

# United States Patent

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[50] Field of Search..... 208/10

[56] References Cited  
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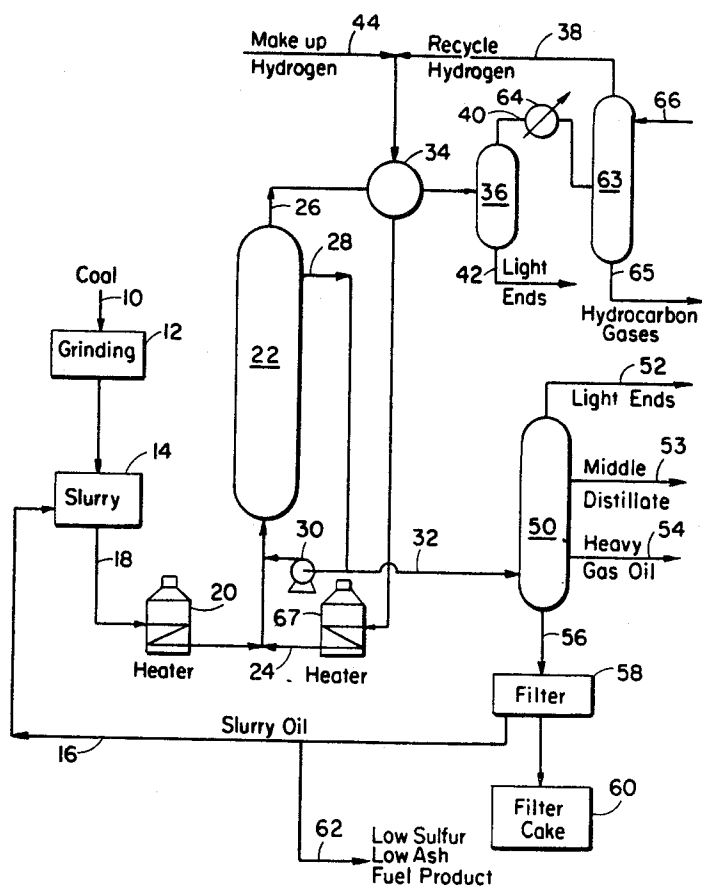
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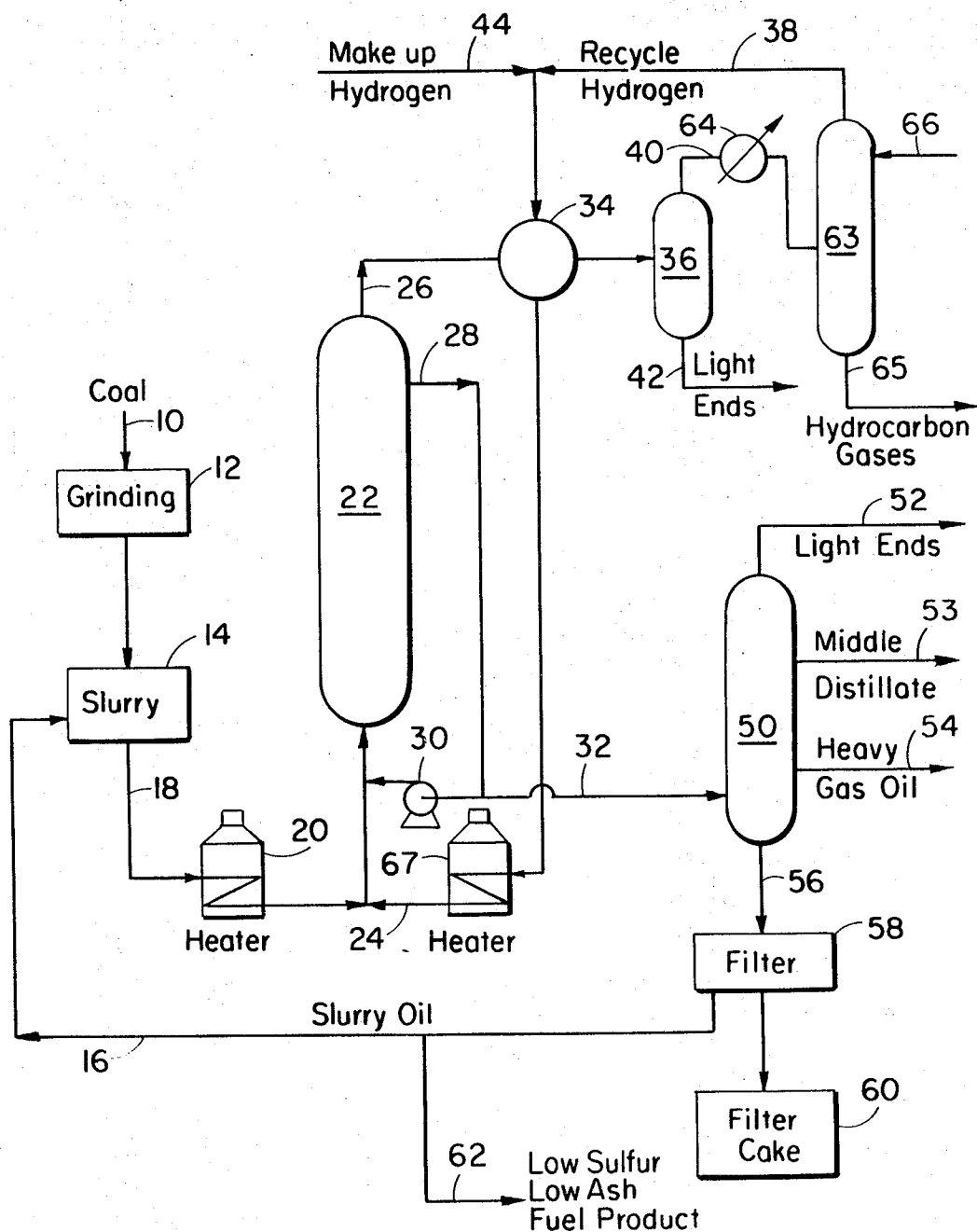
[54] **LOW SULFUR FUEL OIL FROM COAL**  
9 Claims, 1 Drawing Fig.

