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UNITED STATES  
DEPARTMENT OF THE INTERIOR  
BUREAU OF MINES  
OFFICE OF SYNTHETIC LIQUID FUEL 1404  
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W. M. Sternberg  
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PREPARATION OF SLIGHTLY HYDROGENATED COAL

(Addition to the table on experiments for preparation of slightly hydrogenated coal, April 21, 1943, No. 213, 811).

SUMMARY

The processes known today for the preparation of slightly hydrogenated coal are being discussed. It has been established that as the processes are carried out they fail to produce the optimum results in the production of hydrogenated coal for the extracting hydrogenation, which results in an unfavorable balance in the computation of costs. Better operating results are foreseen when larger amounts of gases are used, or by simultaneous production of asphalt-freeer oils in larger amounts.

The addition of coal in the hydrogenation of low temperature tars offers the possibility to produce in the same reaction space some briquetting material (slightly hydrogenated coal) without important effects upon the hydrogenation of tar, and this will reduce the price of the briquetting material.

The operations with a cold catchpot is unfavorable from the heat exchanger angle, and the catchpot temperature must be investigated as related to the quality of the let-down.

Application of solid and ash-containing slightly hydrogenated coals as briquetting materials requires:

1). That the slightly hydrogenated coal has softening points not exceeding 70 - 75°C, with adequate cohesion qualities.

2). The cost of the prehydrogenated coal must not exceed the cost of the originally used amounts of pitch.

These preliminary requirements specify for the production process of slightly hydrogenated coal, that:

1). In the slightly hydrogenated coal a high asphalt and gum content is desired with, however, a low ash and solids content.

2). The process must permit to maintain the pasting oil equilibrium during the production of briquetting materials with a softening point of 70 - 75°.

3). When the slightly hydrogenated coal is the principal production product (e.g. from coal alone), which is the case when only unimportant amounts of asphalt-free oils are obtained in addition to the coal, the slightly hydrogenated coal must carry the total production costs, i.e. the process must be so cheap that the price of pitch is not exceeded.

4). Such processes are adopted to the production of slightly hydrogenated coals, in which such amounts of asphalt-free oils (gasoline, middle oil, fuel oil) are formed together with briquetting materials of the required quality, that the total costs of the process can be charged up to the slightly hydrogenated coal which it may carry from considerations of prices.

Chemical Advantages of the Different Processes

The extraction and the extracting hydrogenation will be the only processes which will be at first discussed. The coal is pasted in these processes with middle oil, with little or no use of hydrogen, avoiding excessive cracking and with the production of filterable products. If filtration is omitted in these processes we obtain slightly hydrogenated coals.

A). Broche extraction method. The pasting oil equilibrium is not reached at the softening point of the slightly hydrogenated coal of over 220°C. The process must therefore be disregarded.

B). Extraction hydrogenation at 300 atm (Unde). The pasting oil equilibrium is not reached in this method of operation when the softening point of the partially hydrogenated coal is higher than 100 - 120°C. This process must again be considered unsuitable.

C). Extraction hydrogenation at 700 atm. (Ludwigshafen). The pasting oil equilibrium is reached with the younger coals from Lower and Upper Silesia with a suitable softening point of the partially hydrogenated coal of 70 - 75°C and the production of a small excess of asphalt-free oil. The process is satisfactory from a chemical standpoint. The cost of the briquetting material is, however, probably higher than the cost of pitch.

It has already been mentioned that the aims of these processes was not the production of partially hydrogenated coal but rather of valuable extraction. The operating conditions have been selected with an eye to these requirements. The filtration ability may be disregarded in the preparation of slightly hydrogenated coal. This offers the chance to conduct the process with larger amounts of gas. This change will presumably cause an easier reaching of the pasting oil equilibrium with a simultaneous production of an important amount of more valuable asphalt-free oils, as well as an increase in the throughput.

Computations of cost of slightly hydrogenated coal from coal alone, i.e. the comparison of costs of such hydrogenated coals with the slightly hydrogenated coal from a combination process (e.g. coal + low temperature tar) must perhaps be done by consideration of the following processes:

1). Extraction hydrogenation with large amounts of gas at 300 atm.

