United States Patent

Bull et al.

[15] **3,640,816**

[45] **Feb. 8, 1972**

[54] MULTIPLE STAGE PROCESS FOR PRODUCING LIGHT LIQUIDS FROM COAL

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[22] Filed: **Dec. 8, 1969**

[21] Appl. No.: **883,182**

[52] U.S. Cl. **208/10**, 208/59

[51] Int. Cl. **C10g 1/00**

[58] Field of Search 208/10

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UNITED STATES PATENTS

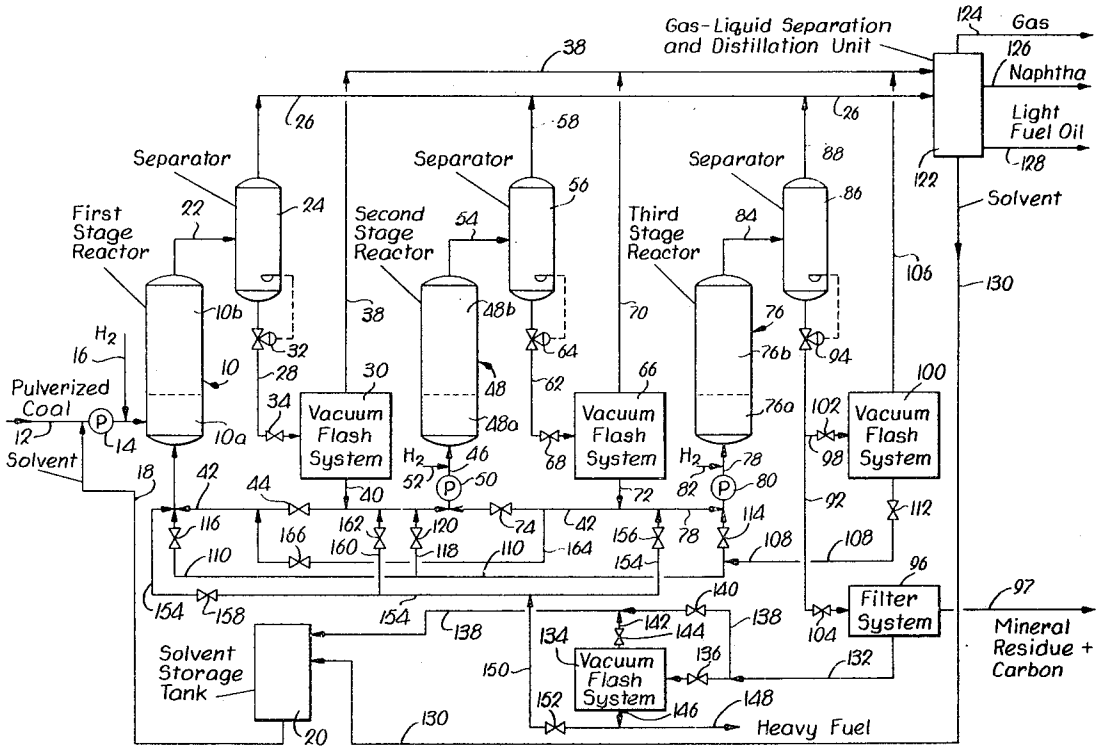
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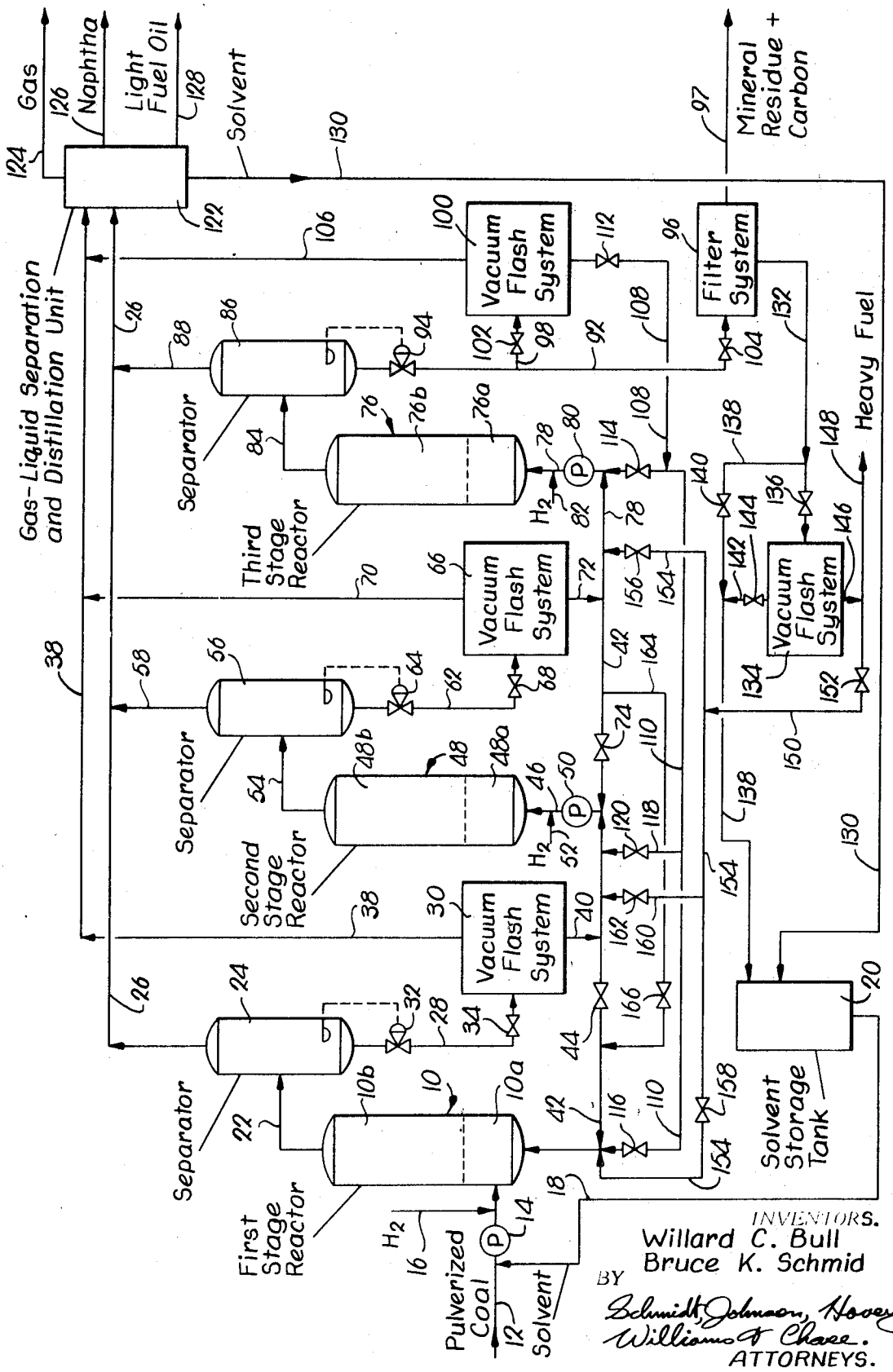
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[57] **ABSTRACT**