[52] U.S. Cl..... 208/10

[51] Int. Cl..... C10g 1/06

**ABSTRACT:** The economic production of a low sulfur residual fuel oil by the hydroconversion of coal is accomplished in an "ebullated" bed system in the absence of downstream processing and the use of a minimum of hydrogen to produce a fuel of high B.t.u. value and low sulfur content.





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## LOW SULFUR FUEL OIL FROM COAL

## BACKGROUND

The increase in government concern regarding air pollution, especially with regard to sulfur emission into the atmosphere, has resulted in legislative action which could eliminate the use of some of our conventional coals in the utilities market. This is particularly true for coals such as Pittsburgh No. 8 which contains about 4 percent sulfur, as well as Illinois No. 6. A process which can economically convert coal to a fuel which will meet anticipated air pollution regulations is of prime economic interest.

While it is known that such coals can be processed to produce gasoline boiling range materials, together with some gas and some heavier boiling products, it has been found that the high consumption of hydrogen and catalyst, and relatively low coal throughput per volume of reactor, is not economical unless gasoline is the principal product made.

Hydrogenation of coal depends on many factors including the characteristics of the coal as to oxygen, ash, and volatile content. Other factors include those controllable reaction conditions of temperature, pressure, throughput, and catalyst. Equally as a controlling factor is the quality of the end product.

Normally, the hydrogenation of coal has proceeded to the ultimate production of gasoline or similar high quality fuels. In such case, the use of hydrogen, a relatively expensive commodity has directed the research into conditions of high conversion, usually requiring two or more reaction stages and separation equipment.

## SUMMARY OF THE INVENTION

This invention is primarily adapted to make an inexpensive, low cost, fuel substitute of low gravity which may be used either as ground-up solids or as a liquid if maintained at a temperature above the melting point. The low sulfur characteristic is especially beneficial for reduction of pollution.

## DRAWING

The drawing is a schematic view of a reactor and auxiliary equipment for a coal hydrogenation process.

## DESCRIPTION OF PREFERRED EMBODIMENT

Coal at 10, appropriately ground at 12 (and not necessarily dried), is mixed at 14 with recycle slurry oil 16 to form a coal-oil slurry. This slurry is passed by line 18 through heater 20 into the lower part of reactor 22. Hydrogen is added at 24. Liquid oil and hydrogen pass upwardly through a bed of catalyst or activated alumina at sufficient velocity to maintain an ebullated bed of catalyst or inert solids such as disclosed in the Schuman U.S. Pat. No. 3,281,393.

A gaseous phase is removed overhead at 26 and a liquid stream is removed at 28. The liquid stream is in part recycled through pump 30 to maintain the desired liquid velocity and a net liquid stream is removed at 32.

Preferably the coal is initially ground to all pass 20 mesh and not more than about 10 percent passing 325 mesh (Tyler). The slurry at 18 is a pumpable slurry with at least equal parts of oil and coal but, if for operating reasons, it is desirable to recycle an additional amount of slurry oil, slurries of one part coal and up to 10 parts oil may be used.

