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Li et al.

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[54] **UPFLOW THREE-PHASE FLUIDIZED BED COAL LIQUEFACTION REACTOR SYSTEM**

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[51] **Int. Cl.** **C10g 1/08**

[58] **Field of Search** 208/8; 23/267, 270

[56] **References Cited**

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Re25,770	4/1965	Johanson	208/10
2,714,086	7/1955	Bluemner	208/8
2,985,698	5/1961	Pechtold et al.	260/679
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3,011,953	12/1961	Foch	208/8
3,488,278	1/1970	Nelson	208/8
3,162,510	12/1964	Meissner et al.	23/267 R

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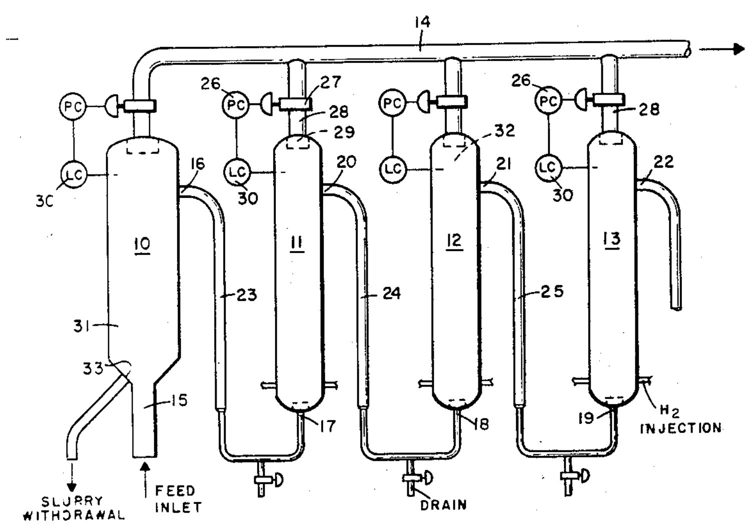
[57] **ABSTRACT**

A coal/liquid slurry is conducted upwardly in a three-phase fluidized state through a plurality of vessels connected in series, with the gas generated in each individual vessel being purged to a common header by a pressure control system included in each stage for ensuring control of flow through the reactors. The lead vessel includes the following features: a narrow, bottom entrance section designed to produce a superficial liquid velocity which is higher than the minimum fluidization velocity of the heaviest, largest particles in the slurry, so that all entering particles can be fluidized in the entrance section; an expanded upper section designed to slow the upflow of the slurry to a superficial liquid velocity sufficiently lower than the aforesaid minimum fluidization velocity to carry upwardly all particles which are essentially organic in composition but insufficient to carry upwardly a major portion of particles which are essentially nonorganic in nature, so that such major portion of essentially nonorganic particles settle from the upper section and do not carry over to a subsequent vessel; and a side opening at the intersection of these two sections for the continuous withdrawal of a slurry of those particles which settle from the upper section. The vessels subsequent to the leading vessel each have a uniform diameter smaller than the diameter in the lead vessel, so that particles passed from the lead vessel will not settle in the subsequent vessels. Each subsequent vessel also has vertical internals to provide a close approach to plug flow performance. Also, entry ports are provided at the base of each subsequent vessel for hydrogen gas addition.

7 Claims, 4 Drawing Figures



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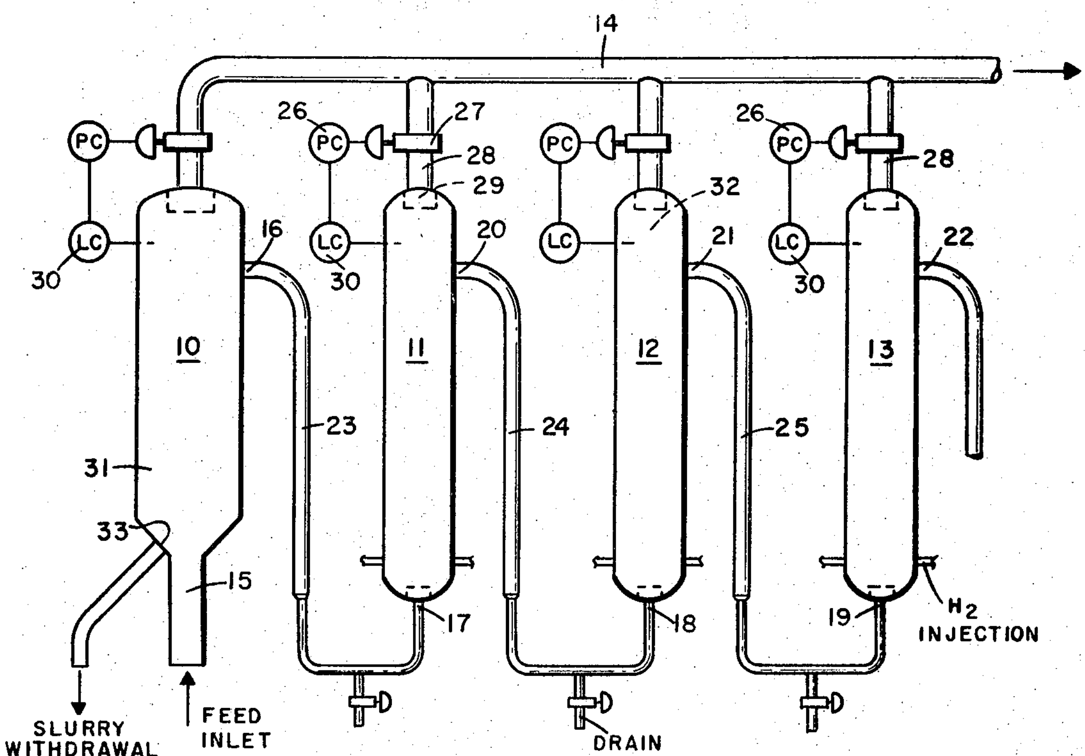

FIG. 1.

