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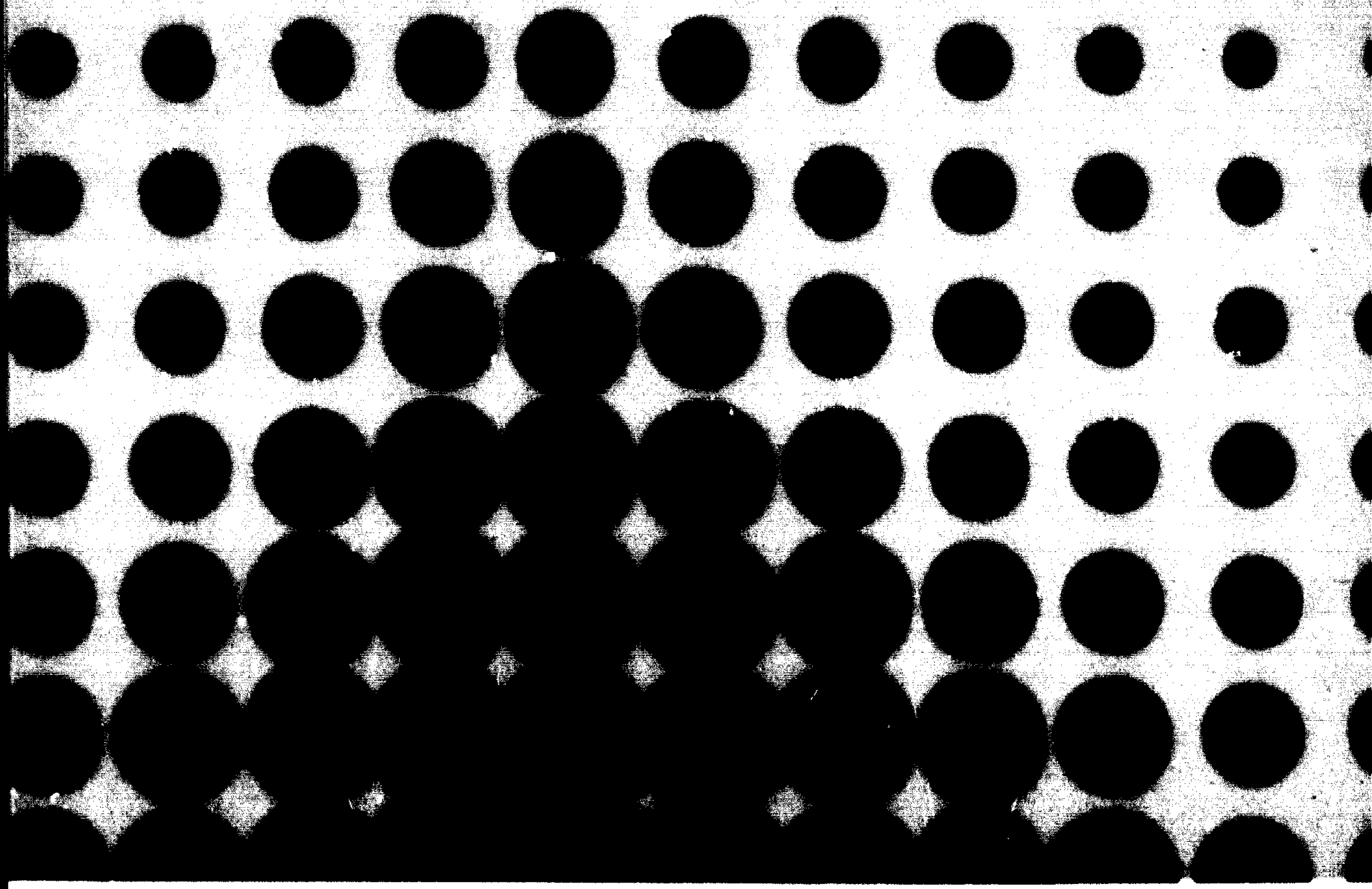

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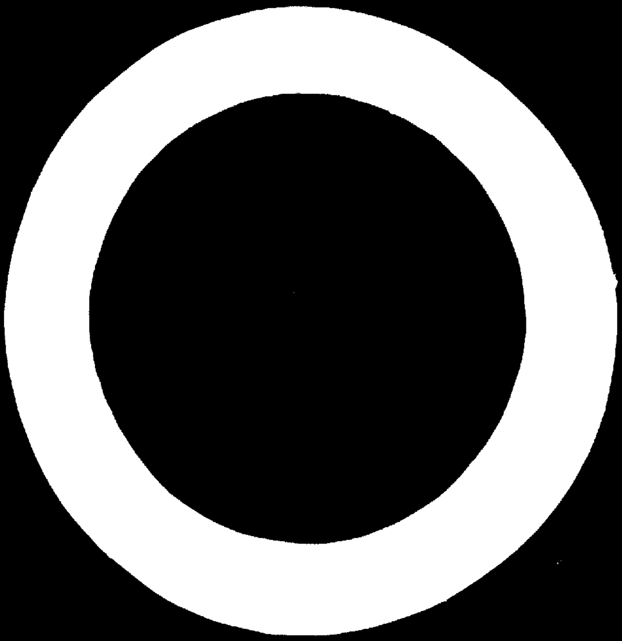

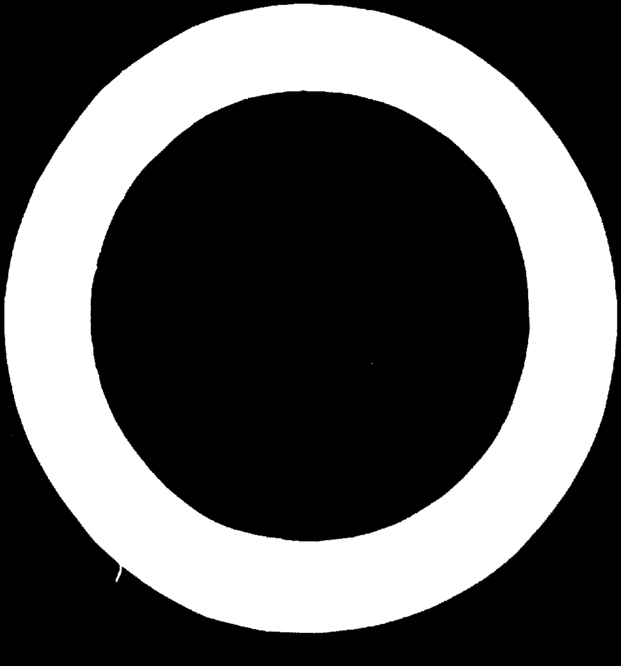

HYDROGENATION OF COAL AND COAL TARS

UNITED NATIONS INDUSTRIAL DEVELOPMENT ORGANIZATION
VIENNA

HYDROGENATION OF COAL AND COAL TARS

Review and annotated bibliography
of works published in 1959—1970

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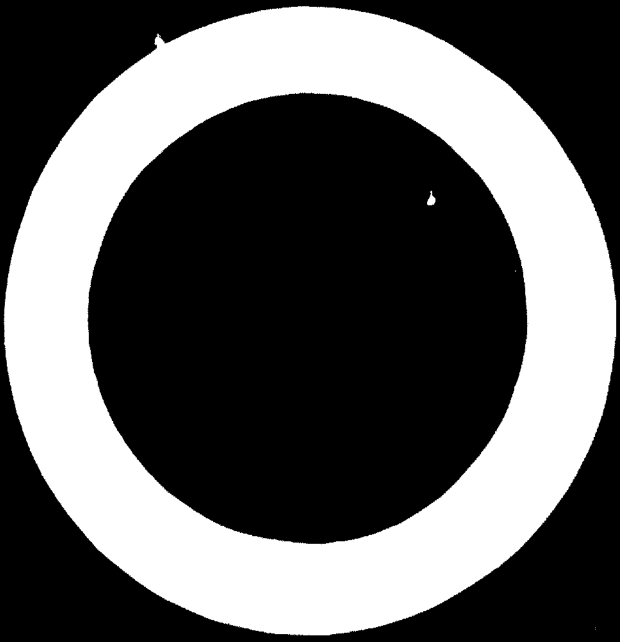
Foreword

This publication gives a brief review of coal hydrogenation processes and their application in industry since the Second World War. It is intended as a guide for those working on the chemical technology of fuel and those concerned with planning in the fuel processing industry. An annotated bibliography is provided, covering works published during the period 1959—1970 on the production of liquid fuels by hydrogenation. The studies reviewed are concerned with the main directions in the development of the hydrogenation of coal and coal tars, the economics of the new processes, and the prospects for their commercialization.

This study has been prepared at the request of UNIDO by the Institute of Fossil Fuels of the Ministry of Coal Industry of the USSR. The primary contributors to the study and to the work of preparing the annotated bibliography were Institute Members R. A. Konyashina, A. A. Krichko, D. P. Pchelina, A. I. Ryabtseva and T. A. Titova.

The views expressed are those of the consultants and do not necessarily reflect the views of the secretariat of UNIDO.



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Dollar ($) and cent (¢) refer to the US dollar and cent unless otherwise specified.

Ton refers to metric ton (1,000 kg) unless otherwise specified.

The following abbreviations are used in this publication:

kcal =kilocalorie
μm =micrometre (micron)
bbl =barrel (US)
SCF =standard cubic foot

Chapter 1

DEVELOPMENT OF THE COAL HYDROGENATION INDUSTRY

Hydrogenation processes were used in Germany and some other countries in the 1930s and the 1940s for the production of gasoline from coal and coal tar. At the beginning of the Second World War, Germany had a very large hydrogenation industry. The production of twelve German plants in 1943—1944 totalled 5.5 million tons of motor fuel, meeting one third of Germany's petroleum requirements [1]. Hydrogenation plants supplied about 50 per cent of the automobile gasoline and diesel fuel and 90 per cent of the aviation gasoline.

In the United Kingdom a plant in Billingham hydrogenating bituminous coal and coal tar had an annual production of 150,000 tons of automobile gasoline [2]. In Italy two industrial coal-tar hydrogenation plants were built in 1936 with a combined annual production of 240,000 tons of gasoline [3].

In 1942, a coal hydrogenation plant that produced 110,000 tons of gasoline and diesel fuel annually was put into operation in Korea [4]. Coal was hydrogenated at 480°—490° C under high pressure (250 atm for brown coal and 600—700 atm for bituminous coal) in the presence of iron and tin catalysts. The conversion rate of the organic component of the coal was 95—98 per cent; 55—61 per cent of the product consisted of liquid distillate products with a boiling point below 320°—350° C. In the next stages these products were converted into gasoline or gasoline and diesel fuel, with a yield of 45—50 per cent of the coal input. The hydrogen consumption was 8—10 per cent of the coal input. The liquid products of coal hydrogenation contained 10—15 per cent phenols and 3—5 per cent nitrogen bases, as well as other groups of compounds that could be used as chemical raw material. These were not extracted from the liquid-phase hydrogenates but were converted into hydrocarbons. The low yield of liquid petroleum products is explained by the high losses of heavy oil in the sludge treatment (low-temperature carbonization) in which 10—20 per cent of the coal organic component was converted into coke and gas. Thus, many of the advantages of deep hydrogenation of coal under high pressure were lost. This method was used because it was necessary to recycle the high-boiling fractions of hydrogenate.

Attempts to increase the production efficiency by replacing the 2 to 3-stage treatment of liquid-phase hydrogenates by a single-stage process at higher temperature (510° – 520° C) and pressure (500 – 600 atm) were unsuccessful for obtaining aromatic type gasoline. This process was used, however, by the hydrogenation industry of Germany [5], and analogous processes were developed in the Union of Soviet Socialist Republics (USSR) [6, 7] and in the United States of America [8].

Improvement of the techno-economic indexes of coal conversion was achieved only by reducing the pressure to 70 – 100 atm, increasing the temperature to 520° – 530° C and reorienting raw motor fuel production for yielding the scarce aromatic hydrocarbons [10, 11] along with high-octane gasoline [9]. However, it should be noted that the cost of gasoline obtained from coal never fell as low as the cost of gasoline obtained from petroleum. This was the main reason for stopping coal conversion at all the plants in the post-war period and changing over to hydrogenation of petroleum products. In the German Democratic Republic (GDR) coal was being hydrogenated only at the Leuna plant until 1959, after which this plant and others processing petroleum products were converted. However, research aimed at improving the coal-hydrogenation processes and finding economic methods for producing motor fuel and chemicals was continued.

At present, the problem of utilization of coal for the production of motor fuels and chemicals is again being discussed widely in the scientific and technical literature. Abroad, particularly in the United States, Governments have allocated considerable funds for basic and applied research on the use of coal as a raw material for various branches of industry. Investigations are also being carried out in India, Japan, Poland, the USSR and other countries.

DEVELOPMENTS IN THE USSR

Extensive research on coal hydrogenation [12] resulted in the completion of two experimental plants in the USSR before the Second World War. However, the war interrupted the development of hydrogenation processes. In the post-war period a chemical factory for hydrogenating coal was built, but for economic reasons no production of liquid fuel was planned.

At the end of the 1940s and in the 1950s, there were investigations aimed at improving the technology of hydrogenating coal to liquid products. On the basis of the progress achieved in the development of processes for hydrogenating hydrocarbon distillates of petroleum origin, an essential simplification, intensification and reorientation of the technology of converting the liquid-phase coal hydrogenates into valuable chemicals (phenols, aromatic hydrocarbons C_6-C_8, naphthalene) was successfully carried out. As a result, the techno-economic indexes of coal conversion were greatly improved, since the process pressure had been reduced from 300—700 atm to 75—150 atm and the hydrogen consumption had been reduced by 30—50 per cent.

The process of solvent extraction of solid fuels was developed in the USSR in the pre-war period [13], allowing the production of motor fuel and some chemicals from bituminous coal, brown coal, peat and oil shales. In the 1950s a semi-commercial plant for solvent extraction of peat was designed but not built.

In this process, liquid products were first obtained from solid fuel by dissolution at high temperature (400—425 C) in the presence of organic solvents. The liquid was then separated from the solid residue by filtration or destructive distillation (coking, pyrolysis, etc.). The conversion rate for the organic component of coal was 70—80 per cent, whereas in the case of peat or oil shale the conversion was 90—95 per cent. Research on solvent extraction of bituminous coal carried out in the United States resulted in a process of ashless pitch production; this process was recommended for technical realization, since ashless pitch would provide a raw material for the production of carbon and graphite materials [14]. The pitch yield was 60 per cent by weight of the coal input. The cost of the plant for processing 3.3 million tons of coal per year was $57.2 million. The cost of a ton of pitch was $2.50, assuming a coal cost of $2.80 per ton.

A process for hydrogenation of bituminous and brown coal to obtain chemicals along with liquid and gaseous fuel is now being developed in the USSR [15]. This process differs from the known German process in that it uses lower hydrogen pressure, fewer stages and less hydrogen. The chemicals obtained (aromatic hydrocarbons, phenols, naphthalene, solvents) are more valuable than fuel. Preliminary techno-economic calculations show that under certain conditions this process of coal hydrogenation should be profitable.

A process is also being developed for combined coal and petroleum hydrogenation at low pressure. In the case of coal, the organic component is converted into a liquid from which black oils with a low sulphur content (0.5—0.8 per cent) chemicals and motor fuel are obtained [16]. The process is based on new principles: It is carried out in the presence of petroleum products boiling above 240° C, various additives and a combination of catalysts. The petroleum products are donors of hydrogen, while the various additives initiate the conversion of the organic component into liquid and prevent secondary polymerization reactions. The use of these donors and initiators permits reductions of 50—60 per cent in pressure, 50—70 degrees in temperature, 50—70 per cent in hydrogen consumption.

and 30—50 per cent in gas formation. Liquid phase hydrogenation also removes much more sulphur from the residue.

The industrial application of liquid-phase hydrogenation of coal and petroleum can solve not only the problem of better coal utilization in the national economy, but also that of producing low sulphur fuels, basically black oils, from petroleum.

DEVELOPMENTS IN THE UNITED STATES

Investigations on coal hydrogenation have been carried out in the United States since the 1920s. The first semi commercial plant was put into operation at Baton Rouge, Louisiana, with the capacity to hydrogenate 25 tons of coal daily.

As part of the 10-year programme of the United States Bureau of Mines, and on the basis of the investigations carried out in Louisiana, a semi-commercial plant with a daily capacity of 50 tons of coal was built in 1949 and operated until 1953 [17]. Although some new processes, particularly reforming, were used in conjunction with the German technology, the scheme was not economically advantageous [18].

A semi-commercial plant for hydrogenation of coal, built by the Carbide and Carbon Chemical Company, with a daily capacity of 300 tons of coal, was operating during the period 1952—1956 [19, 20]. The hydrogenation process (500 C, 200—400 atm) produced mainly aromatic hydrocarbons, phenols and other chemicals. However, the economic indexes of the plant operation were not very satisfactory. Attempts to improve the process were carried out over a period of 5 years at a plant with a daily capacity of 12 tons of coal.

Research and experimental and project work on developing economically advantageous methods of coal hydrogenation have been carried out in the United States since 1960. Coal, petroleum and electrical corporations, as well as government institutions, take part in this work, which is being co-ordinated by the Bureau of Mines in the United States Department of the Interior.

In the literature one finds reports of the development of technical processes for producing gasoline and other fuel and chemical products as cheaply from coal as from petroleum. However, these reports are frequently too general; technological principles are explained but the most important engineering data may be left out. It is therefore difficult to judge the present actual state of the problem and to estimate achievements.

One of the most fully elaborated processes for producing gasoline from coal is the Consol process developed by the Consolidation Coal Company [21, 22]. The basis of this process is a modernized solvent extraction process followed by hydrogenation of the extract into gasoline at 430 °C and 200 atm. Hydrogen is obtained from natural gas and from the waste gas produced during hydrogenation, as well as from the semicoke produced from the residues.

Experiments carried out on a pilot plant with a daily output of 10 m^3 of gasoline resulted in a project for processing 8.3 million tons of coal and obtaining 3.8 million cubic metres of gasoline per annum at a cost of 10.5 cents per gallon, which corresponds to the cost of gasoline of similar quality obtained from petroleum. The cost of the project was $225 million. The high cost was due to the low efficiency of the old technology employed (the Pott-Broche process developed in Germany). It was only because of the low cost of coal in the United States that the cost of the gasoline could be kept so low. There are plans to commercialize the process in 1975.

Another process is the "H-Coal" process developed by Hydrocarbon Research, Inc. [23]. Coal is hydrogenated under high pressure of hydrogen (220—300 atm) in a fluidized bed of an active catalyst. The process is well known in the petroleum industry. It is worked out in two variants: single- and two-stage. The liquid and gaseous products in both variants are refined like petroleum. The products obtained consist mainly of gasoline, gas oil, gas, and small quantities of chemicals and black oil. When the two-stage process is used, the following products are obtained: gasoline, 23 per cent; gas oil, 12 per cent; black oil, 5 per cent; gas with a calorific capacity of 8,400 kcal/m^3, 280 m^3/ton.

In the literature the process of coal hydrogenation in a fluidized bed is described only in a general way; many important technical details are not reported. A pilot plant is now under construction; its daily output will be 9 tons of gasoline from 20—25 tons of coal. Performance data obtained in this plant will be used as the basis for building a full-scale plant for producing gasoline from coal.

The production of gasoline and black oil from coal by this method is stated to be profitable if the coal capacity is between 5,000 and 15,000 ton/day. It will yield a profit of 3.6—8.1 per cent, and the investment will be paid back in 6 to 9 years [26].

COED is the name of a coal-to-crude process developed by the FMC Corporation [24]. The process consists of a stepwise pyrolysis of coal in a fluidized bed followed by liquid products processing. A gas of high calorific value, hydrogen for ammonia synthesis and a semicoke are produced in addition to the liquids. The process was tested in a plant with a coal capacity of 50 kg/h; a plant with a capacity of 10 ton/day is planned.

Basically, there is nothing new involved in these processes. They are either modernizations of old schemes or adaptations of processes used in other fields, especially in petrochemical technology. Nevertheless, some difficult technical problems have been solved, and the resulting simplification, together with the low cost of coal, has made it possible for the price of gasoline from coal to compete with that of gasoline from oil.

THE SITUATION IN WESTERN EUROPE

In Western Europe, the production of motor fuel from coal by means of the existing hydrogenation processes is not profitable. In the Federal Republic of Germany (FRG) the cost of gasoline from coal is three times higher than the cost of gasoline from petroleum [25].

As indicated above, most of the hydrogenation processes developed in the United States are characterized by complex technological equipment, low yield of the final product (only 30 per cent for gasoline from coal, for example), high hydrogen consumption etc. In addition, these processes are multi-stage. Even with a low cost for the raw coal, the production costs remain rather high. These are not the only reasons, however, for the lack of exploitation of coal liquefaction processes. Factories are needed to manufacture the specialized processing equipment for hydrogenation. Building such factories will appear attractive economically only if there is a decision to develop a large-scale coal hydrogenation industry with an annual output of tens of millions of tons of products.

THE SITUATION IN THE DEVELOPING COUNTRIES

In those developing countries where petroleum reserves are strictly limited so that demands for motor fuel and black oil are met primarily by imports, the prospects for the production of liquid fuels from coal are attractive, provided that large deposits of coal are available. Even so, however, the economic resources of these countries are so limited that the building of coal-hydrogenation plants would create serious difficulties, namely: the necessity for large extraordinary capital expenditure, assimilation of complex technology, dependence on the importation of the main equipment, and a shortage of skilled personnel.

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Chapter 2

DISCUSSION OF COAL HYDROGENATION PROCESSES

Some of the processes developed in the USSR and in the United States for obtaining liquid fuels from coal and other solid fossil fuels are discussed in detail below.

SOLVENT EXTRACTION

At the end of the 1930s and at the beginning of the 1940s investigations were carried out in the USSR on developing a technology for obtaining liquid products from solid fuels by solvent extraction. The investigations were carried out on brown coals and coals of various ranks. Fractions of petroleum distillation, products of coal processing, individual chemical compounds and mixtures of compounds were used as solvents.

