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Wolk et al.

[11] **3,856,658**

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[54] **SLURRIED SOLIDS HANDLING FOR COAL HYDROGENATION**

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Related U.S. Application Data

[63] Continuation-in-part of Ser. No. 191,035, Oct. 20, 1971, abandoned.

[52] **U.S. Cl.** **208/10,** 208/8

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

[58] **Field of Search** 208/10; 302/14

[56] **References Cited**

UNITED STATES PATENTS

3,400,984	9/1968	Shellene et al.	302/14
3,519,555	7/1970	Keith et al.	208/10
3,635,814	1/1972	Rieve et al.	208/10

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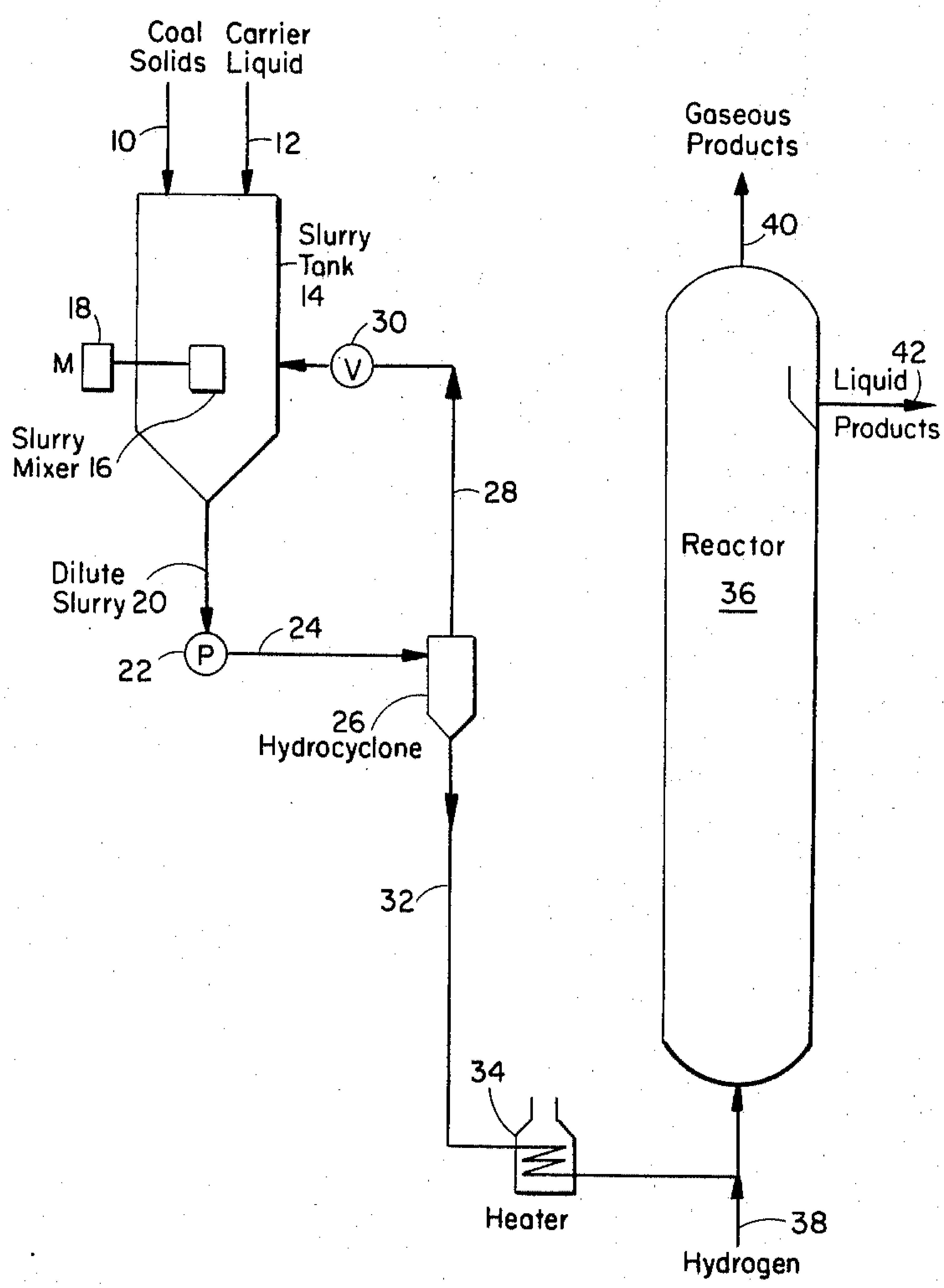