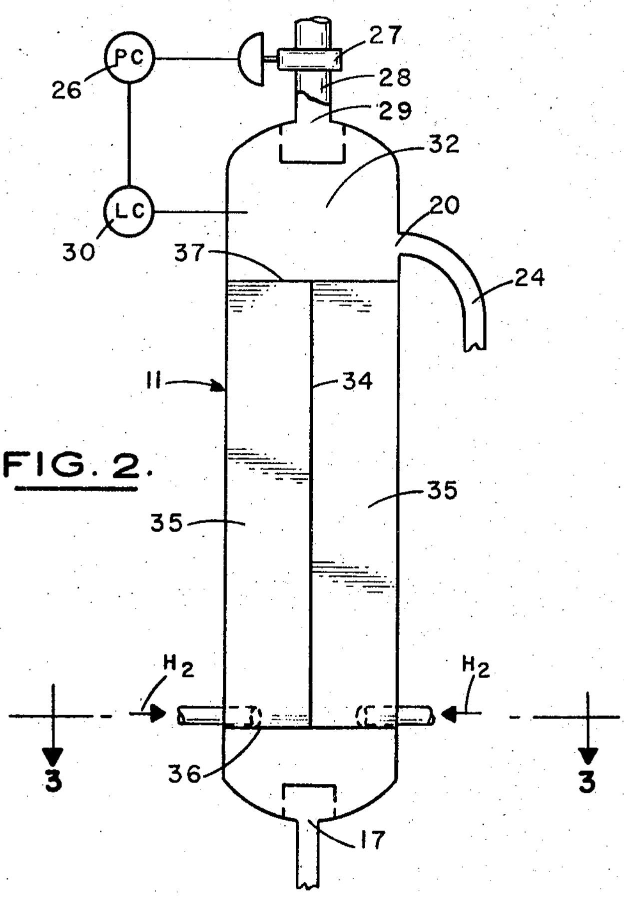

FIG. 2.

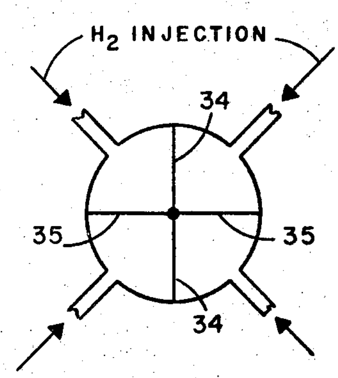

FIG. 3.

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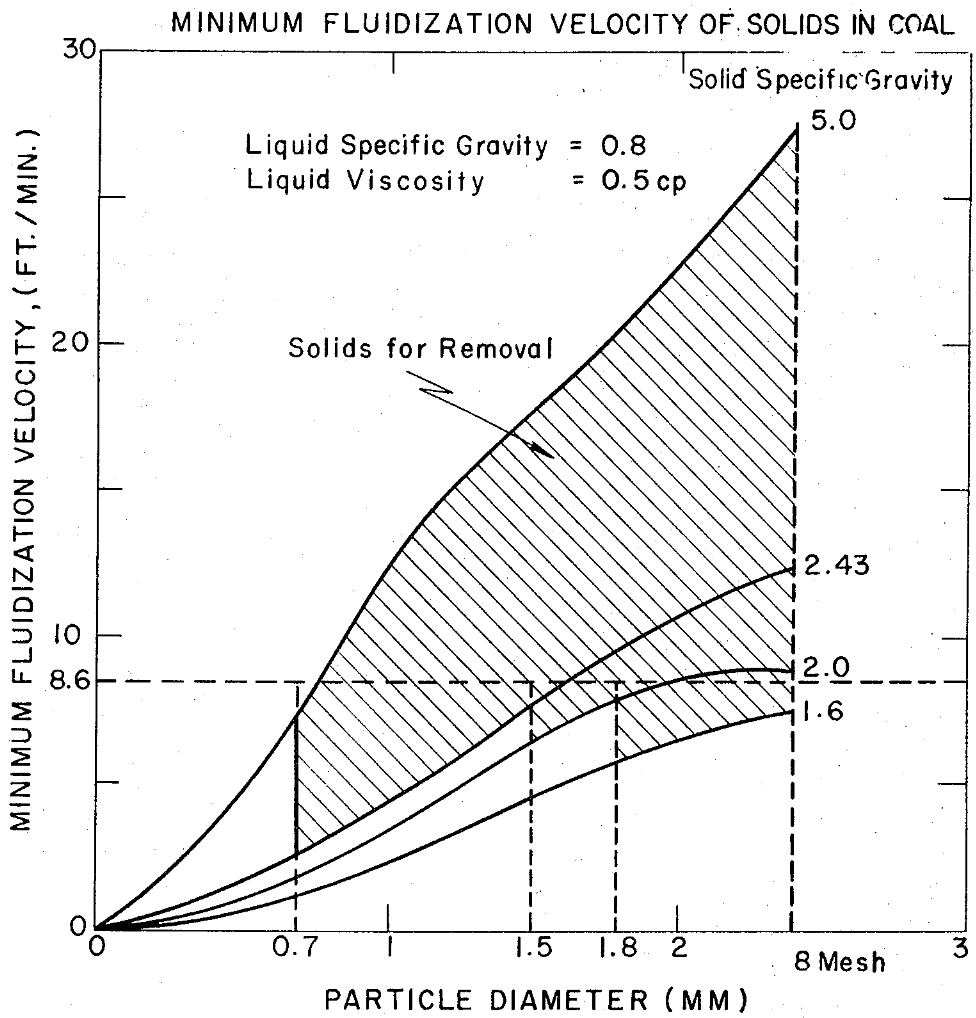

FIG. 4.