The process begins with a reaction of a solvent boiling at 200° to 350° C with a ground solid fuel (coal, peat, oil shale) for a period of 0.5—2 h. The solvent contains mainly hydrocarbons, (including hydrogenates of the solvent extraction products) as hydrogen donors. The reaction takes place under its own vapour pressure (30—40 atm) at a temperature of 380°—450° C. During the process, depolymerization, dissolution and cracking of the fuel take place with a simultaneous removal of oxygen, nitrogen and sulphur. The products resulting from the process are: light oil (200°—230° C), medium distillates (200°—350° C), pitch, unconverted organic component, water and gases (hydrocarbons, carbon monoxide and hydrogen sulphide).

During the thermal dissolution, 60—75 per cent of the organic component of brown coal is converted into light distillates, coke, gas and water. The same products are obtained when 50 per cent of the organic component of coal is converted. Hydrogenation of the extract results in a distillate boiling below 350° C, which is suitable for subsequent conversion into gasoline by the methods used in the petroleum industry. With a 1.8—2.0 per cent hydrogen consumption, the yield of distillate is 36 per cent.

Using hydro-treatment and destructive hydrogenation of this distillate, it is possible to obtain about a 40 per cent yield of automobile gasoline and diesel fuel (on a dry-coal basis with a consumption of 3 per cent hydrogen). The undissolved part of the coal (about 45 per cent) can be used as a high-energy fuel, and the gaseous products (C_1—C_4 hydrocarbons) can be used for hydrogen production.

In processing brown coal and oil shales, the motor-fuel yield can be considerably increased respectively to 50 and 60—65 per cent (based on the organic component).

TWO-STAGE HYDROGENATION OF COAL AT A PRESSURE OF 100—300 atm

In the 1950s a method of hydrogenation of low-rank coal, followed by a single-stage hydrogenation of liquid products in the vapour phase, was developed in the USSR. On the basis of the data obtained, a flow chart was worked out for obtaining both chemicals and fuel. Besides the preparation and production of hydrogen from the gaseous hydrocarbons obtained during the process, the following stages were foreseen:

Liquid-phase hydrogenation (300 atm, 460°—470° C, Fe-catalyst, cleaned long-flaming and gas coals); separation of phenols, nitrogen bases and hydrocarbons from the hydrogenates.

Hydro-treatment (at 400° C) of the hydrocarbon fraction of the hydrogenate boiling in the range of 50°—150° C; high-temperature hydrogenation of the hydrocarbon distillate boiling in the range of 150°—330° C (530°—550° C, 100 atm, stationary Co—Al—Mo-catalyst).

Hydrogen, the consumption of which is only 3—3.6 weight per cent of the organic component, is obtained from methane and (partly) ethane, which are formed in quantity during the coal hydrogenation, by catalytic conversion with water vapour.

Depending on the depth of processing and on the product mix, 45—50 per cent of the output consists of chemicals (aromatic hydrocarbons C_6—C_8, tetralin, naphthalene, an aromatized solvent for the varnish and dyestuff industry, phenols, nitrogen bases, ammonium sulphate), 5—10 per cent is the automobile gasoline component and 15—20 per cent consists of C_3—C_5 hydrocarbon gases.

An approximate technical and economic analysis of the coal processing scheme shows that for a plant processing 2 million tons of coal the cost of production is 16.5 per cent less than the cost of production of similar products in coking and petroleum processing.

A similar scheme may be used for brown coal processing.

HYDROGENATION OF A MIXTURE OF COAL AND PETROLEUM RESIDUES

In the 1960s a technology for hydrogenating a mixture of coal and petroleum residues containing large amounts of sulphur was developed in the USSR. Recommended raw materials are gas coals with low sulphur (0.3—0.5 per cent) and ash (3—5 per cent) content, and residues boiling above 240° C obtained from distillation of sulphur-bearing crude under atmospheric pressure. The use of petroleum residues for making the coal paste increases the degree of liquefaction of the coal and reduces the pressure of hydrogen from 300 to 100 atm and its consumption from 5–8 to 1–2 per cent. In addition, liquid-phase hydrogenation of such a mixture yields final products with a lower sulphur content compared with the higher fractions of petroleum. Hydrogenation of a 1:1 mixture is carried out at a temperature of 425° C, a hydrogen pressure of 100 atm, and in the presence of a catalyst ($Fe^{+3} + Mo$). The organic component is converted at the rate of 90 per cent for coal and 93 per cent for brown coal.

A typical percentage yield from hydrogenation of a mixture of Kuzbass gas coal and Arlan topped oil (40:60) under these conditions is:

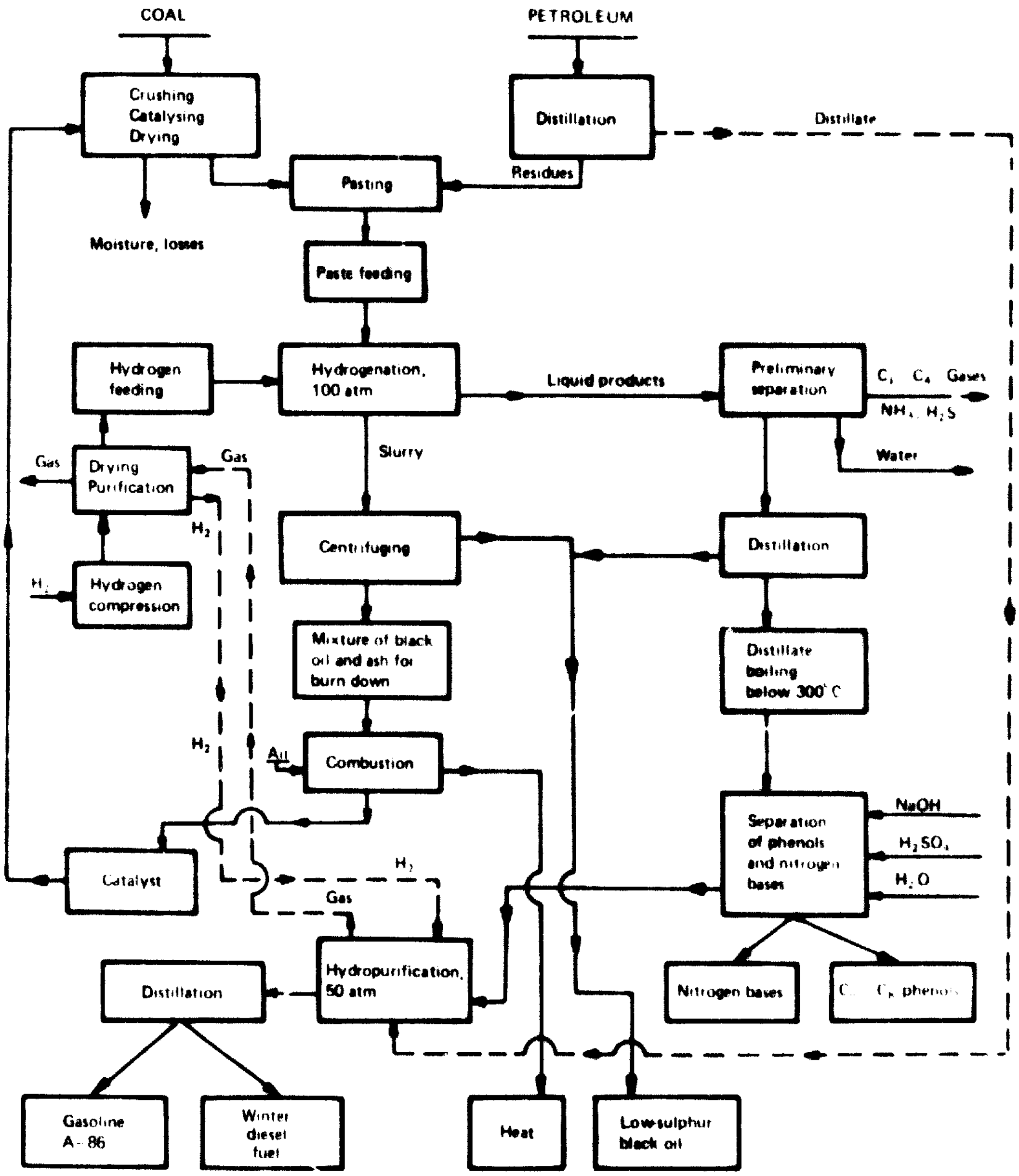


Figure 1. Liquid-phase hydrogenation of a mixture of coal and petroleum residues

black oil, 41; A-86 gasoline, 16; winter diesel fuel, 23; C_6—C_8 phenols, 1.6; hydrogen sulphide, 1; mixture of black oil and ash for burn down, 12; C_1—C_4 hydrocarbon gases, 6.

The black oil contains 0.5—0.8 per cent sulphur. It can be used for the production of a low sulphur electrode coke, the yield of which during coking is 30—35 per cent. The gasoline meets the requirements for high-quality gasoline. The diesel fuel has a high cetane number (49—53) and a low freezing point (−26 °C). Some of its fractions can be used as components of aviation fuel. Sixty per cent of the phenols consists of scarce cresols. The flow chart for this process is shown in figure 1.

Capital costs for construction of a complex coal and petroleum enterprise (with a capacity of about 15 million tons per year) are 20 per cent lower than for construction of an enterprise intended for petroleum processing and production of analogous products.

PRODUCTION OF LIQUID FUEL BY THE "H-COAL" PROCESS

The "H-Coal" process developed in the United States is based on the classic Bergius process and permits the use of both coal and brown coal. The main apparatus is the reactor, where particles of a high-activity catalyst and coal suspension are retained in the fluidized bed. In the petroleum industry the reactor is used to process petroleum residues. The reaction takes place at 425°—480 °C and 210 atm; under these conditions the conversion of the organic component is 80—90 per cent.

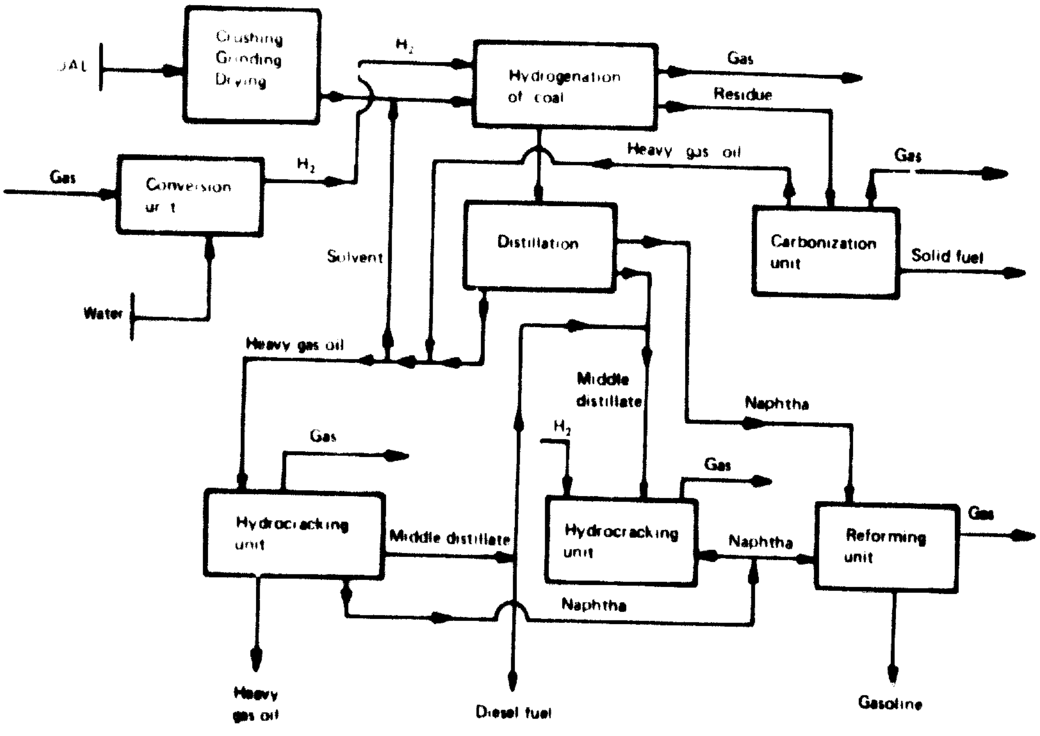


Figure 2. Production of synthetic fuel from coal by the "H-Coal" process

The main product is a light oil that can be converted into gasoline and other products.

The flow chart of the "H-Coal" process is given in figure 2. It was worked out in a plant processing 2 tons of coal per day, the investment cost of which was \$2.06 million. In order to study different technological versions of the process, a semi-commercial plant with a daily capacity of 250 tons of coal is planned.

According to the preliminary estimates the investment for a plant producing about 4,700 m^3/year of gasoline or gasoline and light oil (2:1 ratio) is \$115 to \$120 million. The estimated investment for a plant producing 15,900 m^3/year is \$318 to \$332 million.

PRODUCTION OF LIQUID FUEL BY THE CONSOL PROCESS

The Consol process (worked out in the United States) is a modification of the solvent extraction process in combination with hydrogenation of the extract and its utilization as a hydrogen donor for dissolving the coal.

Only the most reactive part of the organic component (60–85 per cent) is subjected to hydrogenation. This part is separated in the dissolution process and contains enough hydrogen to permit hydrogenation at a moderate pressure (200 atm). Distillation fractions obtained in the process are the feedstock for the hydrocracking units. The flow chart for the Consol process is given in figure 3.

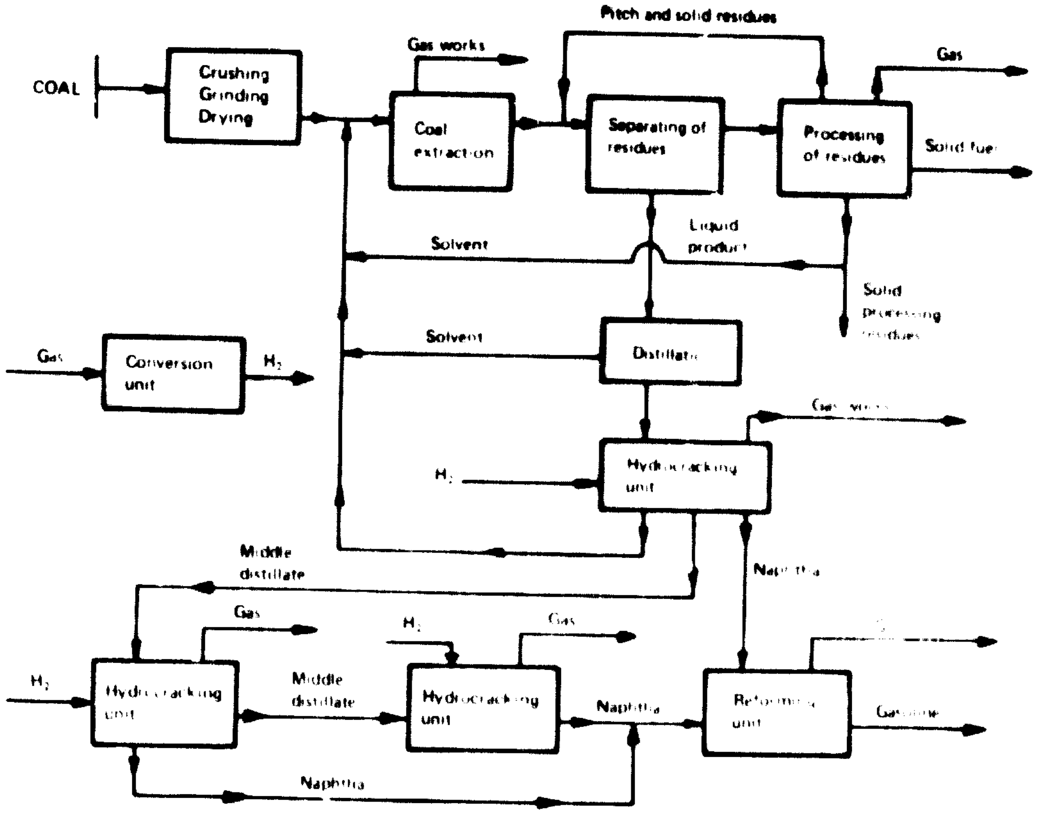


Figure 3. Production of synthetic fuel from coal by the Consol process

Consol has been tested in a pilot plant with a daily capacity of 24 tons of coal. The cost of the plant was $10.93 million.

The projected capacity of the first commercial plant for the production of liquid fuel by the Consol process is 7.5 million tons of coal per year. The investment has been estimated at $225 million.

PRODUCTION OF LIQUID FUEL BY THE COED-FMC PROCESS

The COED-FMC process of producing synthetic fuel, which differs considerably from the processes described above, was worked out in the United States. It is based on a four-stage low-temperature carbonization of coal. As a result only about one third of the organic component is converted into primary tar or light oil, which is hydrogenated subsequently. The process is carried out without a catalyst and at ambient pressure. In addition to the main product, crude oil, a large quantity of fine coke (1:2) is obtained during the process. The technological flow chart for this process is shown in figure 4.

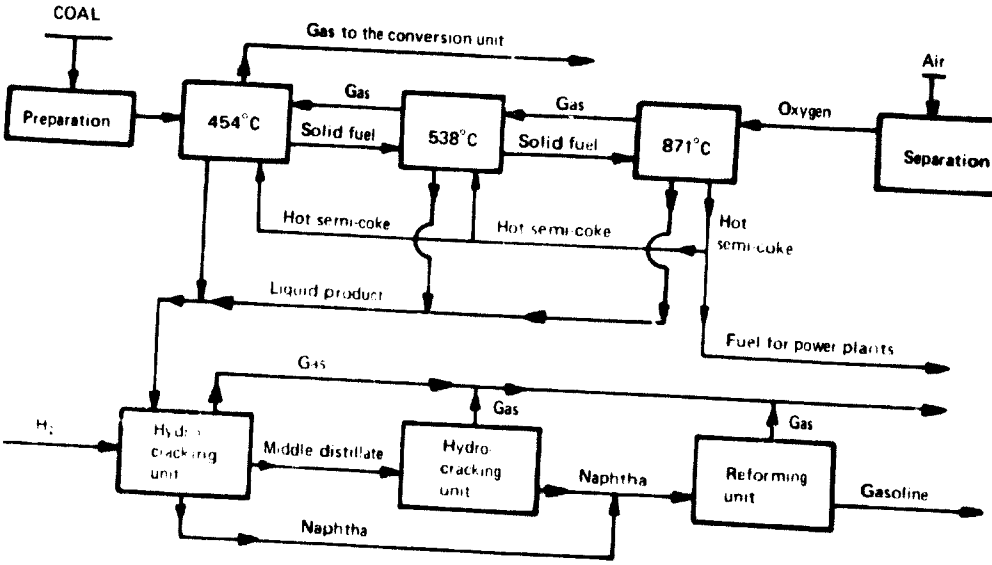


Figure 4. Production of synthetic fuel from coal by the COED process

It was planned that a pilot factory with a capacity of 3 tons of coal per hour would be built by the end of 1970.

On the basis of research, a factory processing 10,000 tons of coal per day should cost approximately $28 million.

COMPARISONS OF COAL HYDROGENATION PROCESSES

A proper comparison of the efficiency and cost of processing the hydrogenate by various methods can be made by studying the composition and properties of the products obtained from coal and those obtained from crude oil, as shown in tables 1 and 2.

Table 1

COMPOSITION OF THE PRODUCT OBTAINED FROM DIFFERENT PROCESSES

Fraction	*Proportion of total liquid product (weight per cent)*			
	"H-Coal"	*Consol*	*COED*	*Crude oil*
Gasoline (36° –204° C)	26.0	8.0	5.0	25—39
Heavy naphtha (204° –360° C)	38.8	92.0[a]	45.0	13—40
Gas oil (360° –524° C)	17.9	—	32.0	25—39
Black oil (524° C)	17.3	—	18.0	1—15

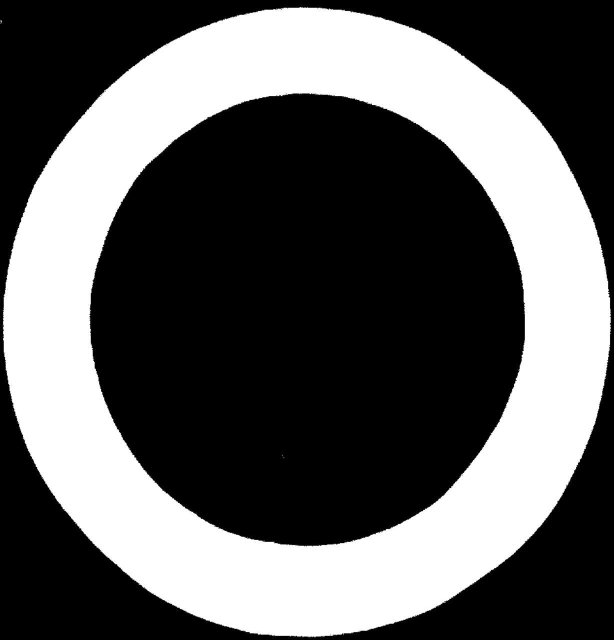
[a] Final boiling point 323° C.

Table 2

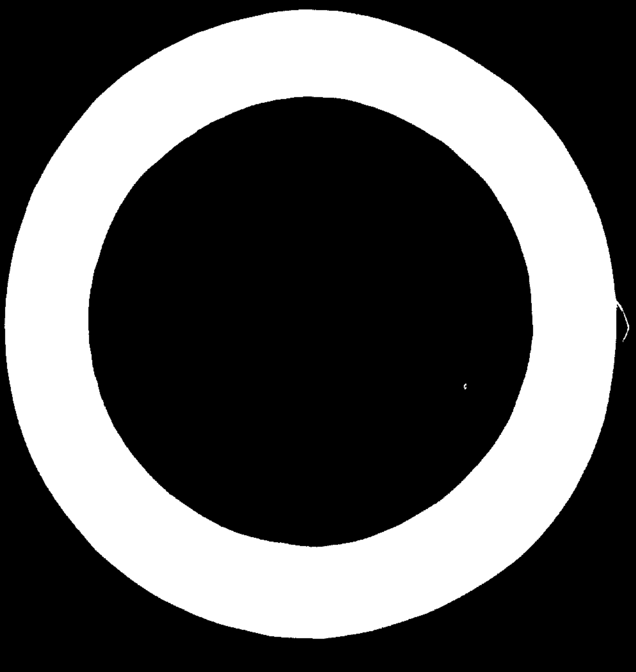
CARBON AND HYDROGEN CONTENT

Product	*Carbon (weight per cent)*	*Hydrogen (weight per cent)*
Hydrogenate from coal	88—93	5—8
Crude petroleum	84—88	11—14

The data in tables 1 and 2 show that the average boiling point of liquid fuels is higher, and the hydrogen content considerably lower, if they are obtained from coal rather than from crude petroleum. In order to obtain products of similar qualities, therefore, the hydrogenates must be subjected to additional thermocatalytic hydrogenation.