A multiple stage process for producing light liquids from coal wherein a slurry of pulverized coal, a solvent therefor and hydrogen is charged under pressure into a first reaction zone where the temperature thereof is elevated and maintained at such level until substantially all of the coal is dissolved. Gases and light liquids produced by partial hydrogenation of the reaction products are separated from the heavy bottoms and the latter are charged to another reaction zone under pressure where the charge along with an added quantity of hydrogen are heated to a higher temperature than present in the first zone so as to hydrocrack the constituents and produce additional quantities of gases and light liquids which are then separated from the heavy bottoms. The gases and light liquids from each stage are selectively segregated in a separation and distillation unit. Two or more reaction zones in series relationship may be employed with the charges thereto being subjected to treatment conditions of successively increasing severity accomplished by successively higher temperatures, pressures or residence times or combinations thereof. The heavy bottoms from one or more of the stages may be recycled back to preceding stages if desired.

10 Claims, 1 Drawing Figure





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MULTIPLE STAGE PROCESS FOR PRODUCING LIGHT LIQUIDS FROM COAL

This invention relates to a multiple stage process for dissolving and hydrogenating coal to produce light liquids therefrom. The light liquids have many advantageous uses and are especially suitable for incorporation into a refinery process for production of gasoline therefrom.

In particular, this invention relates to a process wherein pulverized coal is dissolved in a solvent therefor and hydrogenated at an elevated temperature and pressure to produce a reaction mixture containing gases, light liquids and heavy bottoms with the light liquids being directed to a combination separator and distillation unit while the heavy bottoms are treated in one or more subsequent stages of increasing treatment severity with the light liquids again being removed from the mixture prior to introduction thereof into a subsequent reaction stage. Removal of light liquids from the reaction mixture after each hydrogenation stage prevents cracking of the light liquids to an undesirable low molecular weight, but without decreasing the efficiency of the process in effecting dissolution of the coal and depolymerization thereof.

It has long been known that coal may be converted to fluid fuels, such as gasoline, by processes which involve the steps of transforming at least a part of the coal into liquid form, hydrogenating the dissolved coal, removing the ash therefrom, hydrocracking the ash-free product to produce a raw gasoline, and then reforming the gasoline to increase the octane content thereof. The problem of dissolving coal has heretofore been solved to a substantial extent but efficient hydrogenation has been expensive and difficult. Efforts over the years to provide efficient coal conversion processes have for the most part been directed to development of a process employing catalyst beds for effecting hydrogenation of the coal. Two types of catalytic bed processes have been investigated. In one, the dissolved coal along with undissolvable ash constituents were passed over the bed of catalyst. Although the cost of the catalyst could be economically borne in the initial expenditures for a coal conversion plant, rapid deactivation of the catalyst seriously limited the practicality of such process because of the high costs involved in replacing the catalyst at frequent intervals. Poisoning of the catalyst by the ash constituents presented such an acute problem though that no significant commercial processes have emerged from this work. Efforts to minimize the catalyst poisoning problem by first removing the ash from the liquefied material after dissolution of the soluble constituents in the coal were not satisfactory because of the inability to get all of the ash out of the liquefied product at a reasonable cost.

Noncatalytic coal conversion processes have heretofore been tried because of catalyst poisoning problems but were unsuccessful because of the low efficiency and high initial cost of such processes. In most instances, extremely high pressures were attempted to effect satisfactory solvation of the coal and suitable hydrogenation thereof. Lower pressure plants have been devised which were capable of effecting dissolution of a portion of the coal, but the liquids obtained from the coal were of relatively high molecular weight and of such aromaticity as to make them unsuitable for feed to a gasoline refinery process without further cracking.

In addition, a commercially feasible process for producing desirable light liquids from coal in a form usable for introduction into refinery gasoline-producing processes, must also necessarily be applicable to treatment of various types of coal products without significant change in the plant itself or the operating conditions thereof. In this instance, coal is intended to mean anthracitic, bituminous, subbituminous, as well as lignitic products. The high moisture content of peats and large proportion of ash in shales makes these products for the most part unsuitable for processing in accordance with the present method, primarily from the standpoint of economics rather than ability of the system to handle the carbonaceous fuel.