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UPFLOW THREE-PHASE FLUIDIZED BED COAL LIQUEFACTION REACTOR SYSTEM

BACKGROUND OF THE INVENTION

This invention relates to methods and apparatus for the solvent liquefaction of coal solids, and more particularly, to staged upflow coal liquefaction reactors connected in series and methods of liquefying solvent slurried coal solids in such reactors.

Heretofore, as in U.S. Pat. No. 2,714,086, coal slurries liquefied in upflow reactors connected in series have used rotors and other agitators to impose well-mixed conditions on a slurry passing upwardly in such reactors, with attendant problems of costly power consumption and the associated conversion limitations of a well-mixed reactor. More recently, in U.S. Pat. No. 3,488,278, a countercurrent fluidization of coal particles introduced into the top of a liquefaction reactor was suggested. In this process, however, large, undissolved coal and mineral particles are allowed to settle to the bottom of the reactor for removal. Consequently, there is a strong likelihood that the settled particles will plug the reactor. On the other hand, if all the particles were taken overhead from the reactor, as suggested in U.S. Pat. No. 2,714,086, then either massive liquid cyclones have to be used to effect solids removal from the entire liquid stream or all downstream reactors have to handle very high liquid velocities in order to fluidize the large solid particles so as to prevent them from settling. The latter alternative leads to an unreasonable number of stages to obtain the total required residence time for liquefaction and causes a very high pressure drop in the reactor system. In coal liquefaction, what is needed, and what the prior art has not provided, is a reactor system which removes undissolved solids in such a way as to minimize the likelihood of reactor plugging, and which provides a practical means by which coal solids can be liquefied in a compact reactor volume to a high level of conversion. The present invention fulfills this need.

Prior art considered in the foregoing discussion, in addition to the patents already cited, includes U.S. Pat. Nos. 3,488,278; 2,840,462; 2,985,698 and 3,047,371.

SUMMARY OF THE INVENTION

In short, this is an invention of a method and means by which slurried coal particles are flowed upwardly in a three-phase fluidized state in a liquefaction reactor system at staged superficial liquid velocities. This permits plug flow rheology to be efficiently employed in a compact system. In the bottom of the leading part of the upflow reactor system, the superficial liquid velocity is higher than the minimum fluidization velocity of the heaviest, largest particles in the slurry, so that all particles in the slurry are fluidized. In an upper portion of the leading part of the reactor system, the superficial liquid velocity is lower than the minimum fluidization velocity and is sufficient to fluidize and carry upwardly substantially all of the particles which are essentially organic in composition, but it is insufficient to fluidize and carry upwardly a major portion of the particles which are essentially nonorganic in nature. Residence time in the upper portion is sufficient to permit dissolution of at least a major portion, preferably substantially all, of the dissolvable particles. The essentially nonorganic particles which settle from the upper portion are withdrawn with solvent form the reactor system between the bottom and upper portions at a rate which does not remove more than a predetermined maximum of solvent from the slurry introduced into the reactor system. After the slurry of undissolved particles leaves the leading part of the reactor system, plug flow is imposed on it. The superficial liquid velocity of the slurry in the parts of the reactor system subsequent to the leading part is greater than the superficial liquid velocity in the upper portion of the leading part so that particles leaving the upper portion will not settle in the subsequent parts.

An important feature of the invention is that gaseous material is removed from the tops of the leading and subsequent parts of the reactor system so as to impose a predetermined pressure in the leading and subsequent parts which will cause a positive slurry flow to occur in the system. Removal of the gaseous material eliminates gas accumulation from the leading to the subsequent parts of the reactor system, preventing reactor inoperability caused by occurrence of any excessive gas velocity in the reactor system.

In the general practice of this invention, the coal particles which are fed in slurry form to this coal liquefaction factor system are suitably obtained by grinding bituminous coal, subbituminous coal, lignite, brown coal or other suitable coal, and will have a particle size distribution preferably from about 8 mesh (Tyler screen) and finer, although it has been found that the solvation reaction will result in a size reduction even if particles as large as one-quarter inch on the major dimension are introduced into the liquefaction reactor. Coal particles, as well known in the art, are made up of organic matter and mineral matter. The mineral matter is of high specific gravity, generally from about 2.7 to about 3.0, except for pyrite and other heavy minerals which have specific gravities of up to 5.0 and greater. Organic matter generally has lower specific gravities within the range from about 1.2 to about 1.4, usually about 1.3 or so. Coal particles obtained by grinding and produced by disintegration when coal dissolves in solvent have specific gravities which range all the way from about 1.3 to about 5.0. That is, some particles which are essentially organic in composition have a specific gravity which may extend upwardly to about 2.4 and perhaps slightly higher. Particles which are essentially nonorganic in nature have specific gravities which extend down to about 2.5 and perhaps slightly lower. As a result, this invention preferably requires that the bottom of the leading part of the reactor system be made so that the superficial liquid velocity in it is at least sufficient to fluidize and carry upwardly particles having a particle diameter of 8 mesh and smaller with a specific gravity of 5 and lower, and that the upper portion of the leading part of the reactor system be made so that the superficial liquid velocity in it is sufficient to fluidize and carry upwardly particles having a particle diameter of 8 mesh and smaller with a specific gravity of about 2.4 and lower, but insufficient to fluidize and carry upwardly particles which have a particle diameter of 1.6 mm. and larger with a specific gravity of about 2.5 and higher.