A temperature of 800°-900° F. preferably about 850° F. and a hydrogen partial pressure of 800-2500 p.s.i. and preferably about 1,890 p.s.i. is maintained in the reactor.

The vapors 26 leaving the upper part of the reactor are cooled at 34. Condensed light ends are separated in drum 36 and removed at 42. Vapors leaving drum 36 at 40 are cooled in exchanger 64 and scrubbed with an absorber oil in column 63 to remove light hydrocarbon gases at 65 and hydrogen leaving at 38 is recycled. The recycle hydrogen stream 38 plus makeup hydrogen at 44 also passes through heat exchanger 34, through heater 67 and becomes the hydrogen feed line 24.

The liquid leaving the reactor at 32 is separated in one or more fractionation columns 50 into a light ends stream 52, a middle distillate at 53, a heavy gas oil at 54 and a heavy ends at 56.

The heavy ends at 56 contain a substantial amount of solids which are sent to a filter 58. The filter cake is recovered at 60. The filter cake comprises a char and ash product and may contain up to 20 percent of oil which can be recovered thermally. A centrifuge could also be used.

The filtrate leaving filter 58 provides the slurry oil recycle stream 16 and the fuel oil product, 62.

The low sulfur fuel oil product 62 will have an API gravity of about -14° with a B.t.u. value in excess of 16,600 BTU's per pound. Normally this product has a boiling point not less than 400° F. and must be kept hot in order to permit pumpability or distillates can be removed to give a product boiling at 900° F. and higher.

Alternatively, the fuel oil product can be cooled, solidified and ground and used as a solid combustible fuel low in ash and sulfur, especially when free of distillates.

The reactor 22 may be operated under varying conditions depending upon the maximum sulfur desired in the fuel oil product. The following table illustrates experimental results from the operations. Approximately three barrels of fuel oil with a sulfur content of less than 0.5 weight percent sulfur and 0.56 barrels of naphtha have been produced per ton of Illinois No. 6 coal.

Treatment of coal from the Pittsburgh No. 8 seam has yielded 3.48 barrels of fuel oil per ton and 0.21 barrels per ton of naphtha. In this case the fuel oil product contained approximately 1 percent sulfur.

The versatility of the reactor is indicated in the table below in which, in one case, 93 pounds of Illinois No. 6 coal per hour per cubic foot was the coal feed rate utilizing a cobalt molybdate on alumina catalyst. In such case the hydrogen consumption was approximately 3.75 s.c.f./lb.

The Pittsburgh No. 8 coal was operated at a throughput rate of 187 pounds of coal per hour per cubic foot with activated alumina and only 2 s.c.f./lb. of hydrogen was consumed.

## EXAMPLE OF TYPICAL COAL ANALYSES

|                        | Illinois No. 6 | Pittsburgh No. 8 |
|------------------------|----------------|------------------|
| Carbon                 | 70.51          | 74.29            |
| Hydrogen               | 5.14           | 5.49             |
| Nitrogen               | 1.28           | 1.49             |
| Sulfur                 | 3.39           | 4.03             |
| Oxygen                 | 8.08           | 6.40             |
| Ash                    | 11.60          | 8.30             |
| Volatile Matter (MAF)* | 44.56          | 46.46            |
| Organic sulfur         | 1.24           | 2.10             |

## REACTOR CONDITIONS

|                                 | Illinois No. 6  | Pittsburgh No. 8 |
|---------------------------------|-----------------|------------------|
| Pressure, total—p.s.i.g.        | 2,250           | 2,250            |
| Temperature °F.                 | 850             | 850              |
| Hydrogen Consumption s.c.f./lb. | 3.75            | 2                |
| Catalyst                        | CoMo on Alumina | Alumina          |
| Throughput lbs./hr./cu.ft.      | 93              | 187              |

\*MAF - Moisture and Ash Free

## EXAMPLES OF YIELDS

(Pounds per 100 pounds of dry coal)

|                 | Illinois No. 6 | Pittsburgh No. 8 |
|-----------------|----------------|------------------|
| CO <sub>2</sub> | 0.92           | 1.0              |
| CO              | 0.31           |                  |
| C <sub>1</sub>  | 1.66           | 4.81             |
| C <sub>2</sub>  | 0.80           |                  |
| C <sub>3</sub>  | 1.28           |                  |