It is, therefore, the primary object of the present invention to provide a process for producing desirable light liquids from coal using a multiple stage treatment procedure which is operable without the utilization of catalytic vessels or chambers and which results in the production of liquid constituents which are suitable for direction to a refinery for production of gasoline therefrom without major further processing thereof being required. The present process is, however, useful for treatment of coal to produce fluid fuels therefrom even if further processing of the output is necessary or desirable before introduction of the light liquids into a subsequent refinery processing stage.

A particularly important object of the invention is to provide a process for producing light liquids from coal as described which does not require the use of catalytic beds, thus solving the problem of catalytic poisoning while at the same time completely obviating the necessity of removing undissolved constituents from the process stream during treatment thereof.

Another important object of the invention is to provide a process for producing light liquids from coal operable to permit treatment of various types of coal materials including those of an anthracitic, bituminous, subbituminous, or lignitic character with the only significant limitation being the cost of fabricating and operating the plant as compared with the value of the light liquids removed from the particular raw coal available for conversion.

Another very important object of the invention is to provide a process as described, wherein dissolution and hydrogenation of the coal is carried out without the necessity of employing catalytic beds as heretofore required by virtue of the utilization of multiple reaction stages operated under increasingly severe conditions, and wherein excessive cracking of the light liquids produced therein, to gases is avoided by removal of the same before the heavy bottoms are introduced into the next stage, thus permitting treatment of the dissolved coal at all times under conditions for most effective hydrogenation and hydrocracking thereof without adversely affecting the reaction products produced by the process.

A further important object of the invention is to provide a multiple stage process for producing light liquids from coal which especially lends itself to continuous operation wherein pulverized coal is dissolved, hydrogenated in successive reaction zones, light liquids removed therefrom after each stage, and undesirable constituents such as ash and heavy bottoms such as tars, removed, while at the same time producing a sufficient amount of a middle oil having the property of dissolving additional quantities of coal introduced into the process, to permit satisfactory operation on a continuous basis without the necessity of making up solvent from time to time.

A further important object of the invention is to provide a process of the characteristics above wherein hydrogen may be used as the hydrogenating agent, and unreacted hydrogen is recoverable for reintroduction into the process to increase the operating efficiency of the overall conversion process.

A still further important object of the invention is to provide a process for producing light liquids from coal wherein the heavy bottoms from each reaction stage may be selectively recycled either back to the immediately preceding, or any other preceding stages as desired for most effective processing of a particular coal product.

Another very significant object of the invention is to provide a multiple stage process for producing light liquids from coal without using catalytic beds and wherein all of the products to be recovered for introduction into say a refinery gasoline-producing process or the like go overhead, thus effectively eliminating all solids from the product output which could contaminate catalysts used in the gasoline plant.

Also an important object of the invention is to provide a process for producing light liquids from coal wherein multiple stages of increasing treatment severity are employed so that even though the partial pressure of the hydrogen available for hydrogenating the charge to each stage progressively decreases along the length thereof, the use of multiple treatment zones permits introduction of hydrogen at a number of points along the processing train to maximize the partial pressure of the hydrogen for most efficient use thereof.

Briefly, the present invention involves introducing pulverized coal, a solvent therefor, and hydrogen under pressure into a first reaction zone where the temperature is raised and maintained at such level until at least about 95 percent of the MAF coal is dissolved and partially depolymerized, whereupon the reaction products are passed through a separator for removing gases and light liquids and then into a vacuum flash system where lowering of the pressure on the admixture and pulling of a vacuum thereon causes additional quantities of gases and light liquids to be flashed off for removal from the system. Light liquids and gases removed from the first reaction zone products are directed to a combination separation and distillation unit for selective segregation thereof into specific fractions including gases, light liquids of the naphtha type, light fuel oil and middle oil useful as a solvent in subsequent processing of coal in accordance with the process. The heavy bottoms from the first reaction zone are introduced into a second reactor in conjunction with pressurized hydrogen and the temperature of the mixture raised to a level above the temperature level of the first reaction zone in order to subject the dissolved coal to more severe conditions than were imposed thereon in the first reactor. The mixture in the second reactor is maintained at such elevated temperature and pressure for a time period to further hydrogenate the dissolved coal without excessive cracking of the light liquids produced to gases. The effluent from the second reactor passes through a separator and another flash system to effectively remove gases and light liquids from the heavy bottoms from the second reactor. Light liquids and gases removed from the second reactor stage are also directed to the separation and distillation unit for selective segregation along with the products received from other reactors. As many reaction stages as deemed necessary may be employed, limited only by the capital costs of the processing plant as compared with the efficiency thereof represented by the output of desired light liquids from the distillation stage. The heavy bottoms from the final stage are filtered to remove ash therefrom and then passed through another flash system before being directed out of the plant as heavy fuel output. The overhead from the heavy fuel vacuum system may be employed as a part of the solvent for the pulverized coal.

The above system lends itself to operation on either a batch or continuous basis, although the latter is obviously the most efficient for large scale operations where the capital cost of the facility can be better justified from an economic standpoint.

The single DRAWING hereof is a schematic representation of apparatus for carrying out a continuous multiple stage process for producing light liquids from coal in accordance with the present invention, with it being understood that the equipment shown is representative only of components useful in carrying out the process and omitting many conventional controls, valves, pumps, and other miscellaneous units which would be employed in operation of the facility, the selection and disposition of which would be well known to those skilled in this art.