The present staging of superficial liquid velocities permits utilization of plug flow rheology in the reactor system to provide higher conversion levels of dissolved coal in a compact reactor volume. As shown in Table I, below, bench scale laboratory studies indicate that at identical operating conditions, coal conversion in a plug flow reactor, as measured by percent MEK (methyl ethyl ketone) solubles, is slightly higher than that in a stirred tank. But more importantly, cyclohexane conversion of the liquid product obtained in a plug flow reactor is substantially higher than for the stirred tank product. (Because a product slurry with cyclohexane solubles is easier to process, cyclohexane conversion is considered an index of product quality.)

TABLE I

Comparisons of Plug and Stir Tank Reactor Performance on Coal Liquefaction*

Reactor Type	Coal Size	Conversions (wt.%MAF** Basis)		
		MEK	Be nzene	Cyclohexane
Plug Flow	200 mesh	80.5	59.4	30.1
Stirred Tank	100 mesh	73.6	42.5	6.5

*Liquefaction conditions: Nominal Residence Time =30 minutes; Tetralin/Coal =1.2/1; H_2 Added: 3.5 (wt.)% on dry coal; Temperature: 750° F.; and Pressure: 1,000 p.s.i.

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**Moisture and ash-free basis.

Before proceeding to a detailed description of a specific form in which the invention may be embodied, it will be helpful in the understanding of the invention to set forth the reaction conditions which preferably are employed in the liquefaction reactor system.

In a coal liquefaction reactor system, a slurry of coal particles in a liquid which is a solvent for dissolvable organic portions of the coal is heated under pressure to speed dissolution of the dissolvable coal and to cause the dissolved coal to depolymerize to form free radicals. The free radicals can recombine to form material which is insoluble even in pyridine unless a "chain stopper," preferably hydrogen, is present. Preferably, a solvent is employed which itself contains hydrogen-donor compounds. Thus, a hydrogen-donor stream suitably may include one or more hydrogen-donor compounds in admixture with nondonor compounds or with one another. Suitable hydrogen-donor compounds are indane, C_{10}–C_{12} tetralins, C_{12} and C_{13} acenaphthenes, di-, tetra-, and octahydroanthracene, and tetrahydroacenaphthene. A suitable hydrogen-donor solvent boils within the range from about 300° F. to about 900° F., preferably from about 375° F. to about 800° F., at atmospheric pressure, so as to remain in the liquid phase at the elevated temperatures employed in the liquefaction zone.

The degree to which recombination is prevented in the liquefaction zone depends upon the amount of hydrogen donor which is present. Desirably, the donor solvent will contain at least 30 weight percent, preferably at least 50 weight percent, of hydrogen-donor compounds. Where the donor solvent contains about 50 weight percent of hydrogen-donor compounds, and the solvent/coal ratio is from about 0.8:1 to about 2:1, which is preferred, the concentration of hydrogen-donor constituents in the liquefaction zone can be seen to range initially from about 22 weight percent to about 33 ⅓ weight percent and will be depleted by the liquefaction reaction. However, molecular hydrogen can be added to the liquefaction zone to partially replenish the hydrogen-donor solvent by a situ hydrogenation of the depleted molecules. For example, naphthenes may be hydrogenated to tetralins. Thus, it is preferred to add to the liquefaction zone a hydrogen-rich gas which provides from about 0.1 to about 10 weight percent of hydrogen.

Reaction conditions employed in the liquefaction zone include a temperature high enough to permit the hydrogen-transfer reaction between the hydrogen-donor solvent and the moisture and mineral free portions of the coal. The pressure is high enough to prevent excessive vaporization of the solvent at the temperatures employed in consideration of the gas generated and introduced into the liquefaction zone. Suitable liquefaction conditions include a temperature within the range from about 700° F. to about 1,000° F. and a pressure within the range from about 350 p.s.i.g. to about 3,000 p.s.i.g. Thus, in a low severity process in which the pressure may range from about 350 p.s.i.g. to about 500 p.s.i.g., a temperature of 775° F. may be employed in the liquefaction zone. In a moderate severity condition, in which the pressure may range, fro example, from about 1,500 p.s.i.g. to about 3,000 p.s.i.g., a temperature of about 850° F. is suitably employed.

The foregoing will provide a background of the conditions used in a liquefaction reactor system made in accordance with this invention, as illustrated in the drawings.

DESCRIPTION OF THE DRAWINGS

FIG. 1 is a schematic elevational view of an upflow reactor system embodying this invention for solvent liquefaction of a slurry of coal solids;

FIG. 2 is a longitudinal sectional view of one of the vessels subsequent to the leading vessel in the reactor system of FIG. 1; and

FIG. 3 is a cross section of the vessel of FIG. 2 taken along the lines 3—3.

DESCRIPTION OF THE PREFERRED EMBODIMENT

Referring to FIG. 1, an upflow reactor system for the solvent liquefaction of a slurry of coal solids is illustrated. The system is comprised of a leading reactor vessel **10** and subsequent downstream reactor vessels **11**, **12** and **13**, and includes a gas header **14** which serves as a discharge line for removal of gaseous materials from the top of each of the reactor vessels **10–13**. As illustrated, each of the reactors is an elongated, essentially vertically disposed vessel. Each vessel has an inlet in the bottom of it for introduction of slurry into it and an outlet in an upper portion of it for discharge of treated slurry from it. In leading vessel **10**, the bottom inlet is a narrow elongated entrance **15**. The outlet in leading vessel **10** is indicated by reference numeral **16**. In the vessels subsequent to reactor vessel **10**, the inlets are indicated respectively by reference numerals **17**, **18**, and **19**, and the outlets are indicated respectively by reference numerals **20**, **21**, and **22**. The reactor vessels are connected in series by confined fluid transfer lines extending from the outlet of a preceding upstream vessel to the inlet of a next subsequent downstream vessel. Confined transfer line **23** interconnects the leading vessel **10** to the next subsequent vessel. The downstream vessels **11**, **12**, and **13** are successively interconnected by transfer lines **24** and **25**, respectively. The terminal downstream vessel **13** discharges liquefied product containing some undissolved coal solids for further treatment in the production of liquid fuel products from solid coal.