ANNOTATED BIBLIOGRAPHY OF WORKS PUBLISHED IN 1959—1970



1959

1. Krichko, A. A., A. V. Lozovoi and D. P. Pchelina (1959) **A new low-pressure hydrogenation process for semicoke-oven tars** (Novaya tekhnologicheskaya skhema gidrogenizacionnoi pererabotki polukoksovykh ugolnykh smol pod nevysokim davleniem) *Trudy Instituta Goryuchikh Iskopaemykh AN SSSR*, Vol. 9, p. 37. (in Russian)

   A simplified technological process for low pressure hydrogenation of primary coal tars is described. The process yields valuable chemical products and high-grade motor fuel.

2. Krichko, A. A. and R. A. Konyashina (1959) **Hydrogenation of coals mined in the Cheremkhovo coalfield and concentrated by the method of centrifugal separation in heavy liquids** (Issledovanie gidrogenizacii uglei Cheremkhovskogo mestorozhdeniya, obogashchyonnykh metodom tsentrobezhnoi separatsii v tyazhyolykh zhidkostyakh) *Trudy Instituta Goryuchikh Iskopaemych AN SSSR*, Vol. 9, p. 62. (in Russian)

   This article discusses the hydrogenation of Cheremkhovo coals in an autoclave at 460° C and 300 atm after a one-hour treatment by the method of centrifugal separation in heavy liquids.

3. Blonskaya, A. I., A. V. Lozovoi, M. G. Gonikberg *et al.* (1959) **Hydrogenation of lean coals and anthracites at hydrogen pressures exceeding 1,000 atm** (Issledovanie gidrogenizacii toshchikh uglei i antratsitov pri davlenii vodoroda vyshe 1,000 at) *Trudy Instituta Goryuchikh Iskopaemykh AN SSSR*, Vol. 9, p. 50. (in Russian)

   The behaviour of anthracite, semi-anthracite and lean coal during destructive hydrogenation in an autoclave at 480°—600° C and 1,200—1,700 atm.

4. Blonskaya, A. I., A. V. Lozovoi, D. L. Muselevich *et al.* (1959) **Two-stage process for production of chemical intermediates, motor oil and gases by hydrogenation of Cheremkhovo coal tar** (Dvukhstupenchataya skhema gidrogenizacionnogo proizvodstva khimicheskikh poluproduktov, motornogo topliva i gazov iz smoly cheremkhovskikh uglei) *Trudy Instituta Goryuchikh Iskopaemykh AN SSSR*, Vol. 9, p. 5. (in Russian)

   A simplified technological process for combined treatment of semi-coked coal tar by hydrogenation was worked out. Chemical intermediates, motor oil and hydrocarbon gases were the products.

5. Dyakova, M. K., A. B. Vol-Epstein, M. N. Zharova *et al.* (1959) **Conversion of higher phenols into phenols, cresols, and xylenols** (O prevrashchenii vysshikh fenolov v fenol, krezoly i ksilenoly) *Zhurnal prikladnoi khimii*, Vol. 32, No. 9, p. 2120. (in Russian)

   The origin of these higher phenols was Cheremkhovo coal tar.

6. Okladnikova, Z. A., A. S. Nakhmanovich, N. I. Shchergina (1959) **An infra-red spectroscopic investigation of the mechanism of transformation of the high-molecular fraction of semi-coked tar during destructive hydrogenation** (Issledovanie khimizma prevrashcheniy vysokomolekulyarnoi chasti polukoksovoi smoly v usloviyakh destruktivnoi gidrogenizacii pri pomoshchi infrakrasnoi spektroskopii) *Trudy Vostochno-Sibirskogo Filiala AN SSSR*, seriya khimicheskaya, No. 26, p. 39. (in Russian)

   Infra-red absorption spectra of neutral components separated from the pitch of Cheremkhovo semi-coked tar.

7. Davtyan, N. A., M. K. Dyakova (1959) **Research on the structure of the higher phenols obtained from Cheremkhovo coal tar** (Issledovanie struktury vysshikh fenolov gidrogenizata smoly cheremkhovskikh uglei) *Trudy Instituta Goryuchikh Iskopaemykh AN SSSR*, Vol. 9, p. 26. (in Russian)

   This article describes the research carried out on the composition and structure of phenols obtained from the heavy fraction boiling at 230°–310° C.

8. Pinchin, F. J. (1959) **The mechanism of coal hydrogenation** *British Coal Utilization Research Association* (BCURA), Monthly Bulletin, Vol. 23, No. 12, pp. 465–476. (in English)

   A review of data concerning the mechanism of the hydrogenation process as applied to improve the coking qualities of coal, to convert coal into liquid, to produce partial gasification of coal and hydrogenation of the resulting organic compounds.

9. Cockram, C. and E. W. Sawyer (1959) **Hydrogenation at Billingham in Retrospect** *Industrial Chemist*, Vol. 35, No. 411, pp. 221–229. (in English)

   Tracing the development of hydrogenation processes at the Billingham coal-tar factory, the article reviews the processes that the factory employed for production of synthetic liquid fuel from coal and creosote oil. The article also describes the major plants and their capacities, as well as the composition of the products.

10. Kawa, W. *et al.* (1959) **Agglomeration studies in the low-pressure hydrogenation of coal in a fluidized bed** *U.S. Bureau of Mines Bulletin, No. 579*, 11 pp., Washington, D.C. (in English)

    Equipment and procedures for agglomeration studies. Reduction and elimination of agglomerates by slow heating or by coal pretreatment at 400° C with or without hydrogen. To maximize the yield of synthetic liquid fuels, the time spent at 400—500° C must be sufficiently long.

11. **An American coal-hydrogenation plant** (1959) *Chemical Products*, Vol. 22, No. 7, pp. 271—272. (in English)

    A pilot plant for coal hydrogenation is described and a simplified flow chart presented. The plant has a capacity of 100 tons of coal per day. Operated in West Virginia by the Union Carbide Corp., this plant operates at 450° C to 550° C to produce phenol, cresols, aniline, quinoline, naphthalene and toluene.

12. Keith, P. C. and F. Ringer (1959) **Coal hydrogenation,** *U.S. Pat. No. 2,885,537;* 5 May. (in English)

    The suggested method combines coal hydrogenation with subsequent product cracking in a fluidized bed and gasification of the residual products by vapour-and-oxygen blast. Coal paste is hydrogenated at a temperature of 351° C and a pressure of 70 atm, and 80—85% of the coal organic mass is converted into liquid and gas products. The flow diagram is presented and the concept of the basic apparatus is described.

13. Pelipetz, M. G. and R. A. Friedel (1959) **Hydrogenation products from bituminous coal and sucrose at elevated temperatures; spectral comparison,** *Fuel*, Vol. 38, No. 1, pp. 8—16. (in English)

    Nearly identical products were obtained by similar hydrogen treatments of bituminous coal and sucrose.

14. Švajgl, O. (1959) **Disturbing influence of arsenic in high-pressure hydrogenation of brown-coal tars** (Rušivý arsenu pri vysokotlaké hydrogenaci hnedouhelnych dehtů) *Chemicky Prümysl*, Vol. 9, No. 5, pp. 230—234. (in Czech)

    Peculiarities of the hydrogenation process as applied to Czechoslovakian brown-coal tars. A high content of arsenic passes from coal into tar during low-temperature carbonization in the presence of a fixed-bed catalyst. Methods for eliminating the arsenic are suggested.

15. Švajgl, O. (1959) **Deactivation of WS_2—NiS—Al_2O_3 catalyst by arsenic. 1. Mechanism of deactivation** (Entaktivierung des WS_2—NiS—Al_2O_3-Katalysators durch Arsen. 1. Mechanismus der Entaktivierung) *Collection of Czechoslovak Chemical Communications*, Vol. 24, No. 12, pp. 3829—3835. (in German)

    Deactivation of WS_2—NiS—Al_2O_3 catalyst by arsenic contained in the hydrogenation distillates of brown-coal tar. When absorbed by the catalyst, arsenic reacts with the nickel sulphide and reduces the amount of active sulphide, thus lowering the activity of the catalyst as a whole.

16. Schnabel, B. (1959) **Low-temperature hydrogenation of Northern-Bohemian brown-coal tars** (Nízkotepelná hydrogenace severo-českých hnědouhelných dehtů) *Chemický průmysl*, Vol. 9, No. 1, pp. 10–14. (in Czech)

    A continuous process of low-temperature hydrogenation of brown coal tars was tested on a small-scale apparatus. The aim was to obtain motor fuels, *n*-paraffins, lubricating oils, phenols and cresols.

17. Bose, S. K., A. K. Ganguli, N. G. Basak, A. Lahiri (1959) **Processing of low-temperature tar oil fractions to diesel oils.** *Journal of the Institute of Petroleum*, Vol. 45, No. 428, pp. 252–258. (in English)

    The article contains sketches of experimental equipment and describes the development of an efficient catalyst for Diesel oil production by hydrogenation of tar obtained from Raniganj coal. The characteristics of the tar are given, and the products of hydrogenation for different experimental conditions and for various catalysts are analysed.

18. Sharkey, A. G. *et al.* (1959) **Analysis of liquid products from coal hydrogenation by mass spectrometry.** *Fuel*, Vol. 38, No. 3, pp. 315 to 328. (in English)

19. Fowler, W. A. (1959) **Current trends in the American coal tar industry.** *Industrial Chemist*, Vol. 35, No. 409, pp. 120–123. (in English)

    Tar production on fluid carbonization plants. New products of coal chemistry: high-boiling aromatic hydrocarbons, phenols etc. Prospects for the production of chemicals by coal hydrogenation and hydrogen production from coke-oven gas.

20. Yamada, M. (1958, 1959) **Catalytic reduction of tar components by molybdenum trisulfide. II. Bicyclic hydrocarbons (diphenyl). III. Monocyclic phenols. IV. Condensed-nucleus phenols (α- and β-naphthols)** *Coal Tar* (Koru Taru), Vol. 10, No. 12, p. 727; Vol. 11, No. 1, p. 17. (in Japanese)

    II. Experimental data are presented on the effects of pressure, temperature and residence time on product yields in diphenyl hydrogenation in the presence of MoO_3. Up to 40% of phenyl cyclohexane can be obtained when the initial pressure of hydrogen is 90 atm and the temperature equals 350° C, 400° C or 450° C. Dicyclohexyl (3.5–5.0 per cent) and negligible quantities of C_6H_6 and C_6H_{12} were identified in the products.

    III. Hydrogenation of phenol, *o*-, *m*-, *p*-cresols, *m*-ethyl-phenol and 1,3,5-xylenol. It was demonstrated that an initial hydrogen pressure of 100 atm and temperature of 350° C (320° C for 1,3,5-xylenole) converts most of the phenol. The value of the initial hy drogen pressure notably influences the reaction rate at 300 °C, while a further rise of temperature to 350 °C and 400 °C reduces this influence.

IV. Experimental data are presented on the effects of pressure and temperature on product yields in hydrogenation of α and β naphthols (naphthalene, tetralin, decalin).

1960

21. Schoenemann, K. (1960) **The German contribution to the present situation of chemical technology in the world** (La contribution allemande à la situation actuelle de la technologie chimique dans le monde) *Génie chimique*, Vol. 83, No. 6, pp. 161—169. (in French)

    Achievements of German researchers in ammonia synthesis, coal hydrogenation etc.

22. Krichko, A. A., A. V. Lozovoi, D. P. Pchelina *et al.* (1960) **Production of chemicals from unpyrolyzed tar derived from continuous carbonization of Kuznetsk coals** (Poluchenie khimicheskikh produktov iz nepirolizovannoi smoly protsessa nepreryvnogo koksovaniya kuznetskikh uglei) *Izvestiya Sibirskogo Otdeleniya AN SSSR*, No. 12, p. 88. (in Russian)

    The article gives the characteristics of the investigated tar, describes how it is processed, and shows what yields of aromatic hydrocarbons, solvents and other products can be obtained from it by hydrogenation.

23. Salimgareeva, F. G., B. V. Przhtsinskaya and I. V. Kalechits (1960) **The formation of lower phenols during liquid-phase hydrogenation** (O poluchenii nizshikh fenolov v khode zhidkotaznoi gidrogenizacii) *Metallurgicheskaya i khimicheskaya promyshlennost Kazakhstana*, No. 2, p. 102. (in Russian)

    Semi-coked tar from Cheremkhovo coal was used.

24. Lewis, P. S. and R. W. Hoteshue (1960) **Hydrogenating coal in the entrained state** *Industrial and Engineering Chemistry*, Vol. 52, No. 11, pp. 919—920. (in English)

    The article describes an apparatus for hydrocarbon production by coal hydrogenation at temperatures of up to 800 °C and pressures of up to 420 atm. This apparatus could be employed for other processes taking place under similar conditions. Design of certain components is presented, e.g. the feeder for delivery of powdered coal in the entrained state.

25. Lang, E. W. and J. C. Lacey (1960) **Properties of tar from low-temperature carbonization of American seam coal** *Industrial and Engineering Chemistry*, Vol. 52, No. 2, pp. 137—140. (in English)

    Large quantities of low-cost char and tar may be produced by fluidized carbonization of coal, and economical applications for the tar are sought. The tar can be hydrogenated to obtain low-boiling products. Hydrocracking and reforming of tar distillates yield lower aromatics. The tar could possibly prove usable in plastic engineering. Economic aspects of tar processing are discussed.

26. Dawydoff, W. (1960) **Hydrogenation in a homogenous phase** (Hydrierung in homogener Phase) *Chemische Technik*, Vol. 12, No. 7, pp. 414—418. (in German)

    Single- and double-stage hydrogenation of brown-coal tars obtained by low-temperature carbonization was used for production of C_8-alcohols. The properties of the resulting alcohols are described

27. Lüder, H. and K. Drescher (1960) **Testing of rhenium-containing catalysts for medium and high pressure hydrogenation of brown-coal tar processing products** (Ausprüfung rheniumhaltiger Katalysatoren für die Mittel- und Hochdruckhydrierung von Braunkohlenteerverarbeitungsprodukten) *Chemische Technik*, Vol. 12, No. 1, pp. 16 to 22. (in German)

    Rhenium catalysts on various carriers were tested in a number of reactions and were found useless for the hydrogenation of hydrocarbons boiling within the range of gasoline; however, these catalysts could be efficiently employed as substitutes for tungsten and molybdenum for hydrogenation of brown-coal middle oils under medium and high pressures.

28. Hiteshue, R. W., R. B. Anderson, S. Friedman (1960) **Gaseous hydrocarbons by hydrogenation of coals and chars** *Industrial and Engineering Chemistry*, Vol. 52, No. 7, pp. 577—579. (in English)

    The article contains the results of an experimental investigation of the possibility of commercial production of hydrocarbon gases by hydrogenation of coal and chars at 420 atm and 800° C in the presence of cobalt molybdate. The data obtained can be used for designing a continuous bench-scale hydrogenation unit.

1961

29. Katsobashvili, Ya. R., Yu. N. Garber, E. I. Elbert and Z. G. Belenko (1961) **Hydrocracking of high-boiling coal tar fractions in a fixed bed catalyst at a pressure of 30 atm** (Gidrokreking vysokokipyashchikh

fraktsiy kamennougolnoi smoly v statsionarnom sloe katalizatora pod davleniem 30 at) *Koks i khimiya*, No. 10, p. 48. (in Russian)

The hydrocracking was performed in a continuous laboratory plant with a reactor capacity of 250 ml in the presence of $CoO + MoO_3 + Al_2O_3$ catalyst. The processing of crude anthracene, anthracene fraction and absorbent oil with a diluent at 540°—560° C was studied. The naphthalene fraction yield was 65%; it contained 70—75% naphthalene.

30. Borts, A. G., A. A. Krichko, R. A. Konyashina *et al.* (1961) **Processing of an anthracene fraction by the hydrogenation method** (Pererabotka antratsenovoi fraktsii metodom gidrogenizacii) *Koks i khimiya*, No. 10, p. 53. (in Russian)

Research on destructive hydrogenation of the coal-tar anthracene fraction using molybdena-cobalt-alumina or molybdena-alumina catalysts at 100—200 atm and 520°—550° C.

31. Tyutyunnikov, Yu. B. and Yu. M. Volkov (1961) **New prospects for the chemical treatment of bituminous coals** (Novye perspektivnye napravleniya khimicheskoi pererabotki kamennykh uglei) *Nauchnye trudy Ukrainskogo uglekhimicheskogo instituta* (Sbornik), No. 12, p. 81. (in Russian)

A brief survey of methods intended for chemical treatment of bituminous coals by hydrogenation, oxidation and alkali pyrolysis. Long-flaming and gas coals were investigated.

32. Salimgareeva, F. G., M. F. Ivanova, B. V. Przhtsinskaya, I. V. Kalechits (1961) **Transformation of carbonyl compounds during destructive hydrogenation** (O prevrashcheniyakh karbonilnykh soedineniy v usloviyakh destruktivnoi gidrogenizacii) *Izvestiya Sibirskogo otdeleniya AN SSSR*, No. 5, p. 115. (in Russian)

This article contains data on hydrogenation of benzophenone, benzaldehyde, cyclohexanone, semi-coked tars and coals in the presence of industrial ferric catalysts.

33. Kalechits, I. V., Z. A. Okladnikova, D. Kh. Nikolaeva (1961) **The relative rates of hydrogenation of polycyclic aromatic hydrocarbons** (K voprosu ob otnositelnykh skorostyakh gidrogenizacii politsiklicheskikh aromaticheskikh uglevodorodov) *Trudy Vostochno-Sibirskogo Filiala AN SSSR*, No. 38, p. 112. (in Russian)

The relative rates were measured for the hydrogenation of diphenyl, naphthalene, anthracene, phenanthrene, chrysene, pyrene and caronene in the presence of a skeleton nickel catalyst and industrial ferric catalyst.

34. Sidorov, R. I., Z. P. Trotsenko, A. S. Nakhmanovich (1961) **Composition of industrial liquid-phase hydrogenates. Communication 5. Research on the composition of the mixtures of aromatic hydro-**

carbons of the liquid-phase hydrogenates obtained from medium-temperature Cheremkhovo coal tar (Issledovanie sostava promyshlennykh zhidkofaznykh gidrogenizatov. Soobshchenie 5. Issledovanie sostava smesei aromaticheskikh uglevodorodov zhidkofaznogo gidrogenizata tyazhyologo masla srednetemperaturnoi smoly cheremkhovskogo uglya) *Trudy Vostochno-Sibirskogo Filiala AN SSSR, khimicheskaya seriya*, No. 38, p. 68. (in Russian)

The mixtures contain compounds belonging to the homologous groups of benzene, indan, tetralin and naphthalene.

35. Günther, G. (1961) **Studies of the kinetics of hydrogenation reactions. Hydrogenation of cresol** (Untersuchungen zur Kinetik von Hydrierreaktionen. Die Hydrierung von Kresol) *Chemische Technik*, Vol. 13, No. 12, pp. 720–723. (in German)

Research on the effects of temperature, pressure, residence time, gas-to-oil ratio, and cresol concentration in the original product on the process of high-pressure hydrogenation of tars and oils in the presence of solid catalysts.

36. **Coal holds jet fuel raw material potential** (1961) *Chemical and Engineering News*, Vol. 39, No. 43, pp. 56–57, (in English)

The article contains a flow diagram and a brief description of a process of high-energy jet fuel production by hydrogenation of a coal tar, obtained by high-temperature coking, in the presence of sulphur-resistant catalysts (tungsten and nickel sulphides on alumina or molybdenum sulphide on activated carbon). The process was developed in France. It works best at temperatures of 420°–430° C and hydrogen pressures of 200–250 atm.

37. Feldkirchner, H. L., E. J. Pyrcioch, E. B. Shultz (1961) **Hydrogenation techniques at combined high temperatures and pressures**, *Chemical Engineering Progress. Symposium Series*, Vol. 57, No. 34, pp. 73–80. (in English)

Batch and semicontinuous reactors for hydrogasification of solid fuels and a continuous tubular reactor for hydrogasification of crude oil and tars are described.

38. Hara, H., H. Kudo and M. Kiyoshi (1961) **Hydrogenolysis of coal**, *Journal of the Fuel Society of Japan* (Nenryo Kyokaishi), Vol. 40, No. 411, pp. 545–555. (in Japanese and English)

Investigations show that the speed of hydrogenolysis of coal depends upon the speed of solution of hydrogen in the liquid phase and the number of collisions between hydrogen and coal particles, which is a function of the paste viscosity. It was found that the paste should be pretreated at 310°–360° C for 2–3 h, and the oil-to-coal ratio in the paste should be 1.5 to 2.3:1. The reaction should be carried out at the highest possible temperature and pressure. Minimum density oils are recommended for the paste formulation.

39. Matsuda, T. (1961) **Problems involved in the industrialization of coal hydrogenation processes,** *Journal of the Fuel Society of Japan* (Nenryo Kyokaishi), Vol. 40, No. 411, pp. 531—534. (in Japanese and English)

Experimental data serve as the basis for a discussion of prospects of commercial application of coal-hydrogenation processes composed of two stages, coal liquefaction and liquefied oil reforming. An analysis of production economy is made, assuming that the plant capacity is 1,000 tons of coal per day.

40. Sakabe, T., Y. Ogo, R. Sassa, M. Suzuki, M. Horie, Y. Kanbayashi, T. Ohissa, M. Takahashi, M. Hunsaki (1961) **Liquid-phase coal hydrogenation in an experimental flow plant,** *Journal of the Fuel Society of Japan* (Nenryo Kyokaishi), Vol. 40, No. 411, pp. 535—544. (in Japanese and English)

The test plant for continuous coal hydrogenation under a pressure of 600 atm had a capacity of 50 kg/day. It was used to process Miike coal with Bayer mass catalyst. The following results were obtained (percentages): coal conversion factor — 96.9: yield of light oil (b.p. below 300° C) — 64.1; yield of gas — 25.8; hydrogen consumption — 7.6.