Pulverized coal from a grinding operation is conveyed to the first reactor vessel **10** via line **12** having a pump **14** therein for increasing the pressure of the admixture introduced into vessel **10** to a level within the range of about 500 p.s.i. to approximately 3,000 p.s.i. A hydrogenating agent, such as hydrogen, is introduced into line **12** through a line **16** located between pump **14** and vessel **10**. The pressure of the gas in line **16** (principally or wholly hydrogen) should be equal to or in excess of the pressure of the material forced into vessel **10** by pump **14**. Solvent for the pulverized coal is directed into line **12** through line **18** connected to a solvent storage tank **20**. Reactor **10** is shown only schematically in the drawing and could be made up of a single vessel having a lower preheating zone **10***a*, and an upper solvation and hydrogenation zone **10***b*, or this treatment stage may comprise two separate vessels if desired. The overhead line **22** from zone **10***b* of reactor **10** leads to a gas-liquid separator **24** which has a gas and liquid outlet line **26** connected to the top thereof which leads to a gas liquid separation and distillation unit **122**. Heavy bottoms line **28** extending from the bottom of separator **24** is connected to a vacuum flash system **30**. Liquid controller **32** in line **28** maintains a proper level of liquid in separator **24**, while valve **34** immediately preceding system **30** is preferably operable to lower the pressure on the reaction products flowing through line **28** to substantially atmospheric level or below before flowing into the flash system **30**.

Flash system **30** includes a number of units operable to lower the pressure on the material delivered thereto to a pressure of about 4 inches of Hg or less for efficient separation of gaseous materials from liquids introduced into the system from line **28** and includes a line **38** coupled to gas-liquid separation and distillation unit **122**, as well as a heavy bottoms line **40** which leads to supply line **42** connected to the lower end of first stage reactor **10**. In this manner, heavy bottoms from flash system **30** may be directed back into the reactor **10** if desired by virtue of the control valve **44** in line **42** downstream from line **40**.

Line **46** also joins line **42** to the lower end of a second stage reactor **48** having a preheating zone **48***a* and an upper hydrogenation zone **48***b*. Pump **50** is provided for increasing the pressure on the reaction products flowing through line **46** to a level of from about 500 p.s.i. to approximately 3,000 p.s.i. and a hydrogenation agent such as hydrogen is introduced into line **46** downstream from pump **50** via line **52**. Again the pressure of the hydrogen introduced into line **46** through line **52** should be at least equal to or in excess of the pressure of the reaction products pumped into reactor **48**.

The overhead line **54** in reactor **48** leads to a separator **56** similar to separator **24** and thereby provided with an outlet line **58** leading from the upper end thereof to line **26**. Line **62** from the lower end of separator **56** and having a liquid controller **64** therein extends to vacuum flash system **66** and is provided with a valve **68** therein for lowering the pressure on the reaction products to atmospheric level or below before they flow into system **66**. The gas and light liquid outlet line **70** for flash system **66** leads to line **38**, while heavy bottoms line **72** extends from flash system **66** to line **42**. The heavy bottoms from flash system **66** may be recycled back to second stage reactor **48** via line **42** under the control of valve **74** or returned to reactor **10** by virtue of the control provided by valve **44**. System **66** also includes vacuum units operable to lower the pressure on the material delivered thereto about 4 inches or less Hg.

At least a part of the heavy bottoms from flash system **66** is conveyed to the bottom of a third stage reactor **76** via line **78** connected to lines **42** and **72** at the point of juncture thereof, and provided with a pump **80** therein for increasing the pressure of the reaction products to a level of from about 500 p.s.i. to approximately 3,000 p.s.i. The hydrogenation agent inlet line **82** for introducing hydrogen into line **78** downstream of pump **80** also serves to permit hydrogen to be directed into the reaction product delivered to reactor **76** at a level at least equal to or exceeding the pressure of the reaction products emanating from pump **80**.

The overhead line **84** from reactor **76**, which also has a preheating zone **76***a* and a hydrogenation zone **76***b* extends to separator **86** similar to separators **24** and **56**. The overhead line **88** from separator **86** leads to product outlet line **26**. The reaction product outlet line **92** from separator **86** has a level controller **94** therein associated with separator **86** and leads to a filter system **96** having a mineral residue plus carbon outlet line **97** connected thereto. Branch line **98** between line **92** and vacuum flash system **100** has a valve **102** therein which is operable in conjunction with valve **104** in line **92** adjacent filter system **96** to permit selective control of reaction products to the flash system **100** and filter system **96** or in varying proportions thereto. The outlet line **106** from vacuum flash system **100** is coupled to line **38** while a liquid line **108** leads from flash system **100** to a line **110** between line **42** and line **78**. Valve **112** is provided in line **108** for controlling the flow of liquid to line **110**, while valve **114** in line **110** adjacent

line 78 permits selective control of liquid to line 78 from line 108 and a similar function is performed by valve 116 in line 110 adjacent line 42 for controlling flow of liquid to the line 42. Branch line 118 between line 110 and line 42 has a valve 120 therein which permits selective control of flow of liquid from line 110 into line 42 between reactors 10 and 48.

Vacuum system 100 includes a number of components shown only schematically in the drawing for reducing the pressure on the constituents to as low a level as economically practical, as for example, about 1 inch of Hg.