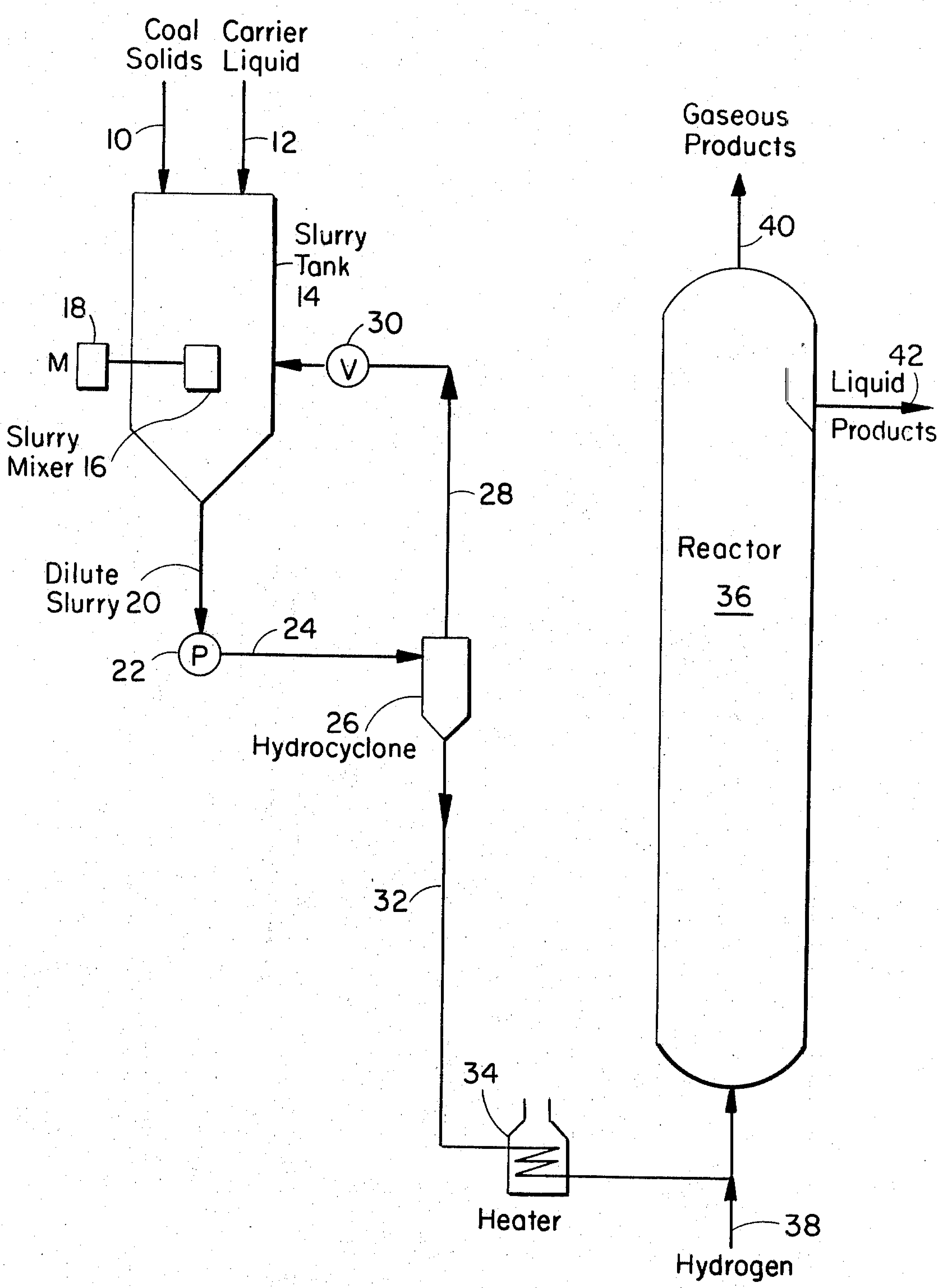
[57] **ABSTRACT**

In the H-Coal process for coal hydrogenation, an initial step is the preparation of a coal slurry for transfer by suitable means into the pressurized reactor. This invention provides an improved method for facilitating such slurry transfer by pumping, and uses a recycled carried liquid to help provide a dilute easily pumpable coal slurry which is pressurized to above reactor pressure and then concentrated in solids content to a desired extent by centrifugal separation means before passing the concentrated slurry to the reaction zone. A clarified liquid stream withdrawn from the slurry concentration step is recirculated back to the mixing step to facilitate the pumping step.