1962

41. Blonskaya, A. I. and A. V. Lozovoi (1962) **Composition of the hydroaromatizate from Cheremkhovo semi-coked coal tar** (O sostave gidroaromatizata iz polukoksovoi smoly Cheremkhovskikh uglei) *Trudy Instituta Goryuchikh Iskopaemykh AN SSSR*, Vol. 17, pp. 187—198. (in Russian)

The composition of the hydroaromatizate resulting from the two-stage hydrogenation of Cheremkhovo semi-coked coal tar was studied. The benzene fraction (60°—205° C) contained: 52.6% aromatic, 4.4% unsaturated, 34.5% naphthene, 8.5% paraffin hydrocarbons. The fraction boiling at 190°—300° C contained: 73.1% aromatic, 5.3% unsaturated, 7.2% normal paraffin, 12.8% naphthene and isoparaffin hydrocarbons.

42. Katsobashvili, Ya. R., Yu. N. Garber, E. I. Elbert, Z. G. Belenko. A. G. Borts (1962) **Method of processing high-boiling coal-tar**

fractions (Sposob pererabotki vysokokipyashchikh fraktsiy kamennougolnoi smoly) *Avtorskoe svidetelstvo SSSR (Russian patent) 143786*, 21 March. (in Russian)

A method is described for processing high-boiling coal-tar fractions into low-boiling aromatic hydrocarbons. The raw material was subjected to hydrocracking at 10—50 atm and 450°—700° C using active cobalt-alumina catalysts in the presence of a liquified gas (propane, butane etc.) followed by a recycling of the unreacted residue. A table of optimum high-boiling cracking conditions for various raw materials is given.

43. Katsobashvili, Ya. R., Yu. N. Garber, E. I. Elbert, Z. G. Belenko (1962) **Method of processing high-boiling coal-tar fractions** (Sposob pererabotki vysokokipyashchikh fraktsiy kamennougolnoi smoly) *Avtorskoe svidetelstvo SSSR (Russian patent) 145561*, 21 March. (in Russian)

This method differs from the one described in Ref. No. 42 above only in that the whole process is carried out at the lower temperature of 300°—350° C. The aim is to obtain the low-boiling hydroaromatic hydrocarbons, which are used as raw materials for organic synthesis, production of plastics etc.

44. Katsobashvili, Ya. R., Yu. N. Garber, E. I. Elbert, A. A. Lukanin (1962) **Coal-tar processing method** (Sposob pererabotki smol) *Avtorskoe svidetelstvo SSSR (Russian patent) 148038*, 21 June. (in Russian)

A no-residue process for converting coal tar into aromatic and hydroaromatic hydrocarbons is proposed. A destructive hydrogenation in a fixed bed of highly active catalysts at 30—50 atm is followed by a separation of hydrocarbons into different fractions by means of a rectifying column.

45. Lozovoi, A. V., D. L. Muselevich, T. M. Ravikovich, T. A. Titova, V. F. Cherkasova (1962) **Two-stage process for production of chemicals by hydrogenation of Cheremkhovo coal tar** (Dvukhstupenchataya skhema gidrogenizacionnogo proizvodstva khimicheskikh produktov iz smoly cheremkhovskikh uglei) *Trudy Instituta Goryuchikh Iskopaemykh AN SSSR*, Vol. 17, pp. 174—181. (in Russian)

A proposed two-stage (liquid- and vapour-phase) process yields 60—66% chemicals and intermediates (aromatics and C_6—C_8 phenols, naphthalene, monomethyl naphthalenes, diluents etc.) and 33—37% hydrocarbon gases C_nH_{2n+2} at a hydrogen expenditure of 5.7—6.0% of the coal-tar weight.

Borts, A. G., A. A. Krichko, R. A. Konyashina, A. V. Lozovoi, L. N. Lvova (1962) **Production of chemicals from the anthracene**

fraction of coal tar by high-temperature hydrogenation (Poluchenie khimicheskikh produktov iz antratsenovoi fraktsii koksovoi smoly metodom vysokotemperaturnoi gidrogenizacii) *Trudy Instituta Goryuchikh Iskopaemykh AN SSSR*, Vol. 17, pp. 250–261. (in Russian)

A study of the hydrogenation in a continuous plant of the first anthracene fraction crystallized from coal tar. A flow chart is proposed for the production of solvents, C_6–C_8 aromatics, naphthalene and other products.

47. Lozovoi, A. V., D. L. Muselevich, T. M. Ravikovich, S. A. Senyavin, T. A. Titova, V. F. Cherkasova (1962) **Silica-alumina catalysts for high-pressure hydrogenation** (Katalizatory s alyumosilikatnoi osnovoi dlya gidrogenizacii pod vysokim davleniem vodoroda) *Trudy Instituta Goryuchikh Iskopaemykh AN SSSR*, Vol. 17, pp. 199–211. (in Russian)

The activity of 13 new catalysts intended for vapour-phase hydrogenation of partially dephenolized liquid-phase hydrogenate (of Cheremkhovo semicoke-oven tar) was tested in a continuous laboratory plant. The catalysts were prepared from a natural silica-alumina-askanite by activation with 40% HF plus 2.0 to 7.6 wt.% of Cr, Mo, Ni, Fe, Zn, W oxides or sulphides.

48. Švajgl, O. (1962) **Method of hydrogenating hydrocarbon-nonhydrocarbon mixture** (Způsob hydroganace uhlovodíků ve smesi s neuhlovodíkovymi příměsmi) CSSR, No. 103, 170; publ. 15 March. (in Czech)

A method is suggested for the hydrogenation of hydrocarbons mixed with nonhydrocarbon compounds, for example, phenols, nitrogen bases or sulphur compounds. The catalysts contain tungsten and nickel sulphides and active alumina. The relative content of aromatic hydrocarbons in the product remains constant or increases, when the ratio of the processed material to the catalyst is changed from 0.5–0.9 to 1–10 kg/litre of the catalyst and the temperature is raised from 340°–380° C to 400°–450° C. All non hydrocarbon admixtures and olefins are thereby converted into saturated or aromatic hydrocarbons.

49. Letort, M. (1962) **High-energy fuels derived from coal** (Les carburants à haute énergie derivés de la houille) *Chimie et industrie*, Vol. 87, No. 3, pp. 371–387. (in French)

The proposal is to produce jet fuels by hydrogenation of coal tar fractions under high hydrogen pressure. Hydrogenation processes in the presence of nickel and combined catalysts (tungsten and nickel sulphides on alumina) are compared and the advantages of the latter are shown. The characteristics of the raw materials and the physical and chemical properties of the fuels produced are presented, as well as a schematic diagram of an industrial installation.

50. Gorin, E. (1962) **Production of hydrogen-rich liquid fuels from coal,** *U.S. Pat. No. 3,018,241*, 23 January 1962. (in English)

A combination process for converting coal into materials suitable for gasoline feedstock. The process comprises the following operations:

1. Treatment with a solvent to dissolve 60—80 wt.% of the coal. Hydrocarbon solvents boiling at 260°—425° C are recommended. The temperature in the extraction zone should be higher (up to 500° C), but not high enough to cause carbonization.

2. Fractionation of the extract into: (*a*) a component rich in benzene-insoluble compounds, and (*b*) a component poor in these compounds and with a precipitant added. The liquid fractions are separated from the insoluble coal residue by means of continuous filtration or centrifugation.

3. Coking at 426°—760° C of the solid residue mixed with the first fraction (*a*).

4. Hydrogenation of a mixture of coke-oven distillate with the second fraction (*b*) on Mo and W catalysts at 410°—455° C and 140—225 atm.

51. Sakabe, T., Y. Ogo, R. Sassa, M. Suzuki, M. Horie, Y. Kambayashi, T. Ohisa, M. Takahashi, M. Hunaki (1962) **Liquid-phase hydrogenation of Miike coal by experimental flow plant. Experimental results with creosote as vehicle,** *Journal of the Chemical Society of Japan, Industrial Chemistry Section* (Kogyo Kagaku Zasshi), Vol. 65, No. 3, pp. 297—303, A21. (in Japanese)

The results of experiments on coal hydrogenation are reported. The pressure was 200—300 atm, temperature 440°—460° C, material feed rate 6 kg/h, hydrogen feed 5.5 Nm^3/h.

52. Münzig, E., H. Blume, E. Pindur (1962) **Research on the improvement of catalysts for hydrogenation of tars and medium oils** (Arbeiten zur Verbesserung der Katalysatoren für die hydrierende Raffination von Teeren und Mittelölen) *Zeitschrift für Chemie*, Vol. 2, No. 3, pp. 73—83. (in German)

A brief history of the development of hydrogenation catalysts. The composition and properties of catalysts 3510, 5058, 6434, 7846 and 8736 are given, as well as those of the new catalysts in use at a factory in Leuna (German Democratic Republic).

The specific features of the new catalysts are as follows: they are mainly oxides; Mo serves as the active component; synthetic materials are preferable for carriers; Co is partly substituted for Ni.

Practical applications of these catalysts are discussed. Highly acid clays are the best carriers for the 8376 and 8197 catalysts. When the raw material contains nitrogen bases an addition of H_3PO_4, HBO_3 and H_2SiO_3 to the carrier is recommended. Sulphonation of the hydrogenating catalysts prior to use is unnecessary.

53. Matsuda, S., Sh. Kirkawa, A. Uchida (1962) **Dealkylation of tar oil in the presence of an alumina catalyst. Effects of pressure and temperature,** *Journal of the Chemical Society of Japan, Industrial Chemistry Section* (Kogyo Kagaku Zasshi), Vol. 65, No. 4, pp. 568—574, A39—A40. (in Japanese)

Benzene, toluene, xylene and naphthalene were formed by the continuous hydrodealkylation of the methylnaphthalene fraction (b.p. 215°—260° C) from tar oil in the presence of Al_2O_3 at a temperature of 600°—650° C and a hydrogen pressure of 30—50 atm. The methylnaphthalene fraction yielded 70.2% liquid products (5.6% benzene, 6.1%, toluene, 38.3% naphthalene, 8.5% methylnaphthalenes, and 11.6% remainder), 12.6% CH_4, and 2.8% coke.

54. Schroeder, W. C. (1962) **Hydrogenation of coal,** *U.S. Pat. No. 3,030,297*, publ. 17 April 1962. (in English)

A hydrogenation process for the production of predominantly single-ring aromatic hydrocarbons. The feature of the process is rapid heating of a suspension of coal particles smaller than 100 mesh. The coal dust, suspended in a hydrogen stream at a pressure of 35—420 atm, is heated in less than 1 min from 300°C to 600° — 1,000° C and kept at that temperature for less than 1 min (2 to 20 sec). The reaction products are cooled rapidly. Ammonium molybdate catalyst (1%) is introduced into the coal prior to reaction. An example is cited in which 90% of the coal (C = 72.12%, H = 5.2%), treated at 800° C, 420 atm, with a flow rate of 2.8 m^3/h, was converted into fluids comprising 39—48% liquids and 32—40% gas. The gas contained 40 vol.% CH_4 and 5% C_2H_6.

55. Kanayama, H. Sh. Nakamura, M. Ogawa (1962) **Composition of udex extract produced from hydrogenate of coal tar light oil,** *Coal Tar* (Koru Taru), Vol. 14, No. 5, pp. 240—245. (in Japanese)

The solvent produced from hydrogenate of light oil and extracted by diethyleneglycol contains 30—40% aromatics, 60—70% hydrocarbons of the paraffin and naphthene series and is practically free of olefins. Chromatographic and spectral analyses show that the aromatics consist mainly of *m*- and *p*-xylenes, ethyltoluenes, and trimethylbenzenes. Higher-boiling aromatic compounds are also present in trace amounts. The paraffin content of the 50°—100° C fraction was determined.

56. Hill, G. R., L. B. Lyon (1962) **A new chemical structure for coal,** *Industrial and Engineering Chemistry*, Vol. 54, No. 6, pp. 36—41. (in English)

A new molecular model for high-volatile bituminous coal is proposed. The idea is that the yield of liquids from coal distillation depends mainly on the structure of the coal, particularly the functional groups, and the process used to convert the coal into liquids and coke. Since complete hydrogenation of coal is uneconomical, the authors suggest that the liquid products of low-temperature carbonization of coal be catalytically hydrogenated either in the presence of hydrogen donor materials or by the hydrocracking processes that Varga and Lozovoi developed for the production of gasoline and other liquids.

57. Coffman, J. A. (1962) **Corona processing under study,** *Chemical Engineering Progress*, Vol. 58, No. 7, p. 15. (in English)

A brief report on research and development in the US in the field of producing synthetic liquid fuels and chemical products by hydrogenation of powdered coal at 200° C in the field of an electrical corona.

58. **New uses for coal urged,** *Chemical Engineering Progress* (1962) Vol. 58, No. 7, pp. 11—17. (in English)

This article summarizes the papers read at a symposium, "New Uses for Coal Through Chemical Processing". Topics covered are: conversion of coal into acetylene by an electric discharge, development of fuels for aircraft and missiles, obtaining raw materials for chemical production from coal, ion-exchange resins, etc. Hydrogen content in coal may be increased and products of low molecular weight may be obtained under the effect of an electrical corona. A mixture of coal and oil heated to 200° C and immersed into an atmosphere of hydrogen, when subjected to corona action, has actually been converted into useful liquid products. The major agent was monoatomic hydrogen. The process might be adjusted to produce gas or stopped at an intermediate stage where the coal has been converted into a resin usable as a plastic moulding compound or in formulation of coatings.

59. Murray, J. V., J. D. Fales, M. A. Eccles (1962) **Hydrogenolysis of coal hydrogenation products,** *Austral.* 240,203, publ. 17 August. (in English)

The end result of coal hydrogenation is a mixture of liquid products. The suggestion here is to subject certain fractions of this mixture to hydrogenolysis without a catalyst at a temperature of 525° to 700° C, pressure of 150—250 atm, hydrogen supply of 4—20 mole/

mole, and residence time less than 15 min. The process is carried out continuously, for instance, in a tubular reactor consisting of 20 coil tubes of 6-mm diameter and 100-cm^3 capacity, these tubes being immersed in a lead bath.

60. Heider, M. (1962) **Liquid fuels made from brown coal** (Treibstoffgewinnung aus Braunkohle) *Betriebs-Ökonom*, Vol. 15, No. 8, pp. 383—388. (in German)

    A review of the properties of liquid fuels obtained from brown coal by low-temperature carbonization, hydrogenation, and Fischer-Tropsch synthesis.

61. Nishio, A. (1962) **Catalytic desulphurization of light distillate,** *Japanese Patent No. 13,173,* publ. 6 September. (in Japanese)

    Light distillate of coal tar was placed in contact with a V_2O_3-containing catalyst at ambient or lower pressure. The catalyst allows this pressure reduction and eliminates formation of adverse residues as compared with the application of H_2SO_4 or high-pressure hydrogenation. Example: raw benzene containing 0.0114% thiophene and 0.0120% CS_2 was passed over the catalyst (V_2O_3: :$Al_2O_3 = 1:9$) at 150 kg litre^{-1} h^{-1} at a temperature of 420° C and a hydrogen pressure equal to the ambient pressure. This purification removed 93% of the thiophene and 100% of the CS_2 from the product.

62. Klimke, R., J. Gondzik (1962) **New aspects in coal-tar processing** (Neue Gesichtspunkte der Teerverarbeitung) *Freiberger Forschungshefte*, A, No. 221, pp. 29—48. (in German)

    Methods for conversion of brown coal into paraffins, phenols, pyridine and electrode coke are described. The most efficient of these low-temperature hydrogenation methods are at a pressure of 40 atm, space velocity of 6 kg litre^{-1} h^{-1}, temperature of 330° C and H_2 consumption of 0.55 m^3/kg. A number of ideas for the most rational methods for complete conversion of various tars are presented.

63. Takeya, G. (1962) **Studies on coal hydrogenation process,** *Journal of the fuel society of Japan* (Nenryo Kyokaishi), Vol. 41, No. 421, pp. 466—477. (in Japanese)

    Coal hydrogenation was carried out in an autoclave and in a continuous apparatus, both in the laboratory and in pilot plants capable of treating 8 litres of coal paste per hour, with the aim of obtaining aromatic compounds. The kinetics of the process was studied by measuring the rate of decrease of the amount of coal which was benzene-insoluble and had not reacted. Product compositions and yields for various coals are presented.

1963

64. Egorova, O. I., L. K. Markov, V. I. Kasatochkin (1963) **Spectroscopic investigation of asphaltenes from coal hydrogenation** (Spektralnoe issledovanie asfaltenov gidrogenizacii kamennykh uglei) *Khimiya i tekhnologiya toplir i masel*, No. 5, pp. 31—34. (in Russian)

    X-ray and IR-spectra of asphaltenes obtained from the hydrogenation of Cheremkhovo fancy coal are reported. According to the spectroscopic and chemical data, hydrogenation is characterized as a process of thermal decomposition, in the presence of hydrogen, of the side radicals of a polymer, with the release of structural units such as asphaltene molecules. Hydrogen also helps prevent the re-polymerization of these structural units.

65. Zabramnyi, D. T., C. Nasritdinov (1963) **The chemical characteristics and structural-group composition of soluble hydrogenates of fusain microcomponents** (Khimicheskaya kharakteristika i strukturno-gruppovoi sostav rastvorimykh gidrogenizatov fyuzenirovannykh mikrokomponentov) *Izvestiya AN SSSR*, Otd. tekhn. nauk., Energetika i transport, No. 2, pp. 238—242. (in Russian)

    The fusain macromolecule has a mean molecular weight of 5234 and corresponds to the empirical formula $C_{350}H_{176}N_3S_2O_{47}$.

66. Taits, E. M., T. M. Bronovets, I. A. Andreeva (1963) **Extraction of plastic materials and binders from coals** (Poluchenie plastichnykh i svyazuyushchikh materialov iz iskopaemykh uglei) *Khimiya i tekhnologiya toplir i masel*, No. 2, p. 24. (in Russian)

    A moderate-temperature hydrogenation process for the preparation of products having valuable plastic and binding properties.

67. Shono, Sh., M. Yamada (1963) **Catalytic hydrogenation of coal-tar components by molybdenum sulphide** *Coal Tar* (Koru Taru), Vol. 15, No. 9, pp. 405—410. (in Japanese)

    Molybdenum sulphide is used widely in hydrogenation of coal-tar light oils. Good yields of benzene are obtained at temperatures below 250° C. Phenol, cresols and ethylphenol are reduced with a yield of up to 40%. The OH-group is eliminated from cyclohexane at 300°—320° C. When hydrogenated at a pressure of 100 atm and temperature of 270° C, α- and β-naphthols convert into tetralin. Hydrogenation of pyridine, carbazole, and quinoline occurs at 250°—450° C and 100 atm. Reduction of carbazole results in partially hydrogenated diphenylene tetrahydrocarbazole.

68. Tanaka, Sh., T. Matsui (1963) **1,1-Diphenylethane in coal hydrogenation products**, *Annual Reports of the Engineering Research Institute*,

Faculty of Engineering, University of Tokyo (Sogo Shikensho Nempo), Vol. 22, No. 1, pp. 15–19. (in Japanese)

An investigation of the neutral oil extracted from coal hydrogenation products, revealed the presence of a compound with a sharp absorption band at 14.3 μm in the infra red spectrum. By means of distillation, liquid chromatography, and determination of physical constants, this compound was identified as 1,1-diphenyl ethane. Its content in the neutral oil was about 5%.

69. Landa, S., M. Urban (1963) **Hydrogenation, chromatography and thermodiffusion of electrostatically purified low-temperature tar** (Über die Hydrierung, Chromatographie und Thermodiffusion des elektrostatisch gereinigten Tieftemperaturteers) *Brennstoff Chemie*, Vol. 44, No. 12, pp. 377–382. (in German)

The primary tar was extracted in an electrostatic tar extractor during low temperature carbonization in Lurgi ovens. The investigation concerned autoclave hydrogenation at 180–350 °C in the presence of MoS_2 catalyst. Both the primary tar and tar free asphaltenes, phenols, acids and bases were tested. The hydrogenation of raw tar and introduction of H_2 began at 220 °C. The H_2 flow rate was 4 litres per 100 g at this temperature. As the temperature rose to 320 °C and 350 °C the H_2 flow increased to 31 and 42 litres, respectively, while the phenol constituent of the distillate dropped to 18.6 and 12.8%, respectively. About 81% of the hydrogenate was gasified at temperatures not exceeding 360 °C.

70. Overholt, D. C., G. D. Roy, R. R. Warren (1963) **Refining of coal hydrogenation product** *U.S. Pat. No. 3,084,118*, Appl. 10 August 1959, publ. 2 April 1963. (in English)

One hundred parts of the hydrogenate are joined with 50–100 parts of an aromatic hydrocarbon and 2–5 parts of a coagulant, such as H_2SO_4. The top and the bottom layers are processed separately.

71. Sakabe, Ts., Y. Ogo, R. Sassa, M. Horie *et al.* (1963) **Liquid-phase hydrogenation of Miike coal by experimental flow plant. Results of runs with heavy oil recovered from product as vehicle,** *Journal of the Chemical Society of Japan, Industrial Chemistry Section* (Kogyo Kagaku Zasshi), Vol. 66, No. 6, pp. 735–746, A49. (in Japanese)

Hydrogenation was performed at a pressure of 300 atm with Bayer mass catalyst. Detailed data on the yield of products, material balances, H_2 consumption, sulphur balances etc., are given.

72. **ACS symposia show coal still has many uses** (1963) *British Chemical Engineering*, Vol. 8, No. 6, pp. 417–418 (in English)

Brief abstracts of papers on preparation of humic acids, manufacture of phthalic anhydride, hydrogenation of shale oil, upgrading iron concentrate etc.

73. Letort, M., A. F. Boyer, P. Payen (1963) **A study of coal hydrogenation by atomic hydrogen** (Essai d'hydrogénation d'un charbon par l'hydrogène atomique) *Bulletin de la Société chimique de France*, No. 8–9, pp. 1589–1593. (in French)

The atomic hydrogen was produced by an electrical discharge acting on a flow of 3–9 litres per hour of H_2 through a glass pipe of 2 m length. The current was 0.1 A; the voltage ~ 9 kV; system pressure 0.4–0.7 atm. The resulting mixture of H_2 and H, which contained up to 45–50% of H, was passed directly through a pipe 15 cm long along with the substance under test. When coal was treated, the resulting gas consisted of CH_4, C_2H_4 and CO. A similar gas was produced by vitrinite, its pyridine extract and coronene. Paraffins, phenantrene and pyrene produced gas of the same composition plus products of high molecular weight. After treatment, the solubility of vitrinite in pyridine increased from 18.8 to 29.6% and its sintering ability was 2–5 times greater. Addition of about 3% H_2O to the H_2 accelerated hydrogenation and facilitated CO formation.