Lines 26 and 38 both lead to the gas-liquid separation and distillation unit 122 which is capable of segregating the products directed thereto into a gaseous fraction removed via line 124, a light liquids line 126 designated in the drawing as naphtha, a light fuel oil line 128, and a bottoms or middle oil liquid line 130 indicated as comprising the solvent for the system. It is to be understood in this respect that unit 122 is designed to handle the high-pressure constituents delivered thereto via line 26 as well as the low-pressure materials received from line 38 by inclusion of suitable equipment well known to those skilled in this art. Line 130 leads into solvent storage tank 20. The liquid outlet line 132 from filter system 96 extends to another vacuum flash system 134 also operable to decrease the pressure on the materials delivered thereto to a level of about 1 inch of Hg. Valve 136 in line 132 controls flow of liquid either directly into the system 134, or in bypassing relationship to the latter into solvent storage tank 20 via line 138 having a control valve 140 therein. The outlet line 142 from system 134 to line 138 has a control valve 144 therein, while the bottoms line 146 from system 134 leads to a heavy fuel outlet line 148. Line 150 provided with a control valve 152 therein extends from line 148 to a line 154 between lines 42 and 78. Control valve 156 adjacent line 78 and control valve 158 in line 154 adjacent line 42, permits selective introduction of liquids into lines 78 and 42 respectively. Branch line 160 leading from line 154 to line 42 between reactors 10 and 48 has a control valve 162 therein so that selective control is maintained over introduction of liquids from line 154 into line 42 between line 40 and line 118.

Return line 164 extending from line 42 between lines 46 and 72 to line 42 downstream of valve 44 has a control valve 166 therein so that liquids from flash system 72 may be selectively recycled back to reactor 10 if desired.

The above equipment shown for purposes of illustration only and in schematic form, is especially adapted for producing light liquids within the range of the boiling point of C_5 and above hydrocarbons up to a boiling point of about 250° to 350° C. and preferably about 300° C. Coal derived hydrocarbons above this range become difficult to utilize in a conventional refinery process for producing gasoline.

In accordance with the preferred operating parameters for the process of this invention, it is desirable that sufficient solvent be admixed with the pulverized coal introduced into reactor 10 to effect dissolution of substantially all of the coal in the solvent under the processing conditions imposed on the mixture in zones 10*a* and 10*b*. Since the solvent used in the system is ultimately that which is produced thereby, the only initial consideration insofar as the solvent in concerned, is selection of a liquid capable of dissolving the particular type of coal to be processed. Anthracitic, bituminous, subbituminous, and lignitic processes are all capable of being processed in the continuous system described herein, and A.S.T.M. classification D-388-38 is incorporated herein by reference as a description of the various types of coal products which can be advantageously treated in accordance with the present process to produce desirable light liquids therefrom. Most efficient results are obtained however when the raw coal contains less than 86 percent dry fixed carbon and has a dry volatile matter of 14 percent by weight or more with both analyses being made on a mineral matter free basis. A preferred raw coal product, however, may be designated as Kentucky No. 11 coal or an equivalent coal product and which is ground in a suitable attrition machine such as a hammermill to a size such that 80 percent of the coal will pass through a 100 mesh (u.S. Standard) sieve. A solvent particularly useful as a startup solvent for Kentucky No. 11 coal is anthracene oil or creosote oil having a boiling range of about 220° to 400° C. However, as noted, the selection of a specific startup solvent is not critical since during the process dissolved fractions of the raw coal form substantial quantities of additional solvent which provide an amount of solvent sufficient to replace any solvent that is lost during the process. Thus, regardless of what the original solvent may have been, it loses it identity and rapidly approaches the constitution of the solvent formed by solution and depolymerization of the raw coal fed into the process. For this reason, the solvent which is employed may be broadly defined as that obtained from a previous treatment of coal in accordance with the steps of this invention. Solvents produced from coal in the present process will be of a highly aromatic nature and generally have a boiling range of about 150° C. to approximately 600° C., with a typical solvent obtained from a raw input such as Kentucky No. 11 coal, comprising middle oil having a boiling range of from about 190° to 300° C. Best results are obtained when the solvent has a boiling range of 280° to 430° C.

The ratio of startup solvent to coal may be varied so long as a sufficient amount is employed to effect dissolution of substantially all of the coal in the solvent within reactor 10, but best results are obtained when the ratio of coal to solvent is about 1:1. Generally, the ratio of startup solvent to coal should be within the range of 0.6:1 to 4:1 and preferably 0.8:1 to 2:1. Ratios of startup solvent to coal greater than 4:1 may be used but provide no significant functional advantage in the solvation process of this invention and suffer the additional disadvantage of requiring added energy or work for subsequent separation from the system.

Because the system shown and described is of a continuous nature, with solvent for the coal being derived from the process streams themselves, the ratio of solvent to coal at any particular time after the system has stabilized, will of course, be established by selective control of processing conditions imposed on the coal. Generally speaking though, best results are obtained by maintaining the ratio of solvent to coal at the inlet to reactor 10 at about 1:1. Again though, the processing conditions can be varied to alter the ratio of solvent to coal on a continuous basis within the range of 0.6:1 to 4:1 and preferably 0.8:1 to 2:1.

The slurry formed by admixture of coal in line 12 with solvent from tank 20 introduced into line 12 via line 18, is pumped into reactor 10 at a pressure of from about 500 p.s.i. to approximately 3,000 p.s.i. and preferably 1,000 to 2,000 p.s.i. The admixture is raised in temperature by the preheater 10*a* to a level within the range from about 350° C. to approximately 500° C. by virtue of introduction of heat to the admixture from an external source. A preferred temperature range of reactor 10 is 350° to 425° C. At the preferred operating pressure, the reaction products in the first reaction zone should be raised to a temperature level of 375° to 400° C. Hydrogen is introduced into the slurry at a pressure and at a rate to maintain the average partial pressure thereof in the reactor 10 within the range of about 300 to 2,500 p.s.i. and preferably 700 to 1,500 p.s.i. with best results being obtained when the average partial pressure of the hydrogen is maintained at about 900 p.s.i. The charge to reactor 10 flows along the length thereof on a continuous basis and heat is supplied to the reactants for a time period to maintain the admixture at the desired temperature level and for a time sufficiently long to effect dissolution of 95 percent of the MAF coal added to the reactor.