As hereinbefore indicated, temperatures of above about 700° F. are employed in the reactor system. Such temperatures make pumping the slurry from one reactor to another reactor undesirable, because the slurry would have to be cooled for pumping and then reheated. To avoid pumping, pressure is controlled in each reactor vessel at a predetermined level to ensure a positive slurry flow in the reactor system. In one form of pressure control (not illustrated), all the reactor vessels are run at the same pressure, and the reactor vessels are designed so that the hydrostatic head in each reactor guarantees gravity feed to the next reactor. In a preferred pressure control system which is illustrated in the drawings, pressure is controlled in each reactor so that the pressure drop from one reactor to the next is large enough to guarantee gravity feed. Referring to FIG. 1, each vessel, except the terminal subsequent vessel 13, has a pressure controller operatively associated with it to impose a predetermined pressure on it which is sufficient to establish a pressure differential between it and the next subsequent vessel that is adequate to overcome the pressure drop between those vessels. As a result of the pressure differential, a positive slurry flow occurs in the reactor system, and slurry which flows from the leading vessel **10** subsequently passes through vessels **11**, **12** and **13**. The pressure in terminal vessel **13** is maintained at system pressure by a downstream control unit. Taking the pressure control system of reactor **11** as exemplary of the pressure control system in vessels **10** and **12**, reference is made to FIG. **2**. As depicted in FIG. **2**, means by which the pressure in vessel **11** may be regulated to a predetermined value includes a pressure controller **26** which controls a butterfly valve **27** in a gas discharge line **28** connected into a discharge orifice **29** in the top of vessel **11**. A level control **30** set for actuation on attainment of a predetermined level above the outlet **20** is operatively connected to pressure controller **26** to reset pressure controller **26** when the gas volume in a gas-disengaging space **32** between discharge outlet **29** and opening **20** shrinks to a predetermined value. If the system pressure in the reactor vessels is one which fluctuates significantly, differential pressure controllers are preferably utilized.

Referring now to FIG. **1**, feeding vessel **10** is designed to produce the superficial liquid velocities which separate the particles that are essentially organic in composition from the particles that are essentially nonorganic in makeup. The narrow elongated entrance **15** has a diameter selected, relative to a designed slurry feed rate range, to produce a superficial liquid velocity that is higher than the minimum fluidization

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velocity of the heaviest, largest particles in the slurry feed, so that all particles entering the reactor system are fluidized in the entrance. Above the bottom entrance, leading vessel **10** is expanded in cross-sectional area into an upper portion **31** which extends upwardly in uniform cross-sectional area to at least outlet **16**, providing means by which the slurry received from entrance **15** can be flowed upwardly in the vessel to outlet **16** at a superficial liquid velocity that is lower than the minimum fluidization velocity. The cross-sectional area is selected (as hereinafter illustrated by example) to provide a superficial liquid velocity in upper portion **31** which is sufficient to fluidize and carry upwardly substantially all the particles that are essentially organic in composition but which is insufficient to fluidize and carry upwardly a major portion of the particles that are essentially nonorganic in nature. Leading vessel **10** is provided with a length-to-diameter ratio sufficient to give sufficient residence time in leading vessel **10** for dissolution of at least a major portion of the dissolvable coal particles. This residence time will vary depending on the liquefaction conditions used and the extent of dissolution that has already occurred before introduction of the slurry into the reactor system. Where as much as 50 percent dissolution has already occurred, very little residence time is actually necessary; a minute or less will suffice under the more severe liquefaction conditions mentioned hereinbefore. Preferably, however, the residence time is longer in order that substantially all the dissolvable coal dissolves. In this case, the residence time suitably will range from about 2 to about 10 minutes.

The essentially nonorganic particles which settle from upper portion **31** are continuously withdrawn as a slurry through a side opening **32** placed between upper portion **31** and entrance **15**. The settled solids slurry withdrawal rate is selected so that the solvent removed with the settled solids does not exceed a predetermined maximum, suitably no more than 5 weight percent, and preferably no more than 2 weight percent, of the solvent fed into the system in the feed slurry. Although not illustrated, the withdrawn slurry may be fed to a liquid cyclone to separate the solids from the solvent, and the solids recovered and recycled for use to make up slurry fed to the reactor.

The particles which do not dissolve in the leading reactor are conveyed in a slurry into the bottom of the next subsequent vessel by means of transfer line **23**. Transfer line **23**, subsequent vessel **11**, and subsequent vessels **12** and **13** have cross-sectional configurations which are smaller than that of the upper portion **31** of the leading vessel **10** in order that the superficial liquid velocities in these subsequent vessels and in the transfer line is greater than the superficial liquid velocity in the upper portion of the leading vessel. This prevents the particles passed from the upper portion of the leading vessel from settling in the transfer lines or in the subsequent vessels.

The cross-sectional area of the subsequent vessels preferably is also selected to minimize the number of reactor vessels which will provide a predetermined minimum total residence time of the slurry in the system to reach a desired level of conversion. Thus, the cross-sectional area or diameter chosen should not be too small, as this would lead to an unreasonable number of vessels to achieve the total required residence time. Neither should the cross-sectional area or diameter be so large that it decreases the superficial liquid velocity in the subsequent vessels to a level which, in order to provide the necessary staging of superficial liquid velocities in the lead reactor and in the subsequent vessels, makes it necessary to so increase the slurry withdrawal rate in the first vessel that too much solvent is taken from the slurry introduced into the system.