74. Huntington, M. G. (1963) **Continuous distillation of condensable volatiles** *U.S. Pat. No. 3,107,985*. Appl. 8 July 1960, publ. 22 October 1963. (in English)

A system of coal processing that includes low temperature carbonization, hydrogenation of the volatiles and gasification of the char is described. A single vertical reactor is used for the process.

75. Perry, H., M. A. Elliott, M. R. Linden (1963) **Techniques for the conversion of coal into liquid and gaseous fuels in the United States** (La tecnica della conversione del carbone in combustibili fluidi negli Stati Uniti) *Gas*, Vol. 13, No. 9, pp. 240–248. (in Italian)

Coal gasification and purification of the gas. Production of city gas of high calorific value. Hydrogasification. Fischer-Tropsch process for liquid fuel synthesis. Coal hydrogenation and low-temperature distillation.

76. Howell, J. H., E. W. Doughty, P. L. Alspaugh (1963) **Coal hydrogenation process.** *Austral. 242,754*, Appl. 22 October 1959, publ. 16 October 1963. (in English)

The process features medium temperatures and pressures in combination with low space velocities. Coal particles smaller than 2 mm are mixed with oil to make a paste with coal content of 40–70% by weight. Hydrogen is passed through the paste at the rate of 2.5 kg litre^{-1} h^{-1}. In the reaction zone the pressure is 175–840 atm and the temperature is 490–560°C. The reactor used for the process consists of a large number of horizontal or vertical pipes.

77. **Electrochemical hydrogenation of coal** (1963) *Gas World*, Vol. 158, No. 4137, p. 690. (in English)

    Information on experimental work at the U.S. Bureau of Mines. Liquid fuel was produced with a relatively high hydrogen-to-carbon ratio by means of electrochemical hydrogenation of coal at ambient temperature and pressure. Ethylenediamine saturated with lithium chloride served both as the electrolyte and as the source of hydrogen.

78. Sakabe, Ts., Y. Ogo (1963) **Agitation in liquid-phase coal hydrogenation process and its scale effect,** *Journal of the Chemical Society of Japan, Industrial Chemistry Section* (Kogyo Kagaku Zasshi), Vol. 66, No. 12, pp. 1875—1880, A117. (in Japanese)

    In a continuous coal-hydrogenation process the coal-oil paste in the converter was agitated by introduction of H_2. In the experimental flow plant (converter dimensions 8 × 100 cm), the H_2 input under pressure was 50 litre/h. It was found that the agitation was satisfactory under the following conditions: pressure, 300 atm; temperature, 445—465° C; paste feed, 0.6—0.7 kg litre^{-1} h^{-1}; H_2: paste ratio, 2.0—2.4 Nm^3/kg.

79. Sakabe, Ts. (1963) **High-pressure liquid-phase hydrogenolysis of coal in continuous-type experimental apparatus. I.,** *Chemical Factory* (Kagaku Kojo), Vol. 7, No. 13, pp. 77—81. (in Japanese)

80. Sakabe, Ts. (1963) **High-pressure liquid-phase hydrogenolysis of coal in continuous-type experimental apparatus. II.** *Chemical Factory* (Kagaku Kojo), Vol. 8, No. 1, pp. 102—104. (in Japanese)

1964

81. Katsobashvili, Ya. R., E. I. Elbert, V. K. Smirnov (1964) **Hydrocracking of pitch distillates** (Gidrokreking pekovykh distillatov) *Khimiya i tekhnologiya topliv i masel*, No. 2, pp. 5—11. (in Russian)

    The optimum conditions were determined for hydrocracking pitch distillate mixed with an absorbent oil to obtain low- and medium-boiling hydrocarbons. Pitch distillate is a product of the oxidation of coal pitch at moderate temperatures.

82. He Hsueh-Lung, Ling Li-Hu, Wan Feng (1964) **Investigations on pressure hydrogenation of coal tar. I. Production of fuel and raw materials for chemical industries by liquid-phase hydrogenation of Hu-Chen-Tsu coal** *Chung Kuo K'e Hsueh-Yuan Hua Hsueh Hu Li Yang-Tsu-So Yang Tsu Pao-Kao Ts'u-Kang*, No 1, pp. 13—18. (in Chinese)

83. He Hsueh-Lang, Ling Li-Hu, Wan Feng (1964) **Investigations on pressure hydrogenation of coal tar. II. Solvent influence on liquid-phase hydrogenation** *Chung-Kuo K'e Hsueh-Yuan Hua-Hsueh Hu-Li Yang-Tsu-So Yang-Tsu Pao-Kao Tz'u-Kang*, No. 1, pp. 19 – 23. (in Chinese)

84. He Hsueh-Lang, Ling Li-Hu, Wan Feng, Liu Cheng-Yui, Tasai Guan-Yui, Hsun Dai-Phu (1964) **Investigacions on pressure hydrogenation of coal tar. III. Study of major factors affecting the process of liquid-phase pressure hydrogenation** *Chung-Kuo K'e Hsueh-Yuan Hua-Hsueh Hu-Li Yang-Tsu-So Yang-Tsu Pao-Kao Tz'u Kang*, No. 1, pp. 24 – 30. (in Chinese)

85. Gorin, E. (1964) **Production of hydrogen-enriched fuels from coal.** *U.S. Pat. No. 3,117,921*, publ. 14 January 1964. (in English)

    The suggested method of coal conversion into gasoline-type liquid fuel comprises the following operations: solvent extraction; distillation of the extract to obtain a wide fraction boiling up to 400° C; partial hydrogenation of the distillate to boiling point below 325° C; carbonization of the residue boiling above 325° C into ash-free coke and distillate; hydrogenation of the mixture of carbonization and partial hydrogenation distillate into a hydrogen-enriched liquid fuel.

86. Riedel, E., W. Steinberg, (1964) **Method for reactivation of catalysts for oil hydrorefining** (Verfahren zur Reaktivierung von Raffinationskatalysatoren) *G.D.R. 27.161* (Cl. 23b, 1,04 (C 10g)), Appl. 26 September 1960, publ. 15 February 1964. (in German)

    Hydrorefining at 170 – 220 atm of coal tar or other middle oil whose maximum boiling temperature lies within 320 – 360° C causes catalysts to lose their activity due to clogging with oily or tarry substances. To reactivate the catalysts, it is suggested that the catalyst be heated to 320 – 380° C and kept until it becomes clean in the hydrorefining reactor filled with crude gasoline or light oil boiling below 200° C.

87. Hirao, I., Ts. Fujimoto (1964) **Recent progress of coal chemistry. Pt. II. Hydrogenation of coal and utilization of its products,** *Journal of the Society of Organic Synthetic Chemistry* (Yuki Gosei Kagaku Kyokaishi), Vol. 22, No. 3, pp. 177 – 188. (in Japanese)

    This review of works on coal hydrogenation considers the mechanism of the process, the composition of the products obtained, and their application to synthesis of various organic compounds.

88. Reggel, L., I. Wender, R. Raymond (1964) **Catalytic dehydrogenation of coal. II. Reversibility of the dehydrogenation and reduction of coal.** *Fuel*, Vol. 43, No. 3, pp. 229 – 233. (in English)

    The coal was reduced at 110° C with lithium ethylene diamine and dehydrogenated at 347° C with palladium on $CaCO_3$ as catalyst and phenanthridine as vehicle. A vitrain of Pittsburgh coal, containing 81.5% C, 5.2% H, 1.6% N, 1.3% S, 2.6% ash, was alternately dehydrogenated and reduced. It is shown that the processes of reduction and dehydrogenation of coal are reversible and that hydrogen added to coal by reduction can be completely removed by dehydrogenation. On the other hand, hydrogen removed from coal by dehydrogenation can be only partly restored by reduction. Coal can evidently serve as both an acceptor and a donor of hydrogen: which role predominates depends on the rank. Higher rank coals (90% C) are excellent acceptors, while lower rank coals (83% C) are both acceptors and donors of hydrogen.

89. Arich, G., A. Cocco, L. Podda (1964) **Possibilities for utilization of Sulcis coal** (Possibilità di utilizzazione del carbone Sulcis) *Rivista dei combustibili*, Vol. 18, No. 4, pp. 126—139. (in Italian)

A description of an experimental study of partial liquefaction of Sulcis coal by hydrogenating extraction with tetralin, resulting in a liquid fraction and a residue suitable for further use as a fuel. In extraction tests in an autoclave, 40—45% of the product was in liquid form. The properties of the liquefied product and the material balance of the process are presented.

90. **Alpert, S. B., E. S. Johanson, S. C. Schuman (1964) Converting coal to more valuable fuels,** *Chemical Engineering Progress*, Vol. 60, No. 6, pp. 35—40. (in English)

A review of developments in coal hydrogenation for the preceding decade. Economic feasibility of coal conversion into liquid fuels was achieved by hydrogenation and hydrocracking at reasonable pressures (about 100—200 atm) and temperatures of 450° C in the presence of a catalyst in a fluidized bed. Process economy is analysed for a hydrogenation plant converting approximately 30,000 tons of coal per day into heavy oils.

91. Makhonine, J (1964) **Process and plant for distillation of coal and similar materials** (Procédé et installation de distillation du charbon et produits similaires) *Fr 1,367,716*, Appl 14 June 1963, publ. 24 July 1964 (in French)

The method of distillation and hydrogenation of coal and similar products consists in heating the initial product in the presence of a molten material which may or may not be able to dissolve the coal. Examples of solvents are molten cast iron and silver. The dissolved coal is hydrogenated in the usual manner or by nascent atomic hydrogen. The plant for the process is a vertical furnace comprising a generator containing the melt and two ring shaped pipes. The inner pipe feeds coal into the generator, while the outer one serves for the evacuation of the resulting volatiles. Air for heating and water vapour for H_2 formation are supplied through the bottom part of the furnace. A diagram of the plant is included.

92. Lawrence, S. J. (1964) **Estimates on commercial corona processing of coal.** *Chemical Engineering Progress*, Vol. 60, No. 6, pp. 45–51. (in English)

The article projects the results of experimental work into an economic analysis of a plant processing 10,000 tons of coal daily in a corona reactor. The plant produces liquid fuel and chemicals by hydrogenation of coal dissolved in anthracene oil. The calculations show that the profitability of this process is sensitive to the reaction efficiency. Construction of a plant processing 10,000 tons of coal per day is economically sound only if the energy consumption is no greater than 1.1 kWh per kilogram of coal.

93. Didelins, N. R., J. C. Fraser, M. Kawahata, C. D. Doyle (1964) **Corona processing of coal.** *Chemical Engineering Progress*, Vol. 60, No. 6, pp. 41–44. (in English)

A concentric electrode quartz reactor was used for an evaluation of product nature and specific energy consumption in corona processing of coal in a hydrogen atmosphere. Powdered coal (30–50 mesh) was packed between the electrodes and H_2 was passed through the coal with a corona developed in the space between the electrodes. The results showed that the yield was the same as for thermal decomposition, except that tar was not produced. The electrical conductivity of coal increased as it reacted until it shorted out the electric field. Attempts to overcome this problem by fluidization were not successful. More work is necessary in order to increase the yield of useful products and obtain better measurements.

94. Wielopolski, A. (1964) **Coal as a chemical raw material** (Wegiel jako surowiec chemiczny) *Przemysl chemiczny*, Vol 43, No. 8, pp. 442 to 427. (in Polish)

On the basis of a study carried out by highly developed countries, the future of chemical treatment of coal is forecast. Attention is paid to the following problems: coal hydrogenation to obtain raw materials and intermediates for the chemical industry and for production of jet fuels, coal hydrogasification leading to gas fuels of high caloric value, oxidation of coal under mild conditions and formation of cyclic polycarbonic acids, halogenation and sulphonation of coals. Modern views on the chemical structure of coal are presented. Raw materials and the economic aspects of the future intense development of carbochemistry are discussed.

95. Nagai, H., A. Akama (1964) **Hydrogenolysis and structure of Hokkaido coals.** *Journal of the Chemical Society of Japan, Industrial*

Chemistry Section (Kogyo Kagaku Zasshi), Vol. 67, No. 8, pp. 1266 to 1270, A74. (in Japanese)

Studies of coal hydrogenolysis are continued. Taiheiyo coal (O/C = 0.16, H/C = 0.99) has a smaller condensed structure of fairly simple aromatic and aliphatic units. It decomposes into low-boiling aromatic and saturated hydrocarbons, absorbing a small amount of hydrogen.

Yubari coal (O/C = 0.06, H/C = 0.90) is characteristic in its aliphatic structural part in the condensed aromatic unit structure which easily gives saturated hydrocarbons besides aromatic hydrocarbons in high yield on hydrogenolysis, in spite of low value of H/C for this coal.

It is concluded that hydrogenolysis of coal begins with the liberation of both tar acids and saturated hydrocarbons from the condensed aromatic units. Taiheiyo coal, however, is apparently hydrocracked by a one-step reaction into low-boiling oils.

96. Schroeder, W. C., L. G. Stevenson, Th. G. Stephenson (1964) **Hydrogenation of coal.** *U.S. Pat. No. 3,152,063*, Appl. 21 April 1961, publ. 6 October 1964. (in English)

Liquid or gaseous hydrocarbons are produced by passing pulverized coal, lignite, or char through the reactor at a temperature of 450–600 °C and pressure of 35–420 atm. A catalyst is applied by saturating the material with 0.5 to 1 % of ammonium molybdate. The residence time is 20–200 sec.

97. Alpert, S. B., E. S. Johanson, S. C. Schuman (1964) **Can coal compete for liquid fuels?** *Hydrocarbon Processing and Petroleum Refiner*, Vol. 43, No. 11, pp. 193–197. (in English)

During World War II, 15,000 tons of oil per day were produced in Germany by coal hydrogenation. For coal which will be commercially utilized for the production of liquid fuels, the following requirements must be met: an operable, economic coal hydrogenation process must be available; the cost of the hydrogen necessary to hydrogenate the coal must be low; and a satisfactory differential must exist between the price of coal and that of liquid fuels. It is considered that these requirements will be met in this decade and that the commercial conversion of coal to liquid fuel will be in operation by 1970. A process for production of lighter liquid fuels from petroleum residuals will serve as pilot in the development of the liquid fuel from coal process. The former process is in operation at a plant in Louisiana, USA, and will be repeated at another plant in Kuwait, which is to have a production rate of 8,000 tons per day. A flow chart of a future plant for coal hydrogenation and processing of all by-products is presented.

98. Lyon, L. B. (1964) **Liquid fuels from coal, other fossil hydrocarbons,** *Coal Age*, Vol. 69, No. 12, pp. 70–72, 75. (in English)

The article describes a number of processes under development by different companies for commercial conversion of coal into liquid fuels (low-temperature carbonization and hydrogenation for the production of gasoline, char and coke, or hydrogenation in a fluidized bed with solvent extraction and hydrocracking for the production of liquid fuels, etc.). The application of corona discharge for coal conversion into liquid fuels and gas is of special interest. The "H-Oil" process is a unique method of coal processing in a reactor with a fluidized bed of a catalyst. Economic analyses of coal and oil shale processing are presented.

99. Yamasaki, T. (1964) **Plant for coal hydrocracking,** *Japan. 29,440* (Cl. 18C3), Appl. 24 October 1962; publ. 18 December 1964. (in Japanese)

100. Rao, B. S. N., K. M. Murad, R. Vaidyeswaran, A. V. Ramaswamy, M. G. Krishna, and S. H. Zaheer (1964) **Diesel oil from coal tar** *Indian 80,742,* Appl. 14 February 1962; publ. 9 May 1964. (in English)

Tar obtained by low-temperature carbonization of coal or lignite was topped to 230° C at atmospheric pressure. The distillate was extracted with 10% NaOH to remove tar acids and alkali solubles, followed by 10% mineral acid to remove tar bases. The neutral hydrocarbon oil was distilled and the fraction boiling at 200° to 350° C was collected. It was useful as diesel oil. The diesel index of the fraction was increased by hydrogenation with hydrorefining catalysts.

101. Kawa, W., R. W. Hiteshue (1964) **Unconventional methods of hydrogenating coal,** *United States Bureau of Mines Information Circular No. 8215,* Washington, D.C., Government Printing Office. (in English)

The usual method of liquid phase coal hydrogenation at temperatures of 450–490° C and pressures of 100–170 atm with heavy oil recycling is now regarded by the industry as uneconomical. New ways to improve the economy of coal hydrogenation processes are sought in the application of monatomic hydrogen, electric discharges, ultrasonic effects etc. The article reviews research work and patents in this field.

102. Birch, T. G., J. D. Blackwood (1964) **Hydrogenation of reactive groups in Yallourn brown coal,** *Nature*, Vol. 201, No. 4921, pp. 797 to 798. (in English)

A review of a number of papers on hydrogenation of low rank coals. Some conclusions: About 40% of brown coal is rapidly hydrogenated at a pressure of 40 atm and a temperature of 800° C; CH_4 and water are produced, the residual coal undergoing slow hydrogenation into CH_4. The methane-to-water ratio is a constant. For each mole of methane, 0.63 mole of water is produced and 2.7 moles of H_2 are used. During this process the O_2 content decreases from 25% in the original coal to 1% in the residual coal, i.e. only reactive groups of the coal are attacked by hydrogenation. It is suggested that the oxygen groupings in brown coal are similar to those in lignin, and that only coals with high oxygen content will show an initial rapid hydrogenation.

103. Friedman, S., R. W. Hiteshue, M. D. Schlesinger (1964) **Hydrogenation of New Mexico coal at short residence time and high temperature,** *United States Bureau of Mines Report of Investigation No. 6470.* Washington, D.C., Government Printing Office. (in English)

The study was made in a bench-scale continuous reactor. Coal was hydrogenated to yield large amounts of volatiles. The object was to investigate the conditions of formation of a high-energy gas.

1965

104. Fomenko, O. S., M. D. Shapiro, I. N. Ruban, L. N. Artemyeva (1965) **Improvement of semicoke-oven tar by hydrogenation at moderate pressure** (Uluchshenie svoistv smoly polukoksovaniya uglei gidrogenizaciei pri srednikh davleniyakh) *Khimicheskaya tekhnologiya. Respublikankiy mezhvedomstvennyi nauchno-tekhnicheskii sbornik*, No. 1. pp. 90–95. (in Russian)

105. Fridman, G. E. (1965) **Chemical utilization of coal** (Puti khimicheskogo ispolzovaniya uglei) *Sbornik: "Khimicheskaya pererabotka topliv"*, Moskva, "Nauka". (in Russian)

106. Krichko, A. A., A. B. Vol-Epshtein (1965) **Principal trends and problems in the investigations of the processes of hydrogenation of fuels and the products thereof being conducted at the Institute for Fossil Fuels** (Osnovnye napravleniya i zadachi nauchnykh issledovaniy v oblasti gidrogenizacii topliv i produktov ikh pererabotki v Institute goryuchikh iskopaemykh) *Sbornik: "Khimiya i tekhnologiya smol termicheskoi pererabotki tverdogo topliva"*, pp. 267–285, Moskva, "Nauka". (in Russian)

107. Bronovets, T. M., E. M. Taits (1965) **Bituminous and brown coals as a raw material for the preparation of fusible and soluble products**

(Kamennye i burye ugli kak syryo dlya polucheniya plavkikh i rastvorimykh produktov) *Sbornik: "Novoe v briketirovanii i koksovanii uglei"*, Moskva, "Nauka". (in Russian)

108. Gorin, E. (1965) **Process for producing hydrogen-enriched hydrocarbonaceous products from coal.** *U.S. Pat. No. 3,184,401* (Cl. 208-8), publ. 18 June 1965. (in English)

Extraction is carried out by heating coal with a solvent to 300 – 500° C. The resulting extract undergoes catalytic hydrogenation. To avoid rapid deactivation of the catalyst the extract should be pretreated to eliminate ash by heating it with a hydrocarbonaceous vehicle of hydrogen to 250 – 373° C (the temperature must be below that of extraction). After elimination of 60% of the ash, the extract and the solvent are hydrogenated in the liquid phase in the presence of $CoO-MoO_3-Al_2O_3$ catalyst, at a temperature of 440° C, pressure of 245 atm and residence time of 2.8 h. The yields (percentage of the raw material weight) were: C_1-C_3 – 12.5; C_4 – 5.2; C_5 – 32.5–80.6. The solvent used was a mixture of the hydrogenate fractions boiling at 260° – 325° C and 325° – 425° C taken in equal amounts.

109. Gorin, E. (1965) **Method for producing hydrogen-enriched liquid fuels from coal.** *Austral. Pat. No. 258,145* (Cl. 20.2, 20.6 (C10g, C10b)) publ. 24 March 1965. (in English)

A coking coal was extracted at 350° C by decalin (220 kg per 100 kg of coal). The yield of the extract was 23%, the solid residue was 75% of coal weight. Precipitation of the extract was performed with *n*-hexane. The solid residue was carbonized at 470° C. Hydrogenation of the liquid fractions yielded a liquid distillate boiling below 360° C; Their mass constituted 23.2 to 25.8% of the mass of dry, ash-free coal. H_2 consumption was in the range 5.25 – 5.69%.

110. Hawk, C. O. and R. W. Hiteshue (1965) **Hydrogenation of coal in the batch autoclave.** *United States Bureau of Mines Bulletin No. 622*, Washington, D.C. (in English)

Results of experimental research in coal hydrogenation by a "dry" method without a solvent are presented. The experiments were carried out in a rotating autoclave of 1.2 litre volume. The effects of initial pressures of 70 – 280 atm, temperatures of 400 – 500° C, many different catalysts, and other factors were investigated. For the sake of comparison some tests were made with a solvent present. Pittsburgh and Wyoming coals with a high content of volatiles were tested. It is noted that naphthenates of Mo, Ni, Sn, Fe, Co, Zn and Cu show high activity. In experiments at 500° C and 525 atm the presence of 0.1% (metal content) of these catalysts caused an 80 % conversion of coal while being heated to the above temperature. Molybdenum naphthenate provided 90 % conversion when 0.01 % of Mo was present.