At the optimum pressure and temperature conditions in first stage reactor 10, a residence time of about 15 to 30 minutes is sufficient to dissolve over 95 percent of the MAF coal in the presence of a solvent therefor as defined. In order to provide assurance that substantially all of the coal is dissolved in the solvent therefor, a better criterion for solvation is determination of the relative viscosity of the solution rather than

reliance entirely on residence time. Relative viscosity in this sense is the ratio of the viscosity of the solution to the viscosity of the solvent, as fed to the process, both viscosities being measured at 210° F. Accordingly, the term "relative viscosity" as used herein and in the claims is designated as the viscosity at 210° F. of the solution in first stage reactor **10** divided by the viscosity of the solvent at 210° F. introduced into line **12** via line **18**. The relative viscosity of the solution has been found to generally rise above a value of 20 to a point at which the solution is extremely viscous and in a gellike condition. In fact, if solvent coal ratios lower than those specified are used, the slurry sets up into a gel. After reaching the maximum relative viscosity, usually above the value of 20, the relative viscosity begins to decrease to a minimum level. Thus, the pulverized coal and solvent introduced into reactor **10** preferably are maintained in the rector under the pressure and temperature conditions specified and in the presence of the hydrogenation agent until the viscosity has decreased after initially increasing.

The reactants in reactor **10** should normally be maintained therein until the relative viscosity falls to a value of at least 10, and preferably less than 10, with best results in the range of 1½ to 2. The reaction products produced in reactor **10** go overhead through line **22** into separator **24** where at least a certain part of the gaseous constituents including those which are liquids at room temperature, separate to a certain extent from the liquid bottoms and go overhead through line **26** to gas-liquid separation and distillation unit **122**. The overhead from separator **24** is principally light hydrocarbons such as C_1 to C_4 hydrocarbons and naphthas, unreacted hydrogen, H_2S and CO_2. The constituents which are liquid at the temperature and pressure conditions of separator **24** (at essentially the same pressure as reactor **10** but at a somewhat lower temperature) are directed into flash system **30**, after undergoing expansion through valve **34**, where they are then lowered to a pressure of about 4 inches of Hg. A substantial proportion of the gases and light liquids which boil within the range of C_5 hydrocarbons and above up to about 300° C. are thereby segregated from the liquid bottoms under the low pressure conditions imposed thereon and are conveyed to unit **122** via line **38**. The liquid bottoms from the reaction products introduced into flash system **30** are conveyed to second stage reactor **48** after being raised in pressure by pump **50** to a level within the range of about 500 p.s.i. to approximately 3,000 p.s.i. and preferably from 1,000 to 2,000 p.s.i. with best results being obtained as previously noted at about 1,000 p.s.i. The liquid bottoms from flash system **30** are directed to reactor **48** via lines **40**, **42** and **46** but do not recycle to first stage reactor **10** via line **42** because it is assumed in this mode of operation of the apparatus that valve **44** is closed. Sufficient hydrogen is also introduced into the charge to reactor **48** via line **52** under a pressure such that the average partial pressure thereof in reactor **48** is maintained at a level of about 300 to 2,500 p.s.i. and preferably 700 to 1,500 p.s.i. with best results again being obtained at about 900 p.s.i. with the particular coal product referred to in this specific example.

The charge to reactor **48** is again heated by an external source within preheater zone **48***a* to a temperature level of from about 325° C. to approximately 525° C. and preferably to 375° to 450° C. with best results being obtained at about 400° C. when the pressure on the mixture is 1,000 p.s.i. The conditions thus imposed on the charge to reactor **48** effect some hydrocracking of the constituents under the conditions imposed thereon. Controlled hydrocracking of the mixture is enhanced by maintenance of the temperature of the reactants within the temperature ranges specified throughout the length of hydrogenation zone **48***b*. The residence time of the constituents in reactor **48** however, should not be of such duration as to cause deleterious hydrocracking of the light liquids which boil within the range of C_5 hydrocarbons or above up to about 300° C. Therefore, the residence time of the charge to reactor **48** in zones **48***a* and **48***b* thereof should not exceed about 5 to 100 minutes after the temperature of the reactants has been increased to the specified level with the preferred residence time being about 10 to 30 minutes and best results being obtained using a 15 minutes residence parameter. It is to be pointed out that preferred operation of the process involves subjecting the charge to reactor **48** to somewhat more severe treatment than the charge to first stage reactor **10**. Generally, this increased severity of treatment can best be accomplished by raising the temperature of the charge to second stage reactor **48** to a level above the temperature of the charge in first stage reactor **10** although higher pressures and increased residence time or combinations thereof may also be used to accomplish the desired end.