10 Claims, 1 Drawing Figure




Coal Solids
Carrier Liquid
10
12
Slurry Tank 14
18
M
30
V
Slurry Mixer 16
28
Dilute Slurry 20
24
P
22
26 Hydrocyclone
32
34
Heater
Gaseous Products
40
42
Liquid Products
Reactor 36
38
Hydrogen

SLURRIED SOLIDS HANDLING FOR COAL HYDROGENATION

CROSS REFERENCE TO RELATED APPLICATIONS

This is a continuation-in-part of copending application, Ser. No. 191,035, filed Oct. 20, 1971 and now abandoned.

BACKGROUND OF THE INVENTION

Hydrogenation of a coal-oil slurry by the upflow, liquid-phase, random motion ebullated bed process known as H-Coal and described in U.S. Pat. No. 3,519,555 to Keith et al., is accomplished at temperatures in the range of 750°–950°F and pressures in the range of 1,000–3,000 psig with coal throughputs ranging from 15 to 300 pounds per hour per cubic fool of reactor volume, and results in yields in the order of four barrels of synthetic crude oil per ton of coal. There are, of course, other variables involved, primarily due to the rank of the coal to be converted and to the end products desired.

The particulate coal is usually mixed with a carrying oil to form a slurry, which is then pressurized and introduced into the reactor along with heated hydrogen gas. However, introducing the relatively viscous coal-oil slurry into the high pressure ebullated bed reactor presents significant problems. Because the coal particles are quite abrasive, they cause rapid erosion and wear in the pumping equipment, particularly for positive displacement type pumps which are usually used at the high discharge pressures involved and which have close internal clearances.

SUMMARY OF THE INVENTION

It has been found that the economical introduction of coal into a pressurized reactor is a function of the solids concentration in the coal-liquid slurry, as well as a function of the viscosity and temperature of the carrier liquid used. While it is essential to provide a satisfactorily pumpable fluid slurry, any excessive dilution used in preparing the slurry to facilitate the pumping step has involved considerable excess investment costs for the rest of the high pressure reaction system and much higher operating costs. However, a coal-liquid slurry more concentrated in solids becomes far more difficult to pump in that the coal solids tend to settle out in the pump body and thus interfere with its normal operation, and also to accelerate erosion of the pump inner parts.

In accordance with the present invention, the pumping of the coal-liquid slurry to high pressure is accomplished on a dilute (low percentage of solids) slurry stream which is first pressurized to super-atmospheric pressures, preferably to slightly above the desired reactor pressure, following which a concentration of solids in the slurry to the extent desired is accomplished by centrifugal force means at substantially the same pressure level. This concentrated slurry is then heated to near reactor temperature and passed into the reaction zone without further pressurization.

In the slurry concentration step, the undesired liquid portion is conveniently and substantially removed from the dilute slurry by centrifugal force action using a liquid cyclone device, and such liquid is recycled to the initial slurry preparation step. Separating this undesired liquid portion by centrifugal force means such as a liquid cyclone or hydroclone apparatus is quite desirable, in that the apparatus is relatively simple and reliable and no plugging problems ordinarily result due to the solids portion of the slurry, such as would usually occur if filtration means were used for the slurry concentration step. This liquid recycle arrangement permits the pressurizing transfer pump to advantageously handle a dilute slurry material only, with the concentrated slurry material then being constituted so as to flow freely through regular piping and through the remainder of the system. While the carrier liquid used may be any hydrocarbon oil, it is preferably a light hydrocarbon oil which may conveniently be made in the coal hydrogenation process.