In the subsequent vessels, the slurry received from the next preceding vessel is conducted upwardly in plug flow. To impose plug flow on the slurry and at the same time to minimize the number of reactor vessels providing the aforesaid superficial liquid velocity in excess of that velocity in the upper portion of vessel **10**, at least one vertical partition is placed between the inlet and the outlet of the subsequent vessel to substantially prevent backmixing of the three-phase fluidized bed flowing upwardly in the vessel. Referring particularly to FIG. **2**, a longitudinal cross section of subsequent vessel **11** (identical to vessels **12** and **13**) is depicted. In subsequent vessel **11**, intersecting vertical partitions **34** and **35** subdivide (see FIG. **3**) the cross-sectional area of vessel **11** into four channels fluidly communicated at the bottom **36** and top **37** of the partitions. As illustrated, the channels then have a diameter of one-half the diameter of the vessel **11**. The slurry received from vessel **10** is spread by a baffle transversely disposed across inlet **17** and spreads uniformly to flow upwardly through the channels in plug flow. A port located in the bottom of each channel of the subsequent vessel admits hydrogen into each subdivision quarter equally about the circumferences of the vessel. (No hydrogen is added by entry ports in the first vessel, sufficient hydrogen transfer being obtained in this vessel from fresh hydrogen-donor solvent.)

The total combined length of the subsequent vessels in the reactor system is chosen, with respect to the diameters used for such vessels, to provide a total residence time of the slurry in the total liquefaction reactor system of at least about 25 minutes, preferably about 35–40 minutes, or more.

The operation of the foregoing reactor system, and a specific reactor system configuration to provide an MEK conversion level in excess of 80 percent in a total residence time of about 35–40 minutes for a particular coal slurry makeup, is illustrated in the following example.

EXAMPLE

Coal having a mineral matter constituency of about 9.6 weight percent of the coal is ground to a size range of about 8 mesh (Tyler screen) and smaller. A specific gravity analysis of particles in a given screen size show that the particles of a given screen size have the following specific gravity range:

Specific gravity	1.6	1.61–2.01	2.01–2.43	2.43–5
Wt%	6.34	30.1	41	22.53

The particles will be made up in a slurry with a hydrogen-donor solvent boiling within the range from about 300° F. to about 900° F. The solvent/coal ratio will be 1.2/1. The slurry will be introduced into reactor vessels in which the reactor temperature is 775° F. and the reactor pressure in the final reactor is 365 p.s.i.g. The viscosity of the liquid in the reactor system will be about 0.5 centipoise throughout, and the specific gravity of the liquids in the reactor system will be about 0.8 throughout. It is specified that no more than 2 percent of the feed solvent is to be withdrawn from the reactor system. A conversion level of better than 80 percent MEK solubles is further specified. The reactor exit stream will be constituted of about 20 weight percent solids and about 80 weight percent solvent. It is convenient and suitable to constitute the slurry of settled solids withdrawn from the first vessel and the slurry in the transfer lines to have about 20 weight percent of solids and about 80 weight percent of solvent. Finally, it is specified that the coal feed rate to the leading vessel is to be about 575 tons/hr.

Given the foregoing, the minimum fluidization of the solids, as calculated by the correlation method developed by Zenz (see Zenz, F. A., and Othmer, D. F., "Fluidization and Fluid-Particle System," Reinhold Chemical Engineering Series, New York, 1960.), produces values which are plotted to develop the curve shown in FIG. **4**.

Referring to FIG. **4**, the particles which are constituted essentially of organic matter have a specific gravity of up to about 2.43. Particles which are principally composed of mineral matter have specific gravities which extend from about 2.5 or so up to about 5.0. The largest and heaviest particles in the coal feed are 8-mesh particles with a specific gravity of 5.0. As FIG. **4** illustrates, the superficial liquid velocity in the entrance **15** of the leading vessel **10** must then be at least 28 ft./min.

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The selection of the superficial liquid velocity in the upper portion **31** of the leading reactor is made so that substantially all of the particles in the feed coal which are essentially organic in composition, i.e., those particles of from about 8 mesh and smaller having a specific gravity of less than 2.43, are fluidized and carried upwardly in upper portion **31**. In addition, the selection of the superficial liquid velocity in the upper portion **31** is made so that the superficial liquid velocity is insufficient to fluidize and carry upwardly a major portion of the particles which are essentially nonorganic in nature, i.e., those particles having a size of 1.6 mm. and larger with a specific gravity of from about 2.5 and greater. Because it is specified that no more than about 2 percent of the feed solvent is to be withdrawn from the reactor system, with the coal feed rate to be used, and with the coal/solvent ratio to be used, the total weight of settled solids withdrawn from the leading reactor cannot exceed about 6,900 lb./hr. Selecting 8.6 ft./min. as the superficial liquid velocity in the upper portion **31** of the leading reactor, on a conservative basis some of the solids with a minimum fluidization velocity close to 8.6 ft./min. may be included for removal. (For example, referring to FIG. **4**, consider the solids in the size range from 1.8–2.38 mm. having a specific gravity distribution of 1.6–2.0. Some of these solids have a minimum fluidization velocity higher than 8.6 ft./min. and hence must be removed; however, part of the solids whose minimum fluidization velocity is below 8.6 ft./min. can be tolerated in that system. Nonetheless, all solids in the size range from 1.8–2.38 mm. with a specific gravity of 1.6 or higher are specified for removal to obtain a conservative solids withdrawal rate.) Then, on the basis of 100 lbs./hr. of dry coal in which the large, heavy solids constitute 1 lb./hr., one obtains:

Solid Size (mm)	Wt. of Solids in this Size (No.)	Solid Fraction Removed (%)
1.8–2.38	0.2	100%
1.5–1.8	0.1	63.5%
0.7–1.5	0.3	22.5%
0–0.7	0.4	0

From this, the weight of total solids withdrawn is 4,000 lbs./hr., resulting in a solvent withdrawal rate of 16,000 lbs./hr. which is equivalent to only 1.2 percent of the total solvent, well within the preferred maximum of 2 percent. Thus, 8.6 ft./min. is a suitable superficial liquid velocity for upper portion **31**.