The reaction products from reactor **48** are introduced into separator **56** via overhead line **54** whereby gaseous constituents therein are permitted to flow into separation and distillation unit supply line **26** via line **58**. The liquid constituents directed to separator **56** are conveyed to vacuum flash system **66** through line **62** after being reduced in pressure across expansion valve **68** and then subjected to vacuum conditions of about 4 inches of Hg in system **66**. As a result, the additional gaseous constituents and light liquids produced in reactor **48** and which boil within the range of C_5 hydrocarbons and above up to about 300° C. are flashed off and directed to separation distillation unit supply line **38** via line **70** connected thereto. Flash system **66** also operates at a somewhat lower temperature than that of the reactor **48**. The liquid bottoms from flash system **66** are conveyed to third stage reactor **76** via lines **72** and **78** after being increased in pressure to the same limits described with respect to reactors **10** and **48** by pump **80**. Additional quantities of hydrogen are also introduced into the liquid bottoms directed to reactor **76** from flash system **66** at a pressure and a sufficient rate to maintain the average partial pressure of the hydrogen in reactor **76** within the range of about 300 to 2,500 p.s.i. and preferably 700 to 1,500 p.s.i. with best results being obtained at about 900 p.s.i. with the operating conditions previously specified and the particular type of coal set forth in this example.

In this mode of operation of the apparatus illustrated, it is also assumed that valve **74** is closed so that none of the liquid bottoms can flow back into the bottom of second stage reactor **48**. The reactants in third stage reactor **76** are subjected to more severe processing conditions than carried out in second stage reactor **48** so as to hydrocrack those constituents which were not broken down and hydrogenated in the second stage. The charge to third stage reactor **76** is heated to a temperature within the range of about 350° C. to approximately 550° C. and preferably within the range of 375°, 525° C. with best results obtained at 425° C. with a pressure of 1,000 p.s.i. with corresponding residence times of 5 to 100, 10 to 30, and 15 minutes respectively.

The overhead from reactor **76** flows into the third stage separator **86** for separation of gaseous constituents from the liquid components of the reaction products whereby the gases flow into line **26** through line **88** and the remaining liquid bottoms flow into vacuum system **100** and filter system **96** in proportional amounts governed by the respective settings of control valves **102** and **104**. Removal of a required amount of residue as inorganic compounds and carbon is best obtained by operating the system **96** in a manner to cool the input thereto to a level of about 300° C. Preferably, valves **102** and **104** are set to maintain a predetermined proportion of solids in the treatment system since it is contemplated that at least a part of the bottoms from system **100** be recycled back to the first stage reactor **10** or to the second and third stage reactors as desired under the control of valves **116**, **120** and **114** respectively. Thus, valves **102** and **104** are set to assure removal from the system of an amount of filter cake equal to the undissolvable portion of the coal fed to the treatment apparatus through line **12**. However, the valve setting can be changed as desired, during startup or operation, for a period necessary to change the solids level in the overall process as selected by the operator.

The pressure on the bottoms directed to system **100** via line **98** is let down across valve **102** and the pressure on the bottoms introduced into system **100** is lowered to as low a level as practical, usually about 1 inch of Hg to insure removal of as large a proportion of gases and light liquids from the heavy bottoms as feasible when the operating expenditures and capital costs of the flash system are correlated. The overhead constituents from system **100** are introduced into separation and distillation unit **122** via line **106** and line **38**.

The bottoms from filter system **96** are directed into vacuum flash system **134** via line **132** to effect separation of the solvent for the process from heavy constituents such as tars and the like which are removed via heavy fuel line **148**. These heavy constituents are normally solid at room temperature but are in liquid form at the temperature of about 300° C. at which the material is handled in system **134**. In this mode of operation of the apparatus, valve **136** is open to admit liquid to system **134** while valve **140** is closed, preventing flow of liquid to tank **20** in bypassing relationship to the system **134**. As a consequence, letdown in pressure of the liquid introduced into vacuum flash system **134** and decrease of the pressure thereon to as low a level as practical as previously noted (usually about 1 inch of Hg) causes the lighter fraction which is useful as a part of the solvent phase of the present system to go overhead to line **138** via line **142** for delivery to solvent storage tank **20**, while the heavy bottoms go out through line **148** since valve **152** in line **150** is normally closed. The amount of solvent recovered for recycling to the system will generally comprise 100 percent of the amount of solvent required to maintain the desired ratio of solvent introduced into first stage reactor **10**. In the event it is desired to reprocess a part or all of the heavy fuel bottoms from system **134**, valve **152** may be opened to permit such bottoms to flow to any of the reactor stages via line **150** and line **154** provided with control valves **158** and **156** therein or branch line **160** having control valve **162**.

In addition, the liquid from filter system **96** may be bypassed around system **134** directly into solvent storage tank **20** if desired by closing valve **136** and opening valve **140**.

In other modes of operation of the process, a part or all of the bottoms from respective reactors may be recycled back to any preceding reactors by selective operation of valves **44** and **74**, as well as valves **114**, **116** and **120**.

Although the system schematically illustrated and specifically described above involves the utilization of three reaction zones wherein the dissolved coal is subjected to successively more severe processing conditions, it is to be understood that although more than one treatment zone is required, the actual number employed may be varied as necessary to obtain the desired output when compared against the costs of constructing and operating a particular facility. For example, it is not necessary to carry out the process on a continuous basis but a batch operation is usable, particularly in those cases where a large number of stages are desired for optimum results. A material balance for a batch operation is set forth in the Table hereunder with the conditions of processing thereof indicated for each stage.

The continuous process described above is preferred because it is more effective and efficient than the batch process for at least these reasons:

1. Contact between the hydrogenating gas and the liquid is better in the continuous system where the hydrogen is passing upward through the liquid in the vessel whereas in a batch system most of the hydrogen tends to remain in the free space above the liquid.
2. The light liquids continuously are removed from the reactor in a continuous system, thereby minimizing the possibility of further converting the valuable light liquids to gas while in a batch system the light liquids are not removed from the reactor until the end of the cycle, and can be further converted to gases.
3. Continuous removal of light hydrocarbons, CO_2, H_2O, and H_2S from the reactor in a continuous system results in maintenance of a higher average effective partial pressure of the hydrogen than in a batch system at the same total pressure.