The principle advantage provided by this invention is that it materially reduces the amount of diluent oil that must be handled in the remainder of the system downstream of the liquid-solid separation step, i.e., in the heater, reactor, heat exchangers, distillation apparatus and such. Consequently, this arrangement requires a smaller reactor and less heat exchange surface and smaller piping for handling the excess liquid. Other advantages include less wear on the slurry pump, which thus usually can be a less expensive pump.

DESCRIPTION OF THE DRAWING

The drawing is a schematic view of certain process equipment for the hydrogenation of coal.

DESCRIPTION OF THE PREFERRED EMBODIMENT

Referring to the attached drawing, particulate coal solids at **10** and a carrier liquid, which is preferably a light hydrocarbon oil, at **12** are added to a slurry mixing tank **14** in a preferred ratio of about 60 weight percent solids to 40 weight percent liquid. The solids/liquid weight ratios of the slurry, however, may vary from about 0.5:1 to 3:1 of coal to liquid depending on the properties of the carrier liquid. The mixing tank **14** conveniently has a slurry mixer **16** which is driven by motor **18,** which may be either electric or hydraulic and powered by recycle liquid.

The resulting coal-liquid slurry, diluted by a recycle liquid stream as explained below and at a pressure between atmospheric and about 15 psig, is withdrawn from the mixing tank through line **20** to pump **22,** wherein it is pressurized to above reactor pressure or about 500–3250 psig, and preferably to 1000–3,000 psig. This high pressure dilute slurry is then passed through line **24** to liquid cyclone **26** for liquid-solids separation. In the liquid cyclone **26** the separation of the dilute slurry into a substantially clarified liquid overflow and concentrated slurry underflow streams occurs by centrifugal and centripetal force action induced by the pressure drop developed through the static device. The liquid cyclone **26** is selected and the pressure differential across it adjusted by the flow rate therethrough so that about 30 weight percent of dilute slurry stream **24** is returned as clarified liquid through line **28** under control of pressure reducing means **30** back to slurry mixing tank **14** to help provide the dilute slurry therein. The pressure drop across the liquid cyclone device will usually be 50–200 psi. Thus, the remaining concentrated slurry at substantially the same pressure level and preferably containing about 60 weight percent solids and 40 weight percent carrier liquid, is now passed by line **32** through heater **34.** Herein it is heated to at least 500°F and preferably to 600°–800°F and then passed on to the hydrogenation reactor **36.**

As an example of this reactor **36,** reference is made to U.S. Pat. No. **3,519,555** previously mentioned. In such case, by appropriate control of liquid and coal and hydrogen supplied in line **38,** an ebullated bed hydroconversion condition can be provided in the reactor with the ultimate removal of gaseous products at **40** and liquid products at **42.** Carrier liquid **12** is preferably a light hydrocarbon oil boiling in the range of fuel oil, that is, having a nominal boiling range above 400°F, illustratively, one having a nominal boiling range of 400°–975°F and recovered from the process by conventional processing steps (not shown) downstream of liquid stream **42.**

It is also within the contemplation of this invention that the solid particulate matter may be catalyst rather than, or together with, the coal, such as also disclosed in U.S. Pat. No. 3,519,555.

In a coal hydrogenation system of this type, the dilute slurry is pumped to a high pressure, following which the clarified liquid obtained after passing through the liquid cyclone separator is recycled back through pressure reducing means **30** to the slurry mixing tank. If desired, the dissipation of the pressure of this recycled liquid stream may be accomplished by a hydraulic turbine, with the power output therefrom used in the system either for driving the slurry mixer motor **18** or assisting in driving high pressure pump **22.** The concentrated slurry stream at substantially the high pressure level is passed to the process reactor **36.**

In this system utilizing high pressure pump **22,** which is preferably a positive displacement reciprocating plunger type pump, the pump operates only on dilute slurry. Furthermore, the more dilute the slurry, the less the pump wear and greater the pump reliability. No wasteful diluent is required in the remainder of the system and such avoidance of excess liquid processing and additional heat exchange surface is highly beneficial in reducing equipment investment and operating costs. For example, in a coal hydrogenation process as normally operated with high pressure slurry pump **22,** as much as 50–60 weight percent diluent liquid is required in the process. Using this invention, only about half of this diluent liquid is required for transfer of the coal slurry downstream of liquid cyclone **26** through the associated piping, heater, etc., to the reactor.