111. Sakabe, Ts., Y. Ogo, R. Sassa, N. Norie, Y. Kanbayashi, Sh. Takahashi, M. Suzuki (1965) **High-pressure liquid-phase hydrogenation of coal by experimental flow plant,** *Resources Research Institute, Report No. 61.* (in Japanese)

The plant is designed to hydrogenate 50—80 kg of coal per day at a pressure of 700 atm. Miike coal mixed with 2.5 % of Bayer mass catalyst was fed at 300 atm into the first and second converters in succession wherein the temperatures were 445° C and 465° C, respectively. The paste feed rate was 0.6—0.7 kg litre^{-1} h^{-1}. H_2: paste ratio was 1.9—2.3 Nm^3/kg. The results (percentage of the dry, ash-free coal) were: liquefaction, 96—97; yield of middle oil and gasoline, 60—62; gasification, 27—30; H_2 consumption, 6—8. Agitation of the paste has a pronounced effect on the results.

112. Ishii, T., Y. Maekawa, T. Gen (1965) **Reaction kinetics of coal hydrogenation under high pressure,** *Chemical Engineering of Japan* (Kagaku Kogaku), Vol. 29, No. 12, pp. 988—995. (in Japanese)

Autoclave hydrogenation of coal and brown coal (ash content 47.8 and 43.6 %, carbon content — 75.5 and 84.0 %, respectively) was carried out at pressures of 210—230 atm and temperatures of 300—450° C. The process rate was measured by controlling the yields of the residue insoluble in benzene, gas, oil and asphaltenes After 35—40 min at approximately 450° C, coal conversion reached 92—94 %. The determination of these characteristics for various residence times and temperatures made it possible to calculate the values of the reaction rate constants. It was found that the coal hydrogenation consisted of two steps. The rate of reaction in the first step was high and that in the second step was much lower. Both steps could be expressed as first order reactions with respect to the coal remaining. The first step of hydrogenation yielded asphaltenes, which were then hydrogenated in a single step; this was proved by experimental hydrogenation of asphaltenes. The activation energies of the first and second stages of brown coal hydrogenation were equal to 19.6 and 16.3 (without a solvent — 10.4), respectively, that of asphaltenes 25.9 kcal.

113. Boyer, A. F., P. Payen (1965) **Mild hydrogenation of coals and analysis of the hydrocarbons obtained.** (Hydrogénation ménagée de charbons et analyse des hydrocarbures obtenus) *Bulletin de la Société chimique de France*, No 10, pp 2765—2770 (in French)

Catalytic (MoS_2) hydrogenation of two types of low rank coal dispersed on glass cloth was performed by slow heating to 420° C

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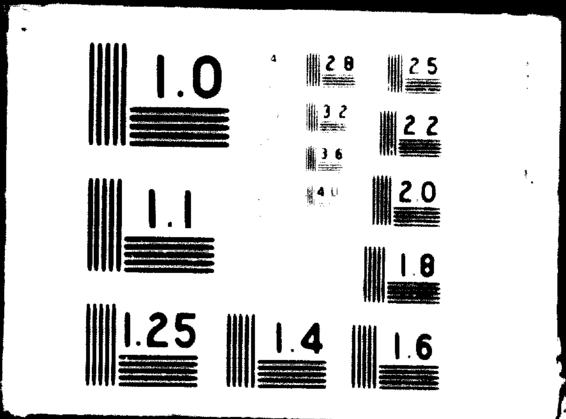

in a H_2 flow under pressure. The distillate products were entrained as soon as they formed to avoid excessive degradation of them. The hydrogen was recirculated, and volatile products were condensed. A flow chart of the plant is presented. The authors succeeded in converting almost 90% of vitrinite and exinite into water and volatile products. An analysis of the products revealed hydrocarbons whose aliphatic chains were long or slightly branched. Hydrocarbons were represented by normal paraffins up to C_{36} but mainly from C_{14} to C_{28}, slightly branched isoparaffins and partly hydrogenated aromatic compounds. The research showed that the coals studied were composed of structural units with two to four condensed rings.

114. Mosely, F., D. Patterson (1965) **The rapid high-temperature hydrogenation of coal chars. Part 1. Hydrogen pressures up to 100 atmospheres,** *Journal of the Institute of Fuel*, Vol. 38, No. 288, pp. 13–23. (in English)

The coal char was prepared from non-caking coals (volatile matter — 35.38%; ash — 3.04%) by heating for three hours in nitrogen to 700° C. The char contained 90.15% C, 1.65% H, 4–5% volatiles, and 4.40% ash.

The experiments were performed in a tube at a hydrogen pressure of 100 atm, temperature of 800–950° C, and hydrogen flow rate of 80–280 litre/h. Twelve grams of the material was introduced as 50–100-mesh powder. It was established that the initial rate of hydrogenation is very large but rapidly decays to a steady low value. The hydrogenation gases comprise CH_4, C_2H_6, and CO. The decay of the rate of hydrogenation was independent of the extent of carbon gasification; it depended only on time and temperature. The rate of CH_4 formation was directly proportional to the H_2 pressure over the whole range of hydrogenation times.

115. Taketani, S., H. Nagai (1965) **Method of coal-paste preparation,** *Japan. 1,248* (Cl. 18C3), publ. 23 January 1965. (in Japanese)

Coal paste for hydrocracking was prepared by mixing pulverized coal with heavy oil in the ratio of 2:8 for lignite and 1.5:8 5 for coal. The mixture is heated to 250°–350° C or 250°–375° C, respectively. For instance, 21 kg of coal of 5% humidity, containing more than 80% of particles of 200 mesh, are mixed with 80 kg of coal fraction 260°–360° C. The mixture is heated in an autoclave to 350° C under H_2 or N_2. Heating takes 6–7 h, and the residence time is 1 h. After that the mixture is cooled to 50° C, and 98.5 kg of product is released. By adding 54.2 kg of dry lignite to it, a paste with coal-to-heavy-oil ratio of 2:3 is formed.

116. Mirza, A., M. A. Masood, A. V. Ramaswamy, R. Vaidyeswaran (1965) **Medium-pressure hydrogenation of neutral oil fractions of**

low-temperature tar into diesel oil (Mitteldruckhydrierung von Tieftemperaturteer-Neutralölfraktionen zu Dieselöl) *Brennstoff-Chemie*, Vol. 46, No. 11, pp. 355–358. (in German)

Tar from low-temperature carbonization was hydrogenated in a commercial plant at 300–450° C, 70–100 atm, and space velocity of tar of 0.3–1.0 h^{-1} in the presence of a commercial catalyst (tungsten and nickel sulphides). Reaction rates, yields and product properties are presented. The final product had a diesel index of 48–50.

117. Jenny, E. J. (1965) **Hydrocracking of tars and other high-molecular-weight hydrocarbons,** *U.S. Pat. No. 3,200,061* (Cl. 208-58), publ. 10 August 1965. (in English)

A process of gasoline production from high-molecular-weight compounds poor in hydrogen (coals, heavy sulphurized crude oils etc.) is described. The process is carried out in the presence of a H-containing gas in the fluidized bed of fine particles of char or coal whose temperature is maintained by their partial combustion and by heat exchange with hot products of the reaction. If the raw material is solid (e.g., coal), it must be pulverized and mixed with a solvent (preferably, a product of the process) prior to being fed into the reactor.

1966

118. Fumich, G. (1966) **Office of coal research continues liquid extraction projects,** *Journal of Petroleum Technology,* Vol. 18, No. 8, pp. 939 to 943. (in English)

The Office of Coal Research awards contracts for fundamental investigations in coal technology. This article by its Director reviews various projects under development for coal conversion into city gas and gasoline. The Consol project (Consolidated Coal Company) calls for dissolution of coal in a liquid product, filtering and catalytic hydrocracking of the filtrate. The distillate is subsequently processed by standard techniques of petroleum refining. The variant of the process under development by Hydrocarbon Research, Inc., is a method for converting 80% of coal into liquid products by catalytic processing in a fluidized bed in the presence of H_2. Again, the liquid product is refined by standard methods. Project Seacoke (Atlantic Richfield Co.) is a combination of fluidized coking and burning of a fine-grain coke. Project COED, undertaken by FMC Corp., involves the multistage fluidized pyrolysis of coal. The resulting char is burned and the liquid product used for gasoline production. This process was tested in 1965 on a pilot plant with a feed capacity of 1 ton/day of coal.

119. Meimarakis, G. (1966) **Investigations in chemical technology.** *Arts et manufactures*, No. 160. (in French)

The article describes the major trends in the studies of chemical processing of coal, including high rate pyrolysis, coal hydrogenation without a solvent, tar analysis and tar processing methods.

120. Turanska, D. (1966) **New investigations in coal liquefaction** (Nowsze badania and upxynnianiem węgla) *Chemik*, Vol. 19, No. 4, pp. 127 to 129. (in Polish)

A review of recent work in the USA on hydrogenation of coal in an electric corona or an electric arc.

121. Vishnoi, D. M. (1966) **On production of liquid fuel from coal in India,** *Metals and Minerals Review*, Vol. 5, No. 8, pp. 10–13. (in English)

The author thinks it necessary for India to develop home industry for liquid fuel production from coal. The most probable technology is considered to be coal gasification, gas refining and application of the Fischer-Tropsch method for liquid fuel production.

122. Lherm (1966) **Conversion of solid fossil fuels into hydrocarbons,** *Journal Techn. Petrole*, Vol. 21, No. 239. (in French)

Brief data are presented on recent activities in various countries in the area of conversion of coals and shales into liquid fuels, city gas and chemicals.

123. Sakabe, Ts., M. Horie, Y. Ogo, R. Sassa, Sh. Takahashi, Y. Kanbayashi (1966) **Production of coal chemicals by the combination of high pressure liquid phase hydrogenation with hydro-dealkylation.** *Resources Research Institute. Reports*, No. 36, 44 pp. (in Japanese)

Studies on the production of aromatic chemicals from coal by means of hydrogenation and hydro-dealkylation have been carried out by the Resources Research Institute (Japan) since 1956. The yields of Miike coal hydrogenation by an experimental flow plant are presented. An analysis of C_6–C_8 phenols in the hydrogenate shows the following components (percentage of the mass of dry, ash-free coal): phenol, 0.98; *o*-cresol, 0.40; *m*-cresol, 0.99; *p*-cresol, 0.32; *o*-ethylphenol, 0.09; *m*- and *p*-ethylphenol, 0.65; 2,3-xylenol, 0.05; 2,4- and 2,5-xylenol, 0.26; 2,6-xylenol, 0.03; 3,4-xylenol, 0.01; 3,5-xylenol, 0.11. Dealkylation of the neutral oil boiling below 300 °C was carried out with $Cr_2O_3 \cdot K_2O \cdot Al_2O_3$ catalyst at 30 atm, 500 °C and 580 °C, space velocity of $0.5\ \mathrm{kg\ l^{-1}\ h^{-1}}$, and hydrogen feed mole ratio of 10 : 1. The yield of the dealkylate was 76.4 mole %, its composition (mole %) being as follows: benzene, 7.5; toluene, 11.7; xylene, 15.6; naphthalene, 37.5, methylnaphthalene, 1.7; fluorene, 2.4

124. Horing, M., L. Rauchle (1966) **The technology of coal and petroleum hydrogenation** (Zur Technologie der Kohle- und Ölhydrierung) *Chemie-Ingenieur-Technik*, Vol. 38, No. 3, pp. 205—208. (in German)

The article commemorates Matthias Pier and reviews his work in the field of high-pressure hydrogenation of coal and tar and thermal processing of petroleum.

125. Huntington, M. G. (1966) **Quadriphase low-pressure method for partial liquefaction of coal.** *U.S. Pat. No. 3,247,092* (Cl. 208-8), publ. 19 April 1966. (in English)

A continuous process of thermal treatment of coal is described. After pulverization the crude coal enters a vertical reactor comprising a series of pressurized gastight chambers. Dried and preheated coal is fed into the distillitation chamber where primary volatiles are separated to be hydrocracked, reformed, and dealkylated simultaneously. Hot hydrogen at high pressure serves as heat carrier. The char produced is separated from the gases and transferred into the oxidation chamber. Here it is heated to partial combustion. After that, the heated char makes contact with CH_4 and decomposes it into C and H_2. The resulting hydrogen is passed into the distillation chamber, while the char is returned to the oxidation chamber.

126. Hellwig, K. C., E. S. Johanson, C. A. Johnson, S. C. Schuman (1966) **Make liquid fuels from coal.** *Hydrocarbon Processing and Petroleum Refiner*, Vol. 45, No. 5, pp. 165—169. (in English)

The process designated as "H-Coal" for the conversion of coal into gasoline and furnace oil by hydrogenation and progressive treatment of the hydrogenate by petroleum processing techniques is described. The process has been tested on a continuously operating reactor with a capacity of 7—12 kg/day of coal. Since 1966, tests have been held on a plant processing 3 tons of coal per day. A commercial plant is to be constructed to start operation by 1970. Economic summaries for three versions of the process are presented. A plant yielding 15,900 m^3 of gasoline per day has a payout period of 6—7 years.

127. Cochran, N. P. (1966) **Gasoline from coal.** *Proceedings of the Illinois Mining Institute*, No. 74, pp. 31—43. (in English)

Flow charts and economic data are presented for projects of coal conversion into gasoline and other products, projects which are being developed by US companies on contracts with the Office of Coal Research. The COED, H-Coal and Consol methods are discussed. In the COED project, coal is processed by multistage pyrolysis yielding high-calorific-value gas, H_2 for ammonia synthesis, crude oil for further hydrogenation and char. The over-all scheme for the H-Coal project is coal hydrogenation, processing of the liquid product by hydrogenation and conversion of the solid residue into char. In Consol, solvent extraction of coal is effected under hydrogen pressure; the extract is separated from the solid residue and hydrogenated, while the residue is carbonized. The latter project is the most advanced in development.

128. Sakabe, Ts., R. Sassa, M. Horie (1966) **Production of chemical raw materials from coal by the combination of high-pressure liquid-phase hydrogenation with hydro-dealkylation.** *Journal of the Chemical Society of Japan, Industrial Chemistry Section* (Kogyo Kagaku Zasshi), Vol. 69, No. 6, pp. 1085—1090, A61. (in Japanese)

The article contains the results of hydro-dealkylation of neutral oil and high-boiling acidic oil extracted from the middle oil and gasoline, which are the main products of liquid-phase hydrogenation of Miike coal. Hydro-dealkylation of the neutral oil was carried out with $Cr_2O_3 \cdot K_2O \cdot Al_2O_3$ catalyst at 30 atm and 500°—580° C. The yield of chemicals from dry, ash-free coal were: benzene, 1.6%, toluene, 3.0%; xylene and ethylbenzene, 4.6%; naphthalene, 13.3%; gaseous hydrocarbons, 44.6%. As for the thermal hydro-dealkylation of the acidic oil based on the Wells and Lond methods, the yield of phenol and cresol were not so high as compared with those proposed by Donath.

129. Zielke, C. W., R. T. Struck, J. M. Evans, Ch. P. Constanza, E. Gorin (1966) **Molten zinc halide catalyst for hydrocracking coal extract and coal,** *Industrial and Engineering Chemistry, Process Design and Development*, Vol. 5, No. 2, pp. 158—164. (in English)

Zinc chloride and extract of Pittsburgh coal, produced by dissolving the coal in tetralin after 45 min of residence at 380° C, were used in the experiments. The extract represented 53% of the coal. Unextracted coal was removed in a pressure filter at 200° C. A 300-ml shaking autoclave served for the study of conditions of hydrocracking and their effect on the product yield. The amount of coal conversion was 44.1%, at 425° C, 300 atm, residence time 1 hour, and catalyst-material ratio of 0.02. Hydrogen consumption was 4.13%.

When coal was hydrogenated in the presence of $ZnCl_2$ at 385° C, 140 atm, residence time 1 h, and catalyst-material ratio of 3.5, the conversion and hydrogen consumption were 74.2% and 8.7%, respectively. Data are presented on the effects of temperature, pressure, catalyst quantity and other factors.

130. Perry, R. C., Ch. A. Albright (1966) **Catalytic hydrogenation of carbonized coal vapors,** *U.S. Pat. No. 3,231,486,* publ. 25 January 1966. (in English)

A method is proposed for the stabilization of liquids produced by low-temperature coal carbonization by their catalytic hydrogenation prior to condensation. Finely ground coal is mixed with H_2 at a pressure of 200 atm, and the resulting suspension is directed to a carbonization zone at a temperature of 450°—600° C, where the suspension is kept in a fluidized bed. The mixture of tar and gas is passed over a hydrogenation catalyst in a different zone at a temperature of 400°—475° C and a pressure of 15 to 200 atm, after which it is condensed. The catalyst contains 0.5% Ni, 1.0% Co and 8.3% Mo, the rest being Al_2O_3. Before use, the catalyst is reduced in a flow of H_2 at 350°—400° C, pressure 1 atm, and residence time 4—12 h. Coal coking in the fluidized bed is prevented either by an addition of non-coking coal of the lignite type or by pre-oxidation. The fluidizing gas is heated to 400° C before it is mixed with the coal. The process is recommended for production of tar rich in low-boiling phenols.

131. Mirza, A., M. A. Masood, M. M. Mallikarjunan, R. Vaidyeswaran (1966) **Catalytic hydrogenation of low-temperature tar fractions,** *Chemical Age of India,* Vol. 17, No. 3, pp. 240—245. (in English)

Fractions of neutral oil of low-temperature coal tar boiling in the range 190°—320° C were hydrogenated to obtain fractions with boiling ranges above 150° C or 200° C. The object was to find the conditions of maximum yield of lighter fractions. The apparatus was a flow-by reactor of 2-litre volume, half filled with the commercial catalyst (WS_2 + NiS + Al_2O_3). The best quality of the product was obtained at 380° C, 100 atm, and space velocity of 2 h^{-1}. Properties of the hydrogenates are presented.

132. Rao, M. J., M. G. Krishna (1966) **Hydrocracking of neutral low-temperature tar fractions,** *Chemical Age of India,* Vol. 17, No. 3, pp. 233—239. (in English)

The object of the investigation was to determine the conditions of gasoline production from fractions 210°—350° C of neutral oil of low-temperature tar produced in the Lurgi plant (650° C). The experiments were carried out in a 1 m length reactor with an internal diameter of 12 mm. The temperatures were 400°—500° C, pressures 30—70 atm, and space velocities 1 h^{-1}. The maximum yields of hydrogenate fractions up to 210° C were obtained at 500° C and amounted to (in volume %): 24.7 for $Al_2O_3 + SiO_2$ catalyst; 29.3 for CoO + MoO_3 on Al_2O_3 + SiO_2 catalyst; 29.0 for NiS on $Al_2O_3 + SiO_2$ catalyst. They contained 36.0, 39.5 and 38.4% aromatics, respectively.

133. Švajgl, O. (1966) **Contribution to the studies on pressure hydrogenation of tar distillates** (Přispevek ke studiu tlakové hydrogenace dehtovych distilatu) *Chimicky průmysl*, Vol. 16, No. 4, pp. 209—214. (in Czech)

Conditions for the selective elimination of nitrogen bases from coal tar distillate without hydrogenation of aromatic hydrocarbons were studied. A tar fraction boiling in the range 91° to 295° C was used in the experiments. It contained 40% (by volume) phenols, and bases equivalent to 3,919 mg NH_3 per litre. The hydrocarbons included (%) 34.5 aromatics, 45.7 olefins, and 19.8 saturated hydrocarbons. A series of experiments was carried out with a 1:2 mixture of this fraction and the diesel-oil fraction of Romashkino crude oil. Various catalysts containing Mo, W, Co, and Ni on Al_2O_3 were tested. The influence of temperature in the range 360°—440° C on the base elimination ratio and the composition of the hydrogenate, at pressures of 100—150 and 320 atm and space velocities of 0.7—1.25 kg litre^{-1} h^{-1} was investigated.

1967

134. **Coal gains favour as new source for liquid fuels,** (1967) *Oil and Gas Journal*, Vol. 65, No. 51, pp. 41—44. (in English)

Coal has taken the lead over shale oil as a supplementary source of hydrocarbon energy in the US. In 3—5 years present difficulties should be eliminated; favourable economics may be 10—15 years away. Coal product yields are about 3 bbl/ton compared to 35 gal/ton for shale. Disposal of waste shale is a problem, whereas two thirds of the coal is converted to liquids and the remaining one third is char which can be used as fuel or in H_2 manufacture.

135. Rao, B. S. N. (1967) **Recent trends in some aspects of coal processing, Non-fuel uses of coal,** *Petroleum and Hydrocarbons*, Vol. 2, No. 1, pp. 5—12; No. 3, pp. 99—102 (Appendix to "Chemical Age of India", Vol. 18, Nos. 4 and 10). (in English)

The articles review research work conducted in the USA on non-fuel uses of coal. These projects are aimed at coal conversion into gasoline, liquid fuel, and gas of high calorific value. Plasma processing of coal and irradiation with a flash of high intensity light are included.

136. Morley, R. J. (1967) **Coal and chemicals,** *Mining Electrical and Mechanical Engineer*, Vol. 48, No. 561, pp. 141—145. (in English)

A review of chemical products obtained from coal by carbonization, hydrogenation and other treatment.

137. Groborz, J. (1967) **Work in progress in the United States on new methods of coal processing** (Prace badawcze w Stanach Zjednoczonych nad nowymi metodami przerobki wegla) *Problemy syntezy organicznej*, Vol. 13, No. 8, pp. 489—510. (in Czech)

A review containing a description of projects for coal conversion into liquid fuels and gas.

138. Zawada, E. (1967) **Reappearance of coal as a chemical raw material** (Wiegiel jako surowiec chemiczny znow na widowni) *Przemysl chemiczny*, Vol. 46, No. 3, pp. 117—120. (in Czech)

To illustrate the new interest in coal as a raw material, this review describes the non-fuel uses of coal under development in the US. These projects call for hydrogenation, gasification and solvent extraction of coal.

139. **Project Gasoline in final development stage** (1967) *Chemical and Engineering News*, Vol. 45, No. 25, pp. 96—98, 102, 104. (in English)

The state of gasoline-from-coal projects under study by various US companies on contracts from the Office of Coal Research. At this time, Project Gasoline was at an advanced state of testing at a pilot plant. Projects COED, H-Coal, and Seacoke were being tested at research facilities. Flow charts and brief descriptions of the four projects are included.