With the foregoing specifications of feed rate and superficial liquid velocities etc., a suitable reactor system may have an entrance diameter in the leading vessel of about 4.5 ft, which provides a superficial liquid velocity of 36 ft./min., higher than the minimum fluidization velocity of 28 ft./min. The expanded upper portion **31** of the lead reactor suitably has a diameter of about 10 ft., producing the selected superficial liquid velocity of 8.6 ft./min. The subsequent vessels suitably have a diameter of about 8 ft. to assure suspension of all solids received from the first vessel. Superficial liquid velocities in the subsequent vessels then range from about 12.5 ft./min. to about 9.3 ft./min. The height of the vessels is selected relative to their diameters to provide the residence time specified. A reactor height of about 125 ft. for all of the vessels provides a residence time in the first vessel of about 5 minutes, during which time substantially all dissolvable coal solids dissolve, and it provides a residence time in the subsequent vessels ranging from about 6 to about 7 minutes, resulting in a total residence time of about 38 minutes.

FIG. 4, of course, merely illustrates the manner by which one can select a suitable set of superficial liquid velocities for a reactor system operated under the process limitations set out in the Example. Different process conditions, as hereinbefore described, will produce different curves and different superficial liquid velocities.

Having now described in detail our invention, modifications and changes in the means and modes of carrying it out will occur to those skilled in the art. To the extent that these changes and variations are embraced within the scope of the appended claims, or are the equivalent in spirit and substance thereto, we deem them covered by our claims.

We claim:

1. In an upflow reactor system of a leading vessel in series connection with and at least one subsequent vessel for solvent liquefaction of a slurry of coal particles, the improvement which comprises:

entrance means in the bottom of the leading vessel for introducing said slurry into said system at a superficial liquid velocity which is higher than the minimum fluidization velocity of the heaviest, largest particles in the slurry, whereby all entering particles in the slurry are fluidized in said entrance means,

slurry conducting means in an upper portion in the leading vessel for flow conducting said slurry upwardly therein to an exit at a superficial liquid velocity which is lower than said minimum fluidization velocity and sufficient to fluidize and carry upwardly substantially all of the particles which are essentially organic in composition but insufficient to fluidize and carry upwardly a major portion of particles which are essentially nonorganic in nature, for a residence time sufficient to permit dissolution of at least a major portion of dissolvable coal particles, whereby a major portion of particles which are essentially nonorganic in nature settle in the upper portion and are not carried over to a subsequent vessel,

means between said entrance means and said conducting means for continuous withdrawal of particles settled from said conducting means,

means in said subsequent vessel for flow conducting the slurry received from said leading vessel upwardly substantially in plug flow at a superficial liquid velocity in said upper portion of the leading vessel, so that particles passed from said conducting means do not settle in said subsequent vessel, and

pressure control means operatively associated with said leading and said subsequent vessels for imposing a predetermined pressure in such vessels, so that a positive slurry flow occurs in said reactor system.

2. An upflow reactor system for the solvent liquefaction of a slurry of coal particles which comprises:

a plurality of elongated, essentially vertical vessels for imposing liquefaction conditions on slurried coal in them, and including a leading vessel and at least one subsequent vessel,

said vessels each having an inlet in the bottom thereof for introduction of slurry thereinto and an outlet in an upper portion thereof for removing treated slurry therefrom,

said vessels being fluidly connected in series by at least one confined fluid transfer line extending from an outlet of a preceding vessel to the inlet of a next subsequent vessel,

said vessels each having in the top thereof a discharge orifice for gaseous material, a discharge line being connected to each of said orifices for removal of gaseous materials from each of said vessels,

said vessels, except the last vessel in the series, each having operatively associated therewith pressure control means for discharging said gaseous material so as to maintain the pressure in each vessel at a predetermined level higher than the pressure in the next subsequent vessel, whereby a positive liquid flow occurs in said reactor system,

said leading vessel further having:

a narrow elongated bottom entrance with a diameter which, for a chosen slurry feed rate, produces a superficial liquid velocity therein that is higher than the minimum fluidization velocity of the largest particles in the slurry feed, whereby all particles entering the reactor system are fluidized in said entrance,

an expanded upper portion below the outlet thereof and sized to provide a superficial liquid velocity which is

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lower than said minimum fluidization velocity and sufficient to fluidize and carry upwardly substantially all of the particles which are essentially organic in composition but insufficient to fluidize and carry upwardly a major portion of particles which are essentially nonorganic in nature, and to permit a sufficient residence time for dissolution of at least a major portion of dissolvable coal particles, whereby a major portion of particles which are essentially nonorganic in nature settle in the upper portion of the leading vessel and are not carried over to a subsequent vessel, and

a side opening between said entrance and said upper portion of the leading vessel for continuous withdrawal of slurried nonorganic particles settled from said upper portion,

said subsequent vessel further having:

a cross-sectional area smaller than the upper portion of said leading vessel such that the superficial liquid velocity therein is greater than in said upper portion, whereby particles passed from the upper portion of the leading vessel to the subsequent vessel do not settle in the subsequent vessel, and

at least one vertical partition between said inlet and said outlet of the subsequent vessel for imposing substantially plug flow upon the slurry admitted through said inlet,

said reactor system having hydrogen-rich gas inlet means at least subsequent to said leading vessel for introducing hydrogen gas into said slurry.