The significance of this coal slurry handling method for ebullated bed coal hydrogenation is evident from the determination that viscosity of coal-oil slurry increases appreciably for each percent increase in solids in the slurry. It is usually preferred practice to limit the viscosity of slurries to be pumped in this pressure range (up to 3,500 psig) to about 3 poise and not more than 10 poise. Pumping concentrated slurries is therefore quite difficult and expensive.

Normal conditions for coal hydrogenation have been established using a start-up coal-oil slurry of about 20 weight percent solids at about 100°F (ambient), which is pressurized to reactor pressure by one or more pumping stages, then concentrated to about 50 weight percent solids. This slurry material will flow readily through regular piping in the heater and to the reactor.

EXAMPLE

From the established data, it is found that for coal hydrogenation, the coal-oil dilute slurry may have the following initial properties:

1. Slurry formed of pulverized coal finer than 20 mesh (U.S. Sieve Series) and hydrocarbon oil boiling in the range of fuel oil, i.e., having a nominal boiling range above 400°F.
2. Temperature of 100°–250°F.
3. Viscosity of 3–10 poise.
4. Materials added to the slurry mixing tank in 60/40-solids/liquid weight ratio.

The dilute slurry is pressurized to 1,000–3,000 psig, then concentrated by centrifugal force means using a liquid cyclone apparatus, after which the clarified liquid portion is removed for return to the mixing tank. The remaining concentrated slurry for reactor feed contains about 60/40-solids/liquid ratio. Such a slurry material will flow through pipes and heat exchanger passages to the reactor.

While we have shown and described a preferred form of embodiment of the invention, we are aware that modifications may be made thereto within the scope and spirit of the disclosure herein and of the claims appended thereto. For example, while a single liquid cyclone device has been shown, it is understood that multiple cyclone units connected in parallel could advantageously be used in systems where the flow rate exceeded the desired capacity of a single cyclone unit.

We claim:

1. A method for introducing particulate coal in liquid phase slurry form into a high pressure reaction zone which comprises:

a. mixing said particulate coal with a hydrocarbon liquid to produce a dilute slurry;
b. pressurizing said dilute slurry to a pressure above the reaction zone pressure;
c. passing said pressurized slurry through liquid-solid centrifugal type separator means without substantial loss of pressure to produce a clarified liquid portion and a concentrated slurry portion;
d. returning the clarified liquid portion from (c) through pressure reducing means to the mixing step (a); and
e. passing the concentrated slurry portion of (c) to the high pressure reaction zone.

2. The method of claim **1** wherein the particulate coal solids are smaller than 20 mesh and the slurrying liquid is a light hydrocarbon oil.

3. The method of claim **1** wherein the particulate solids are a mixture of coal and catalyst and the liquid is oil.

4. The method of claim **1** wherein the initial slurry is formed under ambient conditions and is pressurized to at least 500 psig and the concentrated slurry is then heated to a temperature of at least 500°F.

5. The method of claim **1** wherein the pressure drop across the centrifugal separator means to effect the liquid-solids separation is 50–200 psi.

6. The process of claim **1** wherein the slurry of step (b) has a viscosity less than about 10 poise.

7. The method of claim **1** wherein the slurry pressurizing means is a positive displacement reciprocating plunger type pump.

8. A liquid phase catalytic hydroconversion process for converting coal to liquid hydrocarbons and gas which comprises:

a. slurry said coal with a hydrocarbon oil boiling above 400°F recovered from the process in a ratio of from 0.5 part of coal per part of oil to 3 parts coal per part of oil at substantially ambient conditions;

b. pressurizing said slurry by positive displacement pumping means to a pressure of 1,000–3,000 psig;

c. centrifugally separating at least a substantial part of the liquid portion from the coal slurry by centrifugal force means to provide a more concentrated solids slurry portion;

d. returning the clarified liquid from step (c) through pressure reducing means to step (a);

e. heating the concentrated coal slurry to 600°–950°F; and

f. discharging said concentrated heat slurry to a high pressure reaction zone wherein it is converted to hydrocarbon liquid and gas products.

9. The process of claim **8** wherein said slurry liquid is a hydrocarbon oil recovered from the process and having a nominal boiling range above 400°F.

10. The process of claim **8** wherein the pressure reducing means to step (d) is a hydraulic turbine.

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