140. Bond, R. L., L. G. C. Dryden (1967) **Saleable products from solvent extraction of coal,** *British Chemical Engineering*, Vol. 12, No. 5, pp. 731—736. (in English)

A review of developments in coal utilization by solvent extraction for the period of 1955—1956. Possibilities of coal conversion into liquid fuels, chemical products, and ash-free concentrate for electrode coals are discussed.

141. **Big synthetic oil industry seen vital** (1967) *Oil and Gas Journal*, Vol. 65, No. 10, p. 69. (in English)

In 1980 the US will produce 11.5 million barrels of natural oil per day. That same year the demand will be 17 million bbl/day. The reserves-to-production ratio will drop from today's 17:1 to 9.5:1 by that time. It is supposed in this connexion that, by 1980 the US industry will produce large amounts of synthetic oil. Tar sands, shale and coal are suggested as sources for synthetic oil production.

142. Cochran, N. P., G. I. Staber (1967) **Progress in producing useful hydrocarbons from coal,** *Journal of Petroleum Technology*, Vol. 19, No. 10, pp. 1345—1350. (in English)

A review of processes under development in the US for coal conversion into pipeline-quality gas and liquid fuels, such as gasoline or fuel oil. Flow charts for the processes and approximate estimates of product costs are given.

143. Cochran, N. P. (1967) **Finding fresh uses for coal,** *New Scientist*, Vol. 34, No. 542, pp. 212—215. (in English)

Flow charts, brief descriptions and economic calculations are given for new coal processing methods. These processes were developed by individual US companies on contracts from the Office of Coal Research. The processes for liquid fuel production and a number of versions of fuel gas production that combine solvent extraction with hydrogenation are discussed.

144. Schuman, S. C., R. H. Wolk, M. C. Chervenak (1967) **Hydrogenation of coal,** *U.S. Pat. No. 3,321,393* (Cl. 208/10), publ. 23 May 1967. (in English)

A method of coal hydrogenation in a fluidized bed of a catalyst is described. Bituminous Illinois No. 6 coal pulverized to 270 mesh was hydrogenated at a pressure of 190—200 atm and temperature of 270° C. Alumina, cobalt and molybdenum served as the catalyst. Hydrogen consumption equalled 2.6 m^3/kg of coal. More than 80% of the coal was converted into liquid and gaseous products, the yield of liquids boiling below 500° C being in excess of 500 litres per ton of coal. The amount of coal that could be processed was 300—500 kg per cubic metre of the reactor volume.

145. **Coal liquefaction projects underway,** *Independent Petroleum Monthly*, 1967, No. 3, p. 31. (in English)

A brief review of three methods being developed in the US for production of liquid fuels from coal. The projects are low-temperature carbonization (COED), solvent extraction with mild hydrogenation (Gasoline), and catalytic hydrogenation of coal (H-Coal).

146. **COED coal-to-crude process moves into pilot-plant stage,** *Oil and Gas Journal*, 1967, Vol. 65, No. 44, pp. 140—141. (in English)

The COED process has been developed by FMC Corp. under contract with the US Office of Coal Research. It involves step-by-step pyrolysis of coal in a fluidized bed and further processing of the liquid products. The method was tested on a unit processing 50 kg/h. A pilot capable of 10 tons of coal per day is to be constructed. Liquid products are supplemented with gas of high calorific value, hydrogen for ammonia synthesis, and char.

The process development unit yielded the following products (weight percentage of dry coal): char, 54.3; oil, 23.6; gas, 15.0; water, 7.0. The gas comprised (volume percentage): CO, 22.1; H_2, 51.0; CH_4, 20.9; C_2H_6, 6.0.

147. **Oil from coal,** *Colliery Engineering*, 1967, Vol. 44, No. 525, pp. 425 to 426. (in English)

A pilot plant in Cresap, West Virginia (US), capable of converting 25 tons of coal per day into liquid fuel is described.

148. **Gasoline-from-coal produced on pilot plant,** *Oil and Gas Journal*, 1967, Vol. 65, No. 6, p. 123. (in English)

A plant capable of converting 20 – 25 tons of coal into approximately 10 m³ of gasoline daily was built and began operating in Cresap. The construction of a plant is planned which will process 10,000 – 30,000 tons of coal into 480 – 1,600 m³ of gasoline per day. The yield of gasoline approaches 480 litres per ton of coal. The two stage Consol project is the basic process of the plant. The plant employs 100 men.

149. **Coal's entrance into gasoline market set at 3 yr,** *Coal Age*, 1967, Vol. 72, No. 6, pp. 26 – 28. (in English)

The Consol process of gasoline production from coal is being developed in the US at a pilot plant at Cresap, West Virginia. The process involves thermal dissolution of coal in heavy oil, separation of the extract from coal residue, low-temperature carbonization of the extract and distillation to separate the light products, hydrogenation of the residue, and hydrogenate processing by petrochemical methods. A selective solvent is used; the hydrogenation catalyst is nickel-molybdenum on Al_2O_3. The plant, which cost \$225 million and operates on Pittsburgh coal, will have an output of 22.9 million barrels of gasoline per year. It consumes 25,000 tons of coal per day. The annual operating costs amount to \$73.4 million, which includes \$31.4 million for the coal and \$10.0 million for the catalyst and chemicals. The operating income equals \$131.1 million, including \$101.1 million for the gasoline and \$30.0 million for by-products.

150. Eddinger, R. T., S. K. Reed, (1967) **COED research aims at oil, gas and char from coal,** *Coal Age*, Vol. 72, No. 1, pp. 90 – 92. (in English)

The COED project, in development since 1962 by the FMC Corp. under a contract with the US Office of Coal Research, provides for complete conversion of coal into liquid fuel, fuel oil and electricity. In 1965 the process was tested on a 50 kg/h development unit. A pilot plant is to be constructed in 1967, and 1972 will see the start of a commercial plant capable of a throughput of 3.5 million tons of coal per year. The yield from one ton of Illinois No. 6 coal is 610 kg of char, 210 litres of crude oil, and 136 m³ of gas, or, alternatively, 560 kg of char, 260 litres of oil, and 280 m³ of H_2.

151. **Pilot plant for gasoline production from coal in the U.S.,** *Oil and Gas Journal*, 1967, Vol. 65, No. 23. (in English)

A pilot plant for coal-to-gasoline conversion was put into service in Cresap, West Virginia, in May 1967. The cost of the plant was \$3.5 million. The estimated cost of gasoline is in the range of

10.5–13 cents per gallon. If the pilot plant, processing 25 tons of coal per day, shows these estimates to be correct, a commercial plant based on the Consol project will be built. After separation of the solvent, distillation and refining, the extract is fed into the reactor for hydrogenation on a fixed bed of Co–Mo–Al_2O_3 catalyst at 425 °C and 210 atm. Hydrogen consumption was 900 Nm^3/m^3 of product, or 420 m^3 per ton of coal. The pilot plant is also testing a new catalyst based on $ZnCl_2$ that may be capable of direct conversion of coal into gasoline.

152. **Plant for H-Coal project,** *Petro/Chem. Engineer*, 1967, Vol. 39, No. 11. (in English)

An H-Coal plant in service at Trenton, New Jersey (US), processes 3 tons of coal per day by catalytic hydrogenation in the entrained state. Hydrogen is obtained from coal and char. Construction has begun of a plant capable of producing 5 million cubic metres per year. A 230-ton/day plant being planned will serve for a 2-year long process development effort. The commercial plant capable of producing 5 million cubic metres per year will cost $364 million, and will pay for itself in 7 years.

153. **Economy of liquid fuel production from coal,** *Oil and Gas Journal*, 1967, Vol. 65, No. 6, pp. 58–59. (in English)

Economic aspects of the H-Coal process are discussed. A plant capable of 30,000 bbl/day, producing 3.5 bbl of saleable products from 1 ton of coal, yields a profit of 3.3 to 4.6 %, and pays for itself in 8.3–8.8 years. The profit increases if the process is aimed at the production of high grade gasoline with an octane rating similar to that of gasoline from an oil refinery. In a plant of 100,000 bbl/day capacity the profit will amount to 6.4–8.1 %, and the plant will repay in 6.3–7.1 years. The profit will be even larger if the plant is aimed at gasoline production. The economic estimates are based on prices similar to current market prices: gasoline, $4.62/bbl; gas oil, $3.75/bbl; and heavy oil, $2.00/bbl. The construction cost of a plant for conversion of coal into liquid products will be about three times as high as that of the usual petroleum refinery. Building of a 100,000 bbl/day plant will require a capital investment of $332,400,000 to $346,070,000, depending on the specifications of the commercial products.

154. **Catalytic hydrotreating of coal-derived liquids,** *Mining Equipment News*, 1967, Vol. 9, No. 8, p. 5. (in English)

The US Office of Coal Research reports on the results of a laboratory investigation to determine suitable catalysts and hydrotreating systems for converting the liquid products or tars derived from coal to a suitable raw material for the production of gasoline.

155. Bull, W., L. Stevenson, D. L. Kloepper, Th. F. Rogers (1967) **Solvation process for carbonaceous fuels.** *U.S. Pat. No. 3,341,447* (Cl. 208-8), Appl. 18 January 1965, publ. 12 September 1967. (in English)

Solvent extraction is used to obtain low-ash and low-sulphur fuel from fossil solid fuels — coal and brown coal, lignite and peat. Ground fuel is mixed with a solvent in ratios ranging from 1:1 to 4:1. The paste obtained is heated to 370 – 500° C and held at a constant temperature for a given period of time in the zone of dissolution. Then the solvent is separated from the solid residue and returned to the run. A highly aromatized product, obtained in the process, is used as solvent. The product boiling in the range of 150°—750° C has a density of 1.1 g/cm³ and C:H ratio of 1.0 to 0.3. For the initial run an anthracene oil is used. For greater efficiency the H_2 pressure is kept above 35 atm.

156. Curran, G. P., R. T. Struek, E. Gorin (1967) **Mechanism of the hydrogen-transfer process to coal and coal extract,** *Industrial and Engineering Chemistry, Process Design and Development*, Vol. 6, No. 2, pp. 166—173. (in English)

To study solvent extraction of coal, a coal containing 14.9% ash, 70.21% carbon, and 4.66% hydrogen was used. A mixture (1:1 to 1:3) of coal (100—200 mesh grain size) and tetralin was heated in a 1-litre autoclave at 440° C for about 1 h. Solvent extraction of coal in the presence of the hydrogen donor is found to occur via free radicals. Irrespective of donor origin the maximum amount of hydrogen transferred was 2.6%. The molecular weight of the extracts was in the range of 400—1500. The kinetics of the process corresponded to a first-order reaction and was determined by the rate of rupture of a covalent bond. Two components with different thermal stabilities were determined in coal; their energies of activation in coal conversion process were 30.0 and 38.2 kcal/mole, respectively, and in hydrogen-transfer rate process they were 40.0 and 45.5 kcal/mole. A semi-empirical equation for estimating the rate of the process is suggested.

1968

157. Aronov, S. G., M. G. Sklyar, Yu. B. Tyutyunnikov (1968) **Preparation of phenols and aromatics from coals by directed hydrogenation** (Poluchenie iz uglei fenolov i aromaticheskikh soedineniy metodom napravlennoi gidrogenizacii) *Kompleksnaya khimikotekhnologicheskaya pererabotka uglei*, Kiev. (in Russian)

158. Bronovets, T. M., E. M. Taits (1968) **Research on the properties of low-rank coal and the products resulting from its mild hydrogenation** (Issledovanie svoistv uglei nizkoi stadii metamorfizma i produktov ikh gidrogenizacii v myagkikh usloviyakh) *Khimiya Tverdogo Topliva*, No. 6, pp. 3 – 12. (in Russian)

Gas coals were hydrogenated in a 1-litre rotary autoclave charged with 200 g of coal particles < 0.25 mm in size. The process conditions were the following: temperature 380 – 390 °C, residence time at the final temperature 1.0 – 2.5 hours, initial H_2 pressure 55 – 80 atm. After the treatment the coals caked well and became benzene-soluble; the sulphur and nitrogen content was reduced. Benzene extracts can be used in the synthesis of phenol-type tars.

159. **Coal in 1980** *Colliery Engineering*, 1968, Vol. 45, No. 529, p. 80. (in English)

According to the European Economic Community, the likely energy consumption in Europe up to 1980 would be as follows: In the Common Market countries there will be a gradual adoption of a cheap-energy policy as a result of cheap import from outside Europe. In the United Kingdom, coal will remain the main source of energy, but oil, natural gas and atomic energy will increase their shares of the market. Elsewhere in Western Europe there will be a trend towards cheap energy, e.g. hydro-power and cheap imports of oil. In Eastern Europe, a steady development of all sources of energy seems likely, with coal, however, falling behind oil.

160. Walter, L. (1968) **Petroleum products from coal,** *Colliery Guardian*, Vol. 216, No. 5574, pp. 189—192. (in English)

The article discusses the problem of forthcoming application of coal as raw material for production of synthetic petroleum and petroleum products. A review of coal-conversion projects is presented.

161. Mirza, A., Mrs., M. A. Masood, M. M. Mallikarjunan, R. Vaidyeswaran (1968) **Studies on hydrogenation of low-temperature tar products. Hydrorefining of neutral oil. Part I.,** *Chemical Age of India*, Vol. 19, No. 4, *Petrol and Hydrocarbons*, Vol. 3, No. 1, pp. 13—17. (in English)

The hydrorefining of neutral oil (fraction boiling at 200° – 305 °C) obtained from a non-caking coal was studied in a laboratory plant at 350 —450 °C, under a pressure of 25 – 100 atm and with a space velocity of feeding of 0.2 – 2.0 h^{-1}. The stock-to-hydrogen ratio was 1:500 and the Co—Mo—Al_2O_3 catalyst was used. The studies were also carried out in a pilot plant at 340° C, 50 atm and space velocity of 1.0 h^{-1}, with a recycle gas to fresh hydrogen ratio of 80:20. The degrees of desulphurization and deazotization amounted to 77 and 53%, respectively. When the temperature of the process was increased to 400 °C, the conversion level of the stock decreased, and at 450 °C the catalyst lost its activity.

162. Eddinger, R. T., L. D. Friedman (1968) **Hydrogenation of low-temperature coal tars using iodine as catalyst,** *Fuel*, Vol. 47, No. 4, pp. 320—321. (in English)
To improve the technological properties of low-temperature coal tars, hydrogenation of the tars was carried out in the presence of I_2. The mixture was heated at 430 °C for 2 hours under H_2 at a pressure of 210 atm, with agitation. The hydrogenate was 96% liquid. The sulphur content of the tar was reduced from 0.7 to 0.14%. About 86% of the hydrogenate boiled below 350 °C. The naphthalene fraction amounted to 20%. Iodine was found to be a more active catalyst than Co—Mo or Ni—W.

163. Jackh, W. (1968) **Hydrogenation of coal** (Die Hydrierung von Kohle) *Energy und Techn.*, Vol. 20, No. 7, pp. 218—220. (in German)
The history and current state of the coal hydrogenation problem are reviewed. The economics of gasoline production by coal hydrogenation is examined; the conclusion is that the process is economically unfavourable.

164. Tengler, Sz. (1968) **The present state and trend of coal hydrogenation research** (Aktualny stan i kierunki badan nad uwodornieniem wegla) *Wiadom. gorn.*, Vol. 19, No. 7—8, pp. 223—226. (in Polish)
Investigations on coal hydrogenation carried out in the US are discussed. The purpose of these investigations is to obtain gasoline, a high energy fuel for jet-propulsion engines, and aromatic hydrocarbons. To reduce the hydrogen consumption in all the processes, the coal is not liquefied completely.

165. Tengler, Sz. (1968) **Some trends of new research methods of coal processing in the US and their prospects for Poland** (Niektore kierunke badan nad nowymi metodami przerobki węgla w Stanach Zjednoczenych A. i perspektywy dla Polski) *Gospod. paliw. i energ.*, Vol. 16, No. 8, pp. 2—6. (in Polish)

166. Olpinski, W., M. Ihnatowicz (1968) **Production of liquid fuels from coal in the US** (Otrzymywanie paliw plynnych z węgla w Stanach Zjedhoczonych AP) *Przegl. gorn.*, Vol 24, No. 11, pp. 533—540 (in Polish)
This is the report of a Polish delegation that visited the US in 1968. A Consolidation Coal Company pilot plant for the production of liquid fuel, mainly gasoline, by the method of incomplete coal hydrogenation is described. Its capacity is 1 ton/h. The

greatest difficulties are in the separation of the unreacted residue from the extract on the drum filter at 320 °C and 10 atm. A belt filter is now being developed. The Hydrocarbon Research Company has carried out hydrogenation of coal suspensions in oil in a fluidized catalyst reactor in the plant with a capacity of 3 ton day. The FMC Corporation is developing a 4 stage fluidized-bed process with counter-current pyrolysis of the coal. As a result, liquid products and coke of high calorific value will be obtained.

167. Job, J. G., D. G. Nicklin (1968) **The production of liquid fuels from coal in Central Queensland,** *J. Instn. Engrs. Austral.*, Vol. 10, No. 10—11, pp. 253—256. (in English)

Possibilities of liquid fuel production from Queensland coal are discussed. Two methods are presented: the Fischer-Tropsch synthesis and direct hydrogenation. Data on the operation of the Sasol Company plant (South Africa) were compared with the results of testing the H-Coal process — the process of direct hydrogenation of coal in a paste at 400° — 500 °C and 100 — 200 atm. The latter process was found to be more economical.

168. Hellwig, K. C., M. C. Chervenak, E. S. Johanson, H. H. Stotler, R. H. Wolk (1968) **Convert coal to liquid fuels with H-coal,** *Chem. Engng. Progr.*, Sympos. Ser., Vol. 64, No. 85, pp. 98—103. (in English)

Test results on the continuous catalytic hydrogenation of coal for liquid fuel production at moderate pressure in a special feed-through reactor. The testing was carried out by Hydrocarbon Research, Inc., in 1965—1967 in pilot plants with capacities of 12 kg/day and 3 ton/day. For final data it was planned to build a plant with a capacity of 250 ton/day. The process was supposed to be commercialized in 1970. Coal is ground and mixed with oil to form a paste, which is then fed into the reactor. The yield of products from Illinois No. 6 coal is (weight %): C_1-C_3 gases, 10.2; liquid products, 71.0; coal residue, 10.7; H_2S, water, and NH_3, 14.0. The conversion rate of the coal is 89.3%. Hydrogen is produced by steam conversion of natural gas. The economics of the process for plants with various capacities are given.

169. Wu, W. R. K. and F. H. Storch (1968) **Hydrogenation of coal and tar,** *U.S. Bureau of Mines Bulletin No. 633*, 195 pp., ill., Washington, D.C. (in English)

This bulletin contains a review of investigations on hydrogenation of coal and tar carried out in the US and other countries. There are seven chapters, each with extensive bibliography. The economics of the process and the latest investigations on hydrogenating coal into a rich gas and liquid fuel are discussed in detail.

170. Cudmore, J. F. (1968) **Solvent extraction of some Australian coals.** *Mech. and Chem. Engng. Trans. Instn. Engrs. Austral.*, Vol. 4, No. 2, pp. 173–180. (in English)

The thermal dissolution of coals of various rank in anthracene oil is studied in detail with the aim of producing an ashless coal extract, called Coalex. The latter can be used to make electrodes for aluminium extraction, metallurgical coke, carbon black etc. Under optimum conditions, a 1 : 4 mixture of 40–100 mesh coal and anthracene oil boiling above 330° C was charged into a 4-litre vibrating autoclave and heated to 390–400° C at a hydrogen pressure of 130 atm. The experiments showed that semi anthracite was not suitable for this process. Coal and brown coal yielded 50–70% extract and 20% insoluble residue, the hydrogen consumption was 5%, and the degree of solvent regeneration was 86%. The clean coal obtained was a brittle vitreous product like solid pitch and contained 0.1–0.3% ash on the dry basis, 34–38% volatile matter on the combustible matter basis, 80–92% carbon, and 4.6–5.2% hydrogen.

171. Bloomer, W. J., S. W. Martin (1968) **Coal solvation with non-hydrogenated solvent in the absence of added hydrogen.** *U.S. Pat. No. 3,378,638* (Cl. 208-8); Appl. 25 January 1965, publ. 23 April 1968. (in English)

A method is described for obtaining solutions of practically ash-free carbonaceous matter from coals and brown coals equally with fusain and mineral substance. Coal (less than 100 mesh) is mixed with a highly aromatized solvent boiling above 340 to 450° C in the ratio of 1 : 2 to 1 : 3. The mixture is heated at 315 to 455° C and 1–8 atm until the complete decomposition of the coal results in a suspension of fusain and mineral substance in the solvent. Then the solution is cooled to 200°–370° C and the suspended solid fraction is separated.

1969

172. Val, C., L. del Jose (1969) **Catalytic refinement under pressure of a light oil of coal tar consisting mainly of benzene** (Refino catalitico bajo presion del aceite ligero del alquitran de hulla) *Ion*, Vol. 20, No. 253, pp. 479–484. (in Spanish)

In order to produce high-octane motor fuel, refinement is performed at a temperature of 320° C and pressure of 38 atm of hydrogen-containing gas (coke-oven gas or city gas). The yield of the refined product is 98%, whatever the initial material. The refined product contains no sulphur or sulphur compound and can be used as a motor fuel without any additional distillation. Fractional distillation of the refined product provides commercial products of a purity unattainable by sulphuric acid refining. The refined product contains more xylene and C_6H_6 than the product of sulphuric acid refinement. To obtain chemically pure products (benzene, toluene and xylene), the partial pressure of H_2 is maintained at 18 atm.

173. Appel, H. R., I. Wender and R. D. Miller (1969) **Dissimilar behaviour of carbon monoxide plus water and of hydrogen in hydrogenation.** *Symposium on Chemicals and Oil from Coal, Dhanbad, India*, 6 to 8 December 1969. (in English)

The authors demonstrate the feasibility of using a mixture of carbon monoxide and water for hydrogenating coal to yield an oil which can be converted to more volatile fuels by known techniques. In autoclave experiments at 315 atm and 380° to 400° C, conversion of lignite was nearly 95%, although the conversion of the organic part of bituminous coal did not exceed 75%. Investigations showed that when (CO + H_2O) was used at 380° C the reaction was complete in 10 min, whereas with hydrogen gas, only 50% conversion takes place in this time. Carbon monoxide and water effectively solubilize low-rank coal because: (1) activated hydrogen is formed; (2) carbon monoxide is able to cleave certain types of bonds or inhibit condensation reactions that yield benzene-insoluble materials.