2). Extraction hydrogenation at 700 atm with large amounts of gas and high throughput, i.e. with a simultaneous production of considerable amounts of gasoline and fuel oil as the principal products, with

briquetting materials as a byproduct. There is at present no experimental basis for this method.

The cost estimates based on experimental results for the extracting hydrogenation with small amounts of gas produce too unfavorable a picture.

D). The process with the addition of low temperature tar.

If middle oil-containing low temperature tar be added during the hydrogenation extraction with higher amounts of gas, replacing middle oil and return hot oil let-down, a simultaneous hydrogenation of coal and refining of tar take place. In comparison with the extraction hydrogenation when small amounts of gas are used (cases 3 and 1 in the table) practically equal amounts of coal pass through the high pressure space in the two cases and in addition in the tar process about the same amount of fresh tar as of pasting oil.

	Case 1 Extraction hydrogenation at 700 atm. middle oil	Case 3 Extraction hydrogenation at 700 atm. with low temperature tar, containing middle oil ( 42 percent)
Cubic meters of inlet gas/kg paste	0.53	1.04
Paste thruput	2.41	2.20
Pure coal thruput	0.85	0.82
Tar thruput	-	1.33
Production: gasoline	0.05	0.10
middle oil (new)	0.09	0.07 (0.17)
heavy oil	-	0.20
briquetting material	0.65 (77° K.S.)	1.08 (74° K.S.) *
Utilization of organic solids	92.8 percent	89.4 percent
Percent of H <sub>2</sub> consump- tion for the hydroge- nation to pure coal.	2.6	2.6 2)
Percent H <sub>2</sub> consumption for the tar refining/tar	-	1.1 3)

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- 1). Quality of the fuel oil evidently insufficient
  - 2). The figure assumed on the strength of Case 1
  - 3). Calculated.
- \* Kramer-Sarnow
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The experiment of the extraction hydrogenation with middle oil and higher amounts of gas are unavailable and the comparison here given is not strictly applicable. One may, however, readily see that the major part of the tar process is the briquetting material and only 76 percent of the tar is obtained as an asphalt-free oil, part of which is derived from coal. The same high pressure volume produces in case 3 about double the amount of gasoline and middle oil, with some heavy oil added to it. 1). A hydrogen consumption of about 1.1 percent can be calculated for the refining of the tar. The utilization of the organic solids is somewhat poorer in case 3, but this is compensated with respect to the quality of the briquetting material (solids content) by having it contain some tar constituents. This process has evidently no particular advantages because no important proportion of the tar is converted into asphalt-free oil.

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A more favorable picture is obtained when topped tar is used for pasting (extraction means).

	Case 1 Extraction hydrogenation 700 atm. with middle oil for production of briquetting materials	Case 4 Extraction hydrogenation 700 atm with topped low temperature tar for production of gasoline + middle oil + briquetting materials	Case 5 Extraction hydrogenation 700 atm with topped low temperature tar for production of gasoline, middle oil and fuel oil + briquetting materials
Cubic meters of inlet gas/kg paste	0.53	1.15	1.31
Paste thruout	2.41	2.20	2.19
Pure coal thruout	0.85	0.66	0.67
Tar thruout	-	0.55	0.73
Production:			
gasoline	0.05	0.09	0.12
middle oil (new)	0.09	0.47(0.45)	0.40 (0.37)
heavy oil	-	-	0.20
briquetting mat.	0.66(77°K.S)	0.53(66°K.S)	0.54(55°K.S.)
Utilization of organic solids	92.8	89.2	88.6
Percent hydrogen consumed for hydrogenation referred to pure coal	2.6	2.6 <sup>2)</sup>	2.6 <sup>2)</sup>
Percent hydrogen consumed for hydrogenation of tar	-	3.9 <sup>3)</sup>	3.5 <sup>3)</sup>

- 1) . Quality of the fuel oil evidently insufficient  
2) . The figure assumed on the strength of case 1.  
3) . Calculated  
\*) . Kramer-Sarnow.

The comparison is again not strictly applicable for the same reason as given above. In practically the same reaction volumes a little less coal is put through, producing accordingly less briquetting material by the additional tar thruput, which results in the production of nearly double the amount of gasoline, a 5-fold amount of middle oil or the 4-fold amount of middle oil with some appreciable amounts of heavy oil 1). The reaction space in this process is used simultaneously for the hydrogenation of tar and slight hydrogenation of coal. In fact, in case 4, a 1,000 kg addition of tar resulted in the production of about 1,000 kg of gasoline + middle oil (part of which is produced from coal) i.e. 74 percent of the dry coal reappears in the briquetting material.