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|                              |        |        |
|------------------------------|--------|--------|
| C <sub>4</sub> -400          | 7.71   | 2.85   |
| 400-650                      | 18.19  | 3.83   |
| 650-975                      | 9.19   | 16.41  |
| Residuum (975° F. plus)      | 35.31  | 53.15  |
| Coal Residue                 | 7.34   | 6.90   |
| Ash                          | 11.60  | 8.30   |
| H <sub>2</sub> S             | 1.75   | 1.80   |
| NH <sub>3</sub>              | 0.72   | 0.20   |
| H <sub>2</sub> O             | 5.27   | 1.80   |
|                              | 102.05 | 101.05 |
| Yield Summary                |        |        |
| Fuel Oil 400° F. plus B/T    | 3.06   | 3.48   |
| Naphtha B/T                  | 0.56   | 0.21   |
| Sulfur (product) % by weight | 0.46   | 1.02   |

\*B/T—Barrels per ton

The relatively low consumption of hydrogen, the high throughput of the reactor, and the substantial absence of downstream refining processing makes it possible to produce the low sulfur residual fuel oil at a cost of about one half that for production of gasoline.

Although but two examples of coal conversions are given, it is our experience that operating ranges will be as follows:

|                 |                                         |
|-----------------|-----------------------------------------|
| Temperature     | 800-900° F.                             |
| Pressure, total | 1,000-3,000 p.s.i.g.                    |
| Feed rate       | 75-200 lbs./hr./cu.ft. of reactor space |

When the fuel oil recovered boils at 400° F. plus it is preferably maintained as a liquid fuel although it can be used as a solid fuel. Where the fuel oil recovered is free of distillates, it is preferable that the remaining fuel oil boil at 900° F. plus. This 900° F. plus fuel oil is suitable for use as a liquid fuel although it is preferably allowed to cool and used as a solid fuel.

Ammonia and hydrogen sulfide produced in the coal hydrogenation step are recovered and the hydrogen sulfide may be converted to elemental sulfur by the Claus process providing ammonia and sulfur as byproducts.

While we have shown and described a preferred form of embodiment of our invention, we are aware that modifications may be made thereto within the scope and spirit of our disclosure and the claims hereinafter attached.

We claim:

1. The process of producing a low sulfur fuel oil comprising less than about 1 weight percent of sulfur by the hydrogenation of a coal comprising greater than one weight percent of sulfur which comprises:

- passing an oil-coal slurry upwardly through a reaction zone operating under a hydrogen partial pressure between about 800 and about 2,500 p.s.i., and under a temperature between about 800° and about 900° F.;
- consuming hydrogen, in the reaction zone, at a rate of at

least 2 standard cubic feet (s.c.f.) and not to exceed 10 s.c.f. per pound of coal;

- maintaining a space velocity between about 75 and about 200 lbs. of coal per hour per cubic foot of the reaction zone;
- removing a coal effluent which comprises gases, liquids, ash and unconverted coal solids;
- separating a solids containing fraction from said effluent; and
- recovering a hydrocarbon fraction boiling above about 400° F. plus, the volume of which is at least 2.5 barrels per ton of coal fed to the reaction zone.

2. The process of claim 1 in which the reaction zone contains an ebullated bed of particulated solids wherein said particulated solids comprise cobalt molybdate on alumina.

3. The process of claim 1 in which the reaction zone contains an ebullated bed of particulated solids wherein said particulated solids comprise activated alumina.

4. The process of producing a low sulfur fuel oil comprising less than about 1 percent of sulfur by the hydrogenation of a coal comprising greater than one weight percent of sulfur which comprises:

- passing an oil-coal slurry upwardly through a reaction zone operating under a hydrogen partial pressure between about 800 and about 2,500 p.s.i. and under a temperature between about 800 and about 900° F.;
- consuming hydrogen, in the reaction zone, at a rate of at least 2 standard cubic feet (s.c.f.) and not to exceed 10 s.c.f. per pound of coal;
- maintaining a space velocity between about 75 and about 200 lbs. of coal per hour per cubic foot of the reaction zone;
- removing a coal effluent which comprises gases, liquids, ash, and unconverted coal solids;
- separating a solids containing fraction from said effluent; and
- recovering a hydrocarbon fraction boiling above about 900° F. plus, the volume of which is at least 1 barrel per ton of coal fed to the reaction zone.

5. The process of claim 4 in which the reaction zone contains an ebullated bed of particulated solids wherein said particulated solids comprise cobalt molybdate on alumina.

6. The process of claim 4 in which the reaction zone contains an ebullated bed of particulated solids wherein said particulated solids comprise activated alumina.

7. The process of claim 4 wherein the hydrocarbon fraction boiling above about 900° F. plus is allowed to solidify and ground for use as a particulate solid fuel.

8. The process of claim 1 wherein the hydrogen partial pressure is about 1,890 p.s.i. and the temperature is about 850° F.

9. The process of claim 4 wherein the hydrogen partial pressure is about 1,890 p.s.i. and the temperature is about 850° F.

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