3. The reactor system of claim 2 in which the diameter of said entrance of said leading vessel provides a superficial liquid velocity at least sufficient to fluidize and carry upwardly particles having a particle diameter of 8 mesh and smaller with a specific gravity of 5 and lower, and in which said superficial liquid velocity in said upper portion of said leading vessel is sufficient to fluidize and carry upwardly particles having a particle diameter of 8 mesh and smaller with a specific gravity of about 2.4 and lower but insufficient to fluidize and carry upwardly particles which have a particle diameter of 1.6 mm. and larger with a specific gravity of about 2.5 and higher.

4. In a process for liquefying dissolvable coal particles slurried in a hydrogen-donor solvent, the improvement which comprises, continuously:

introducing a slurry of said particles in said solvent into the bottom of a leading part of an upflow reactor system at a superficial liquid velocity higher than the minimum fluidization velocity of the heaviest, largest particles in the slurry, whereby all particles in the slurry are fluidized and carried upwardly into an upper portion of said leading part,

flowing the slurry upwardly in said upper portion to an exit at a superficial liquid velocity which is lower than said minimum fluidization velocity and sufficient to fluidize and carry upwardly substantially all of the particles which are essentially organic in composition but insufficient to fluidize and carry upwardly a major portion of particles which are essentially nonorganic in nature, and which is lower than any superficial liquid velocity in a subsequent part of said reactor system, for residence time sufficient to permit dissolution of at least the major portion of dissolvable coal particles in said slurry, whereby a major portion of particles which are essentially nonorganic in nature settle from said upper portion and whereby all particles existing from said upper portion will not settle in a subsequent part of said reactor system,

withdrawing slurried settled nonorganic particles from an opening between the upper portion and the bottom of said leading part of said reactor system at a rate which will not vary the liquid-to-coal ratio for the slurry in the leading part of the reactor system more than a predetermined extent,

introducing a hydrogen-rich gas into said reactor system subsequent to said leading part thereof,

transferring said slurry in the upper portion of said leading part from said exit into the bottom of a subsequent upflow part of the reactor system,

conducting said slurry and said hydrogen-rich gas upwardly in said subsequent part of said reactor system in plug flow for a total residence time in said subsequent part sufficient to permit a selected level of coal liquefaction,

removing gaseous material from the tops of said leading and said subsequent parts of said reactor system so as to impose a predetermined pressure in said leading and said subsequent parts so that a positive slurry flow occurs in said reactor system.

5. The method of claim 4 in which said superficial liquid velocity in said bottom of the leading part of the reactor system is at least sufficient to fluidize and carry upwardly particles having a particle diameter of 8 mesh and smaller with a specific gravity of 5 and lower, and in which said superficial liquid velocity in said upper portion of said leading part is sufficient to fluidize and carry upwardly particles having a particle diameter of 8 mesh and smaller with a specific gravity of about 2.4 and lower but insufficient to fluidize and carry upwardly particles which have a particle diameter of 1.6 mm. and larger with a specific gravity of about 2.5 and higher.

6. A process of liquefying coal which comprises continuously:

forming a slurry of coal particles having a size range of about 8 mesh (Tyler) and smaller in a hydrogen-donor solvent boiling within the range from about 300° F. to about 900° F. and containing at least 30 weight percent of hydrogen donors chosen from the group consisting of C_{10} to C_{12} tetralins, indane, C_{12} and C_{13} acenaphthenes, di-, tetra-, and octahydroanthracene and tetrahydroacenaphthene, and mixtures of two or more thereof, at a solvent/coal ratio within the range from about 0.8:1 to about 2:1,

introducing the slurry into the bottom of a leading part of an upflow reactor system at a superficial liquid velocity greater than about 28 ft./min., whereby all particles in the slurry are fluidized and carried upwardly into an upper portion of said leading part,

flowing the slurry upwardly in said upper portion to an exit, at a superficial liquid velocity sufficient to fluidize and carry upwardly particles having a particle diameter of 8 mesh and smaller with a specific gravity of about 2.4 and lower but insufficient to fluidize and carry upwardly particles which have a particle diameter of 1.6 mm. and larger with a specific gravity of about 2.5 and higher, under liquefaction conditions including a temperature within the range from about 700° F. to about 1,000° F. and a pressure within the range from about 350 p.s.i.g. to about 3,000 p.s.i.g., for a residence time sufficient to permit dissolution of at least a major portion of dissolvable coal particles in said slurry, whereby a major portion of particles which are essentially nonorganic in nature settle from said upper portion of said leading part of said reactor system,

withdrawing said essentially nonorganic particles settled in slurried form from said upper portion from an opening between the upper portion and the bottom of said leading part of said reactor system at a rate such that the amount of solvent removed through said opening does not exceed about 5 percent of the solvent in the slurry introduced into the system,

transferring said slurry in the upper portion of the leading part from said exit therein into the bottom of a subsequent upflow part of the reactor system,

introducing a hydrogen-rich gas providing from about 0.1 to about 10 weight percent of hydrogen into the bottom of said subsequent part of said reactor system,

conducting said slurry and said hydrogen gas upwardly in said subsequent part of said reactor system in plug flow for a total residence time of at least about 25 minutes, and removing gaseous material from the tops of said leading and said subsequent parts of said reactor system so as to impose a pressure differential between said leading and said subsequent parts sufficient to overcome a pressure drop therebetween, so that a positive slurry flow occurs in said reactor system.

7. The process of claim **6** in which the superficial liquid velocity in the upper portion of the leading vessel is within the range from about 8 ft./min. to about 10 ft./min.

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