Mild hydrogenation of a number of pure compounds in the presence of carbon monoxide, water and hydrogen was studied. The ease of conversion of cellulose and glucose to benzene-soluble products with carbon monoxide and water suggests that the same structural units as those derived from carbohydrates exist in low-rank coals.

174. Mukherjee, D. K., J. K. Sama, S. K. Mukherjee, P. B. Chowdhury and A. Lahiri (1969) **Effect of reaction variables on hydrogenation of Baragolai (Assam) coal.** *Symposium on Chemicals and Oil from Coal*, Dhanbad, India, 6 to 8 December 1969. (in English)

In the hydrogenation of Assam coals, the consumption of hydrogen depends most strongly on the reaction temperature. For the range examined, the authors observed a linear rise in conversion of the organic component with reaction time. It was found that 90% of the total sulphur in the coal could be removed. The production of gaseous hydrocarbons decreased with pressure and increased with temperature.

From the conversion data (hydrogen consumption, gas formation, hydrogen enrichment of the liquid products, yield of distillate boiling between 200°C and 300°C and the amount of saturates in the distillate) the authors deduce that at 250 atm reaction pressure, the most favourable temperature range for hydrogenation is 400–420°C. Under these conditions the middle oil distillate yield is maximum. The loss of carbon as gas is somewhat higher at temperatures above 400°C but this is compensated by increased conversion of the organic component, increased yield of middle boiling distillates containing higher amounts of saturates and decreased residence time.

175. Hiteshue, R. W., W. Kawa and C. O. Hawk (Pittsburgh Coal Research Center, Pennsylvania, USA) (1969) **Low pressure hydrogenation of coal in a continuous pilot plant.** *Symposium on Chemicals and Oil from Coal*, Dhanbad, India, 6 to 8 December 1969. (in English)

    Highly volatile bituminous coals were hydrogenated at 465°C, 136 atm and at a space velocity of 9–22 lb/h. The following catalysts were used: molybdenum (0.1–0.5%) and iron (1–2%). The total conversion of coal was 90%. The yield of distillate oil boiling below 325°C was 56%. The heavy oil produced was just sufficient for the paste preparation. A large reactor volume was used in the plant, but the cost of the reactor is a minor item.

176. Mukherjee, D. K., P. Samuel and A. Lahiri (1969) **On the kinetics of coal hydrogenation.** *Symposium on Chemicals and Oil from Coal*, Dhanbad, India, 6–8 December 1969. (in English)

    The authors re-analyse two sets of data obtained by the U.S. Bureau of Mines on the hydrogenation of Rock Springs and Pittsburgh coals. They conclude that hydrogenation reactions cannot be represented by an elementary mechanism. They point out that the data are better interpreted if the correction factor for benzene insolubles is not included. The negative values obtained in certain cases if this correction is taken into account suggest that the correction does not have adequate validity. The observed change in order from a lower to a higher value, resulting in a decrease in rate as the reaction progresses, is interpreted as the result of an internally generated interference in the system, e.g. opposing reactions that result in the formation of benzene insolubles or other components that resist hydrogenation. The over-all hydrogenation reaction consists of a series of parallel and consecutive reactions like pyrolysis, depolymerization, hydrogenation, hydrogenolysis, dehydrogenation, condensation etc. Depending on the reaction conditions and concentration of the reactants, one or more of these reactions may predominate and thus determine the end result.

177. Brahmachari, B. B., M. R. Saha, P. B. Chowdhury and A. K. Ganguli (1969) **Catalytic hydrogenation of low-temperature tar fractions,** *Symposium on Chemicals and Oil from Coal*, Dhanbad, India, 6 to 8 December 1969. (in English)

The structural changes that take place during the hydrogenation of a (200 – 350 C) fraction on a molybdenum sulphide catalyst are described. The experiments were conducted in a 250-cm³ continuous bench scale unit at 105 atm and at various temperatures and liquid space velocities. Data on carbon distribution and ring analysis are given.

The hydrogenated product did not contain any olefinic hydrocarbons and showed an increase in the paraffinic carbon at the expense of the aromatic carbon, the naphthenic carbon content remaining more or less at the same level. It is inferred that the conversion of aromatic to paraffinic carbons takes place via the formation of naphthenes.

The smoke point of the kerosene fraction was below 20 mm and therefore did not conform to ISI (Indian Standards Institution) specifications. The diesel fraction, however, had a diesel index of more than 45 and could be used as high-speed fuel.

178. Mirza, A., M. A. Masood, M. M. Mallikarjunan and R. Vaidyeswaran (Regional Research Laboratory, Hyderabad, India) (1969) **Possibilities of producing middle distillates from low-temperature tar fractions,** *Symposium on Chemicals and Oil from Coal*, Dhanbad, India, 6 to 8 December 1969. (in English)

Hydrogenation of a neutral oil fraction (200°–300° C) of low-temperature tar was carried out in a 60-cm³ bench-scale unit as well as in a 1-litre pilot plant using two types of commercial catalysts: cobalt molybdate on alumina, and tungsten sulphide-nickel sulphide on alumina.

1. Using cobalt molybdate on alumina for hydrorefining, about 55% and 40% of sulphur and nitrogen, respectively, could be removed. Although the aromatic content could not be brought down sufficiently, the product conformed to grade II diesel as per ISI specifications.

2. With tungsten sulphide-nickel sulphide on alumina and at a space velocity of 0.2–0.3, both hydrorefining and saturation could be accomplished. The aromatics could be reduced to less than 20%.

3. In the two-stage operation a product with a smoke point of 22 mm could be obtained at unit space velocities for both operations by first hydrorefining with cobalt molybdate and subsequently subjecting the product to hydrogenation in a second stage with tungsten sulphide–nickel sulphide catalyst.

4. Recycling of tail gases helped to maintain H_2S concentrations in the gas, thus advantageously keeping the catalyst in sulphide form.

5. The hydrogen consumption was estimated to be 170 m^3/ton and 150 m^3/ton respectively for the 1st and 2nd stages of operation.

6. The use of the tungsten sulphide-nickel sulphide catalyst in two stages was found to be more effective than in a single-stage operation.

179 Morparia, N. P. and S. Sarkar (Indian Institute of Technology, Bombay, India) (1969) **Studies on low-temperature coal-tar hydrogenation. Pt. 1. Hydrogenation of neutral middle distillates in a continuous bench scale unit,** *Symposium on Chemicals and Oil from Coal,* Dhanbad, India, 6 to 8 December 1969. (in English)

The paper deals with the hydrogenation of neutral oil fractions boiling in the range of 200°–350° C to produce diesel oil. With a commercial cobalt-molybdenum on alumina catalyst (3 × 3 mm) under the optimum conditions of 400° C, 100 kg/cm^2 and 0.5 liquid space velocity, the yield of the desired product (200°–350° C) was 68.6%. At lower temperatures the degree of hydrogenation was lower, while higher temperatures resulted in excessive cracking. Better results were obtained at lower space velocities. The cetane number of the product was 35.5 and the smoke point was 13 mm, which satisfies the ISI specifications for diesel oil and kerosene.

180. Mukherjee, D. K., J. K. Sama, P. B. Chowdhury and A. Lahiri (1969) **Hydrogenation of coal with iron catalysts,** *Symposium on Chemicals and Oil from Coal,* Dhanbad, India, 6 to 8 December 1969. (in English)

The object here was to develop inexpensive iron-based catalysts having activities for coal hydrogenation matching those of tin and molybdenum catalysts. Iron oxide is more active in combination with sulphur. At 400° C and 100 atm the conversion, yield and quality of products were comparable to that of the molybdenum catalyst. Although the concentration of iron catalyst is higher (nearly 3.7 times that of molybdenum) it is believed to be within practical and economic limits.

Under the conditions of hydrogenation, the most stable form of iron in the presence of hydrogen and hydrogen-sulphide is ferrous sulphide. The catalytic activity of iron increases with increasing S/Fe ratio or H_2S partial pressure.

181. Hill, G. R. and S. A. Qader (Fuels Engineering Department, University of Utah, USA) (1969) **Liquefaction of Utah coals,** *Symposium*

on Chemicals and Oil from Coal, Dhanbad, India 6 to 8 December 1969. (in English)

The conversion of coals to liquid fuels is respectively 35 %, 96 % and 82 % by pyrolysis, solvent extraction and hydrogen. The coal hydrogenation process is the most promising. The investigations demonstrate the feasibility of producing gasoline as the main product, with diesel oil and high BTU gas as the principal by-products, by the hydrogenation of dry, impregnated caking coals in an entrained bed type of reactor. To avoid agglomeration, coke is mixed with the coal feed. Best results were obtained at 515 °C, 136 atm, and with 15 % $SnCl_2$ catalyst.

182. Develotte, J., M. Mazza, P. Payen (1969) **Hydrogenation of coal and its derivatives. Comparative study of model substances** (Dégradation ménagée par hydrogenation du charbon et de ses dérivés. Etude comparative de substances modèles) *Bull. Soc. chim. France*, No 1, pp. 341–348. (in French)

Hydrogenation of coal, coal-tar and individual substances was carried out at 340°–430° C and under an H_2 pressure of 200 bar with MoS_2 catalyst. Investigation of the hydrogenates showed that the structural units of coal consisted of polycyclic aromatic hydrocarbons, among which there were pyrene structures and some structures containing 6–7 cycles. In low- and high-temperature tars C_{20}–C_{40} polycyclic condensed hydrocarbons were found.

183. Hausigk, D., G. Kölling, F. Ziegler (1969) **The dissolution of bituminous coal in tetrahydroquinoline. Part 2** (Lösen von Steinkohle in Tetrahydrochinolin. II.) *Brennstoff-Chemie*, Vol. 50, No 1, pp. 8–11 (in German)

When a young coal (0.06 mm) with organic component containing 40.6 % volatiles, 81.3 % carbon, 4.91 % hydrogen and 10.0 % oxygen was heated in a rotary autoclave with 30 parts of 1,2,3,4-tetrahydroquinoline at 300 °C for 1–16 h, the yield of the extract (50 % of which is pyridine-soluble) amounted to 90 %. The dissolution of the coal was accompanied by hydrogenolysis at the expense of hydrogen donor with a loss of 40.6 % of the oxygen as water. Similar results were obtained with a coal containing 35.4 % volatiles; 99 % of its extract was pyridine soluble. The molecular weight of the extract was 593, implying that the extract consists of bi- and tri-cyclic aromatic compounds bonded with aliphatic or hydro-aromatic bridges.

184. Dent, F. J., B. H. Thompson, H. L. Conway (The Gas Council) (1969) **Hydrogenation of coal to give methane-containing gases.** *British Patent No. 1,154,322* (Cl. C 5E, C 10b). Appl. 6 October 1966, publ. 4 June 1969, 5 pp. (in English)

Hydrogenation of caking coal to produce CH_4 containing gas was carried out by introducing a fine grained coal into a circulating fluidized bed of non caking material at 600 – 1,000 °C, preferably 700 – 900 °C, in the presence of H_2 at a pressure of 25 – 100 atm. This method of introducing the coal into the fluidized bed solves the problem of heating it and eliminates caking and agglomeration. The fine grained semicoke obtained in the process is used for hydrogen production.

185 Rader, S. A., G. R. Hill (1969) **Synthetic liquid fuel from coal** (Synthetisches Öl aus Kohle) *Brennstoff-Chemie*, Vol. 50, No. 6, p. 167. (in German)

The results of hydrogenating high volatile coal from Flöz and high temperature coke at a pressure of 140 atm over ammonium molybdate, tin chloride and zinc chloride catalysts are given. The catalyst used for hydro-refining of the liquid fuel obtained contained 3 % cobalt oxide and 12 % molybdenum oxide on Al_2O_3. The catalyst for hydrocracking consisted of 6 % NiS and 19 % WS_2 on SiO_2-Al_2O_3.

Experiments on low temperature carbonization and on thermal and catalytic hydrogenation were carried out in a 1-litre vibrating autoclave. A reactor of 50-mm diameter was used in the experiments on hydrogenating pulverized coal. The experiments on hydrorefining and hydrocracking of the liquid product were conducted in a continuous reactor. The reactor was 1.0 m in height and 19 mm in diameter.

The experiments showed that the application of ammonium molybdate increased the yield of liquid products and decreased the yield of gas whereas the application of tin and zinc chlorides decreased the yield of liquid products and increased the yield of gas.

The hydrogenation of a liquid-phase hydrogenate in a typical laboratory reactor yielded 30 % gasoline with an octane rating of 80 and 49 % diesel fuel of index 52. The hydrocracking resulted in 70 % gasoline of 82 octane and 12 % diesel fuel of index 50.

186 Friedman, L. D., R. T. Eddinger (1969) **Hydrogenation of coal tar.** *U. S. Patent No. 3,453,202*, Cl 208-40 (C 10d, C 10c), Appl 28 November 1966, publ 1 July 1969 (in English)

The process is conducted in a flow of an equimolar mixture of H_2 and CO in the presence of I_2 catalyst at 70 – 210 atm and 370 to 480 °C. For example, an autoclave is charged with 100 g of a low temperature coal tar, obtained during pyrolysis in a fluidized coal, and 2 g of sublimated I_2. The initial pressure of H_2 + CO gas is 215 atm. The autoclave is rapidly heated to 400 °C for 3 hr.

the pressure rises to 315 atm and then drops to 280 atm. Then the autoclave is cooled. The yield of oil with viscosity less than 100 centistokes is 87% of dry coal tar.

187. Leatle, K. A. (1969) **Solid fuel research.** *Industr. and Process Heat,* Vol. 9, No. 7, pp. 26–29 (in English).

Solid fuel research being conducted at the experimental station in Stoke Orchard is reviewed. Parallel with fundamental research on combustion in a fluidized bed, extraction of chemicals from coal tar and production of metallurgical coke, investigations are being conducted on non-fuel uses of coal — solvent extraction and hydrogasification at high temperatures and pressures. There is also some research on the expanded utilization of coal treatment products in the building industry.

188. Rader, S. A., R. A. Haddadlich, L. L. Anderson, G. R. Hill (1969) **Coal can also yield liquid fuels.** *Hydrocarbon Process.* Vol. 48, No. 9, pp. 147–152 (in English).

Laboratory experiments on hydrogenation of coal to liquid fuel are described. To avoid caking, the coal, which contained 6.7% ash and 46.8% volatile matter on a dry basis, was mixed with the same quantity of semicoke. The mixture was pulverized to a particle size of 250 μm, added to 15% $SnCl_2$ and passed in a H_2 flow heated to 300°C through a flow reactor. Under optimum conditions (at 515°C and 140 atm) the coal conversion was 75%. At 6% hydrogen consumption the yields were (wt %): gasoline, 30; diesel fuel, 5; gas containing 84% of CH_4, 35; semicoke, 30.

189. Hellwig, K. C., S. B. Alpert, E. S. Johanson, R. H. Wolk (1969) **H-Oil- and H-Coal-processes** (H-Oil- und H-Coal-Verfahren) *Brennstoff-Chemie,* Vol. 50, No. 9, pp. 263–268 (in German).

The use of a fluidized catalyst for hydrogenating petroleum fractions, coal tar and coal is discussed. A number of high volatile and brown coals were examined. The yield of products from Illinois coal was (wt %): C_1–C_3 gas, 10.2; liquid products, 71.0; unconverted coal, 10.7; H_2O, H_2S and NH_3, 14.0.

In the article the economics of the process for treating 27,000 tons of gas coal per day is given. The total investment is $300.7 million.

Product output, ton/day

Gasoline	7 900
Light fuel oil	4 630
Heavy fuel oil	380
Coal residue (heating capacity 4.35 × 10^6 kcal/ton)	5 900
Sulphur	550
Ammonia	205

Input

Dry coal, ton	27,000
Electric energy, kW	244,000
Water, ton/hr	2,080
Chemicals and catalysts, dollars/day	44,000
Labour, men	211
Natural gas, m³ per day	3.900 × 10^6
Production of hydrogen unit, m³ per day	20.200 × 10^6

190. Struth, B. W. (Chem. Systems Inc., New York, USA) **Chemicals from coal,** *UNIDO. Interregional Petrochemical Symposium on the Development of the Petrochemical Industries in Developing Countries.* Baku, USSR, 20–31 October 1969. ID/WG. 34/53. Rev. 1 15 February 1970. (in English)

The paper discusses the economics of a chemical refinery using coal as the raw material instead of crude oil. The author considers this direction to be more economically favourable. The material balance of the process is given with regard to all the processing stages of the products.

The refinery production is 100,000 bbl/day of liquid products.

1970

191. **Eryomin, I. V., I. L. Ettinger** (1970) **Coal science — state of the art (Nauka ob ugle sostoyanie i problemy)** *Vestnik Akademii nauk SSSR,* No. 3. (in Russian)

192. **Angelovich, J. M., G. R. Paston, H. F. Silver** (1970) **Solvents used in the conversion of coal.** *Ind. and Engng. Chem., Process Design and Developm.*, Vol. 9, No. 1, pp. 106–109. (in English)

Selection of the optimum solvent for converting coal to liquid products by solvent extraction or hydrogenation was studied. Fourteen solvents were examined, including *n*-hexane, benzene, cyclohexane, phenol, tetralin, coal-tar fractions and hydrogenated coal oil. The nonpolar solubility parameter was estimated for each solvent. The solvents were examined experimentally for the hydrogenation of a sub-bituminous coal with organic component containing 74.6% carbon, 5.3% hydrogen and 18.3% oxygen in a rotating autoclave. The conditions of the experiments were: coal charge 100 g, 0.5% of ferrous ion catalyst, initial hydrogen pressure 140 atm, temperature of 440° C maintained for 3 hr.

The coal-conversion rate, which increased with increase in the nonpolar solubility parameter, was defined on the basis of the weight of insoluble residue. The results of the experiments showed that tetralin and the coal-tar fraction boiling at 240° C appeared to be most effective in the liquid-phase hydrogenation of coal

193. **Kotowski, W., N. Paterok, Z. Nowak** (1970) **Extraction (peptization) of Polish hard coal** (Ekstrakcja (peptyzacja) krajowego węgla kamiennego) *Koks, Smola, Gaz,* Vol. 15; No. 1, pp. 9–13. (in Polish)

In the experiments, coal of 0.1 mm grain size containing 5.4 % ash on the dry basis, 77.3 % C and 5.1 % H was used. The extraction was carried out in a continuous laboratory plant. Tetralin, anthracene oil and heavy oil obtained from the extract were tested as solvents. Coal was pasted and fed into the reactor at a rate of 0.6 ton/m^3 per hour at 100 atm and 450 °C. Under these conditions the yields of the extract and gas were 61.2 % and 9.6 %, respectively. The extract characteristic was: specific gravity, 1.09; fraction topping below 300 °C, 22.8 %.

194. Kölling, G. (1970) **Dissolution of coal at temperatures less than 200 °C** (Auflösen von Steinkohlen bei Temperaturen unter 200° C) *Angewandte Chemie*, Vol. 82, No. 2, p. 87. (in German)

Coal solubility greatly increased when the coal was alkylated with chlorides of higher fatty acids in the presence of $AlCl_3$ at 50 °C. The corrected molecular weight of the extract appeared to be 3000.

195. Jackh, W. (1970) **Problems in the hydrogenation of coal** (Probleme der Hydrierung von Kohle) *Erdöl und Kohle*, Vol. 23, No. 6, pp. 334 to 337. (in German)

Effective coal hydrogenation depends on successful solutions to the problem of producing cheap hydrogen, eliminating ash and developing the high pressure equipment. The conversion of brown coal and coal into gasoline was found to demand 2,200 m^3 and 2,800 m^3 of hydrogen per ton of the final product, respectively. Calculations of H_2 production from natural gas and petroleum as well as from hydrogenate and slurry processing are given. The possibility of increasing the capacity of the hydrogen plants by increasing the reactor size is discussed.

The author believes that hydrogenating coal to gasoline in the FRG is not justifiable because of the high coal price of $15/ton. In this case the gasoline price is $75/ton whereas gasoline obtained from petroleum costs $25/ton. In countries where coal prices are lower, as in the US ($4/ton), hydrogenation of coal can be economically favourable. Gasoline from coal hydrogenation in large plants, where hydrogen obtained from natural gas is used, is estimated in the US at 11 ¢/gal ($44/ton).

196. Krichko, A. A. (1971)* **Hydrogenation of solid fuel** (Gidrogenizaciya tverdogo topliva) *Khimiya tverdogo topliva*, No. 2, pp. 31—39. (in Russian)

A review of the coal hydrogenation problem.

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* [This paper, which is by one of the consultants, complements the present publication and is therefore retained in the 1970 listing.]

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SUBJECT INDEX

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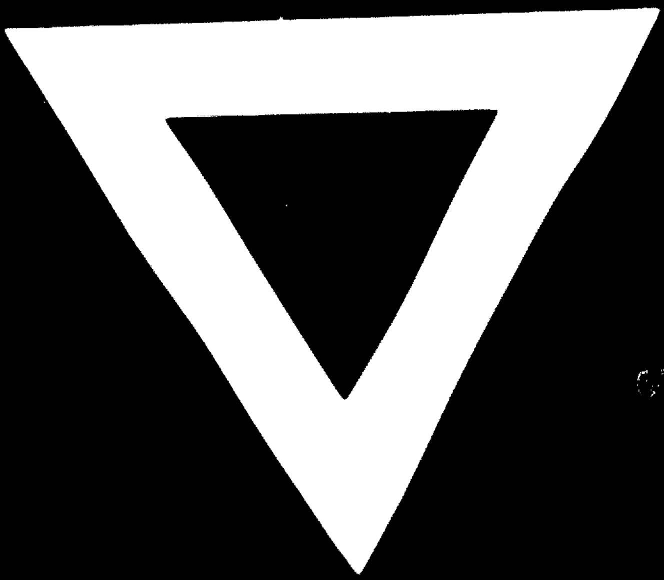

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