If tar alone were used with a thruput of 0.55, and coal alone with a pure coal thruput of 1.0, a 40 percent saving in the reaction space would be produced. This process appears to permit an appreciable lowering of the production costs of the slightly hydrogenated coal in comparison with the middle oil extraction. The proper production costs of the briquetting material are obtained when we base the computations on the hydrogenation of tar to which coal had been added.

Conditions are similar when preheater tar is used instead of the topped tar. The experimental basis for the computation for the production of gasoline, fuel oil and briquetting material from coal and tar with the production of gasoline + fuel oil from tar is shown in cases 6 and 7.

- 1). quality of the fuel oil evidently insufficient
- 2). the figure assumed on the strength of case 1
- 3). calculated.

	Case 6 Extraction hydrogenation 700 atm used with precooler tar for production of gasoline + middle oil + fuel oil + briquetting material	Case 7 gasoline + middle oil + fuel oil from precooled tar at 700 atm
m <sup>3</sup> of inlet gas/kg paste	1.31	1.59
Paste thruput	2.22	1.10
Pure coal thruput	0.69	-
Tar thruput	0.74	0.73
Production: gasoline	0.09	0.07
middle oil (new)	0.34 (0.29)	0.31 (0.26)
heavy oil	0.19	0.24
briquetting mat.	0.61 (82°C K.S.)	-
Utilization of organic solids	84.0	-
Percent hydrogen utilized for slight hydrogenation/ pure coal	2.6 2)	-
Percent hydrogen utilization in the tar hydrogenation/tar	3.4 1)	3.4

With the same tar thruput and in the same reaction space, 0.69 additional coal thruput is obtained in case 6 with production of briquetting material. A comparison of the results with those in the processes with the tar alone shows the same total production of asphalt-free oils of 0.62. One part of the precooled tar is used up in forming briquetting material, while coal produces additional gasoline and middle oil. The softening point of the slightly hydrogenated coal, 82°C, is too high, and a 70 - 75°C softening point for the combination case 6 and will require a small reduction in the production of asphalt-free oil. There is a slight amount of uncertainty about the process for tar alone (case 7) in the operations being carried out at a 4°C lower temperature than in case 6.

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- 1). From case 7
  - 2). Calculated
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The values calculated backwards for the slightly hydrogenated coal (on the basis of case 7), gives a hydrogen consumption in agreement with that in case 1. 1). For the combination process coal + precooled tar for the production of gasoline, fuel oil and briquetting material we get in comparison with the separate tar and coal processes, and with the same assumption as in cases 4 and 5, a savings in the reaction spaces amounting to about 40 percent.

### General

It is worthy of notice from a chemical and of course also from an economic standpoint that in these combination processes the two hydrogenation processes may to a certain extent be considered as superimposed upon each other in the same reaction space, without either process appreciably affecting the other one; or in other words, that the reaction space in the combination process is appreciably smaller than the sum of the reaction spaces for the two processes carried out separately. In the present case the reaction space occupied by a practically unreacting pasting oil can be utilized by the selection of a pasting oil capable of cracking and hydrogenation (e.g. tar instead of middle oil). The application of these principles to the production of gasoline and fuel oil from coal would permit, i.e., to replace the hot oil let down by some available low temperature tar, producing more asphalt-free oils in the same reaction space.

### Technique of the Process

A large scale test is available for extracting hydrogenation with a small amount of gas (cases 1 and 2). There is another large scale run, but lasting only a short time (1 week) for the case 3 (coal + middle oil, containing low temperature tar). The other processes have only been tested in 10 li converters. All methods were run with the use of a cold catchpot, with a heat exchanger connected between the converters and the catchpots. The total liquid reaction product was used for the heat exchange. The reaction products passing through the return pass of the heat exchanger partially plugged it up at the conclusion of large scale runs. For this reason the heat exchange problem does not seem to be entirely solved. It is to be found to what extent the catchpot temperature must be raised to obtain larger amounts of distillate which can be used as heat exchange means.

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