehensive utilisation of the produced coal (due to deep extraction from the raw material and extraction of methane) and increase in the degree of its liquefaction (due both to the elimination of contact with oxygen in the air and addition of ultra-high purity coal concentrate, thoroughly soaked with catalyst of the hydrogenisation process, to the hydrogenisation). This significantly reduces primarily the specific consumption of raw solid fuel for each ton of liquid product.

In addition, the harmful effects of the whole complex of underground mining and production of synthetic liquid fuels on the environment are greatly reduced, as all final tailings of coal beneficiation, as well as solid waste from the production of synthetic liquid fuels, will automatically remain in underground mined-out space.

Moreover, the rejection of lifting raw solid fuel to the earth's surface from the coal mines, but lifting of finished products in the form of synthetic oil (or even the primary products of its processing) obtained in the underground conditions, allows not only to annihilate such powerful consumers of electricity as the cable-skip hoist and coal preparation plants from mine, but also completely abandons railway transport services. In this case, for coal producers it is much more profitable to use a coal pipeline for delivery of their product to the place of its destination, which is about three times cheaper than coal transportation by railway.

The complete economic triumph of the underground liquefaction of coal is determined not only by the progress of chemical science, but also by the progress of mining. In particular, the processes of unmanned development of coal deposits have already been invented, which can reduce the cost of coal by 2-3 times. Only in the combination of these two conditions is the final goal achieved - a fundamental change in the very purpose of the coal industry from the issuance of coal to the earth's surface to the pumping of artificial oil from the

The depth of the coal also plays an important role. As the depth increases, the hydrostatic pressure of the column of oil suspension supplied for hydrogenation increases. Thus, the deeper the mine, the easier the process of coal liquefaction.

On the whole, the main advantage of the proposed technological process is a steady growth of technical and economic efficiency of underground coal liquefaction with growing depth of coal production.

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