Having thus described the invention, what is claimed as new and desired to be secured by Letters Patent is:

1. A process for producing light liquids from coal comprising the steps of:

a. charging an admixture pressurized to at least 500 p.s.i., said admixture consisting essentially of particulate coal, a hydrogenation agent therefor consisting essentially of hydrogen, and a high molecular weight coal derived solvent for the coal to a first reaction zone, the ratio of said

BATCH PROCESSING TABLE

Stage	1 (Aver-age)	2	3	4	5	Total
Temperature ° C	400	400	425	450	450	
Pressure, p.s.i.g	3,000	3,000	3,000	3,000	3,000	
Time, minutes	60	60	60	60	60	300
Feed:						
Hydrogen	0.50	0.60	1.10	2.70	1.30	6.20
Solvent	100.00	49.40	30.81	16.05		100.00
Coal	100.00	58.98	53.84	42.99	25.86	100.00
Ash and insolubles		13.60	13.60	13.60	13.60	
Total	200.50	122.58	99.35	75.34	40.76	206.20
Products out:						
CO_2	1.45	0.14	0.13	0.13	0.07	1.92
H_2S	2.28	0.12	0.22	0.15	0.06	2.83
Gas (C_1-C_5)	2.71	1.75	4.18	12.02	6.14	26.80
H_2O	6.71	0.84	0.49	0.41	0.07	8.52
Cold trap oil	1.60	0.76	1.14	1.61	0.46	5.57
Cut #1 (25°-135° C.)	13.17	2.13	5.79	7.56	1.48	30.13
Cut #2 (135°-180° C.)	50.60	18.59	14.76			86.95
Cut #3 (135°-230° C.)				14.00	4.77	18.77
Total light liquids (including solvent)						141.42
Residue	121.98	98.25	72.64	39.46	27.71	
Solvent	(49.40)	(30.81)	(16.05)			
Deashed coal prod	(58.98)	(53.84)	(42.99)	(25.86)	(14.11)	14.11
Ash and insolubles	(13.60)	(13.60)	(13.60)	(13.60)	(13.60)	13.60
Total	200.50	122.58	99.35	75.34	40.76	206.20
Excess liquids						41.42

NOTES:
1. Hydrogen consumption calculated from elemental balance or by volumetric measurement data.
2. Material balance corrected for loss or gain by proportioning liquid reactor products. Hydrogen sulfide corrected to loss free basis by elemental balance.
3. Numbers in parenthesis calculated by assuming 13.60 lb. of ash and inerts and 100 lb. of solvent.
4. Excess liquids represent all liquid products in excess of 100 lb. of recycle solvent.
5. All quantities in lb. per 100 lb. of original coal charge.
6. All distillation temperatures at 3 mm. Hg pressure.

solvent to said coal within said admixture being from about 0.6:1 up to 4:1;

b. raising the temperature of said admixture in said first reaction zone to at least 350° C. and maintaining said admixture at such temperature for a period not to exceed 100 min. to effect dissolution of a portion of the coal in the solvent and hydrogenation of at least a part of the dissolved coal to produce a first reaction mixture which includes gases, light liquids and heavy bottoms;

c. flash separating by pressure reduction said gases and said light liquids from said heavy bottoms of said first reaction mixture;

d. charging a portion of said heavy bottoms from said first reaction mixture and a hydrogenation agent therefor consisting essentially of hydrogen, under a pressure of at least 500 p.s.i. to a second reaction zone;

e. raising the temperature of said charge to said second reaction zone to at least 350° C. and maintaining said charge at such temperature for a period not to exceed 100 min. to dissolve at least a portion of any undissolved coal in said charge to said second reaction zone and to further hydrogenate a part of said coal dissolved therein to produce a second reaction mixture which includes gases, light liquids and heavy bottoms; and thereafter

f. flash separating by pressure reduction said gases and said light liquids from said heavy bottoms of said second reaction mixture.

2. A precess as set forth in claim **1**, wherein step (b) is continued until at least approximately 95 percent of the charge of coal to the first reaction zone is dissolved in said solvent.

3. A process as set forth in claim **1**, wherein is included the steps of charging said admixture to said first reaction zone in accordance with step (a) under a pressure of from about 500 p.s.i. to approximately 3,000 p.s.i., raising the temperature of said admixture to a temperature level as provided in step (b) of from about 350° C. to approximately 500° C. and maintaining said admixture within said temperature range for a period of about 15 to approximately 30 min.

4. A process as set forth in claim **1**, wherein is included the steps of charging a portion of said heavy bottoms from said first reaction mixture to said second reaction zone in accordance with step (d) under a pressure of from about 500 p.s.i. to approximately 3,000 p.s.i., raising the temperature of said charge to said second reaction zone to a temperature level as provided in step (e) within the range of from about 350° C. to approximately 525° C. and maintaining said charge to said second reaction zone within said temperature range for a period of about 5 to approximately 100 min.

5. A process as set forth in claim **1**, wherein is included the steps of:

g. charging a portion of said heavy bottoms from said second reaction mixture and a hydrogenation agent therefor consisting essentially of hydrogen, under a pressure of at least 500 p.s.i. to a third reaction zone;

h. raising the temperature of said charge to said third reaction zone to at least 350° C. and maintaining said charge at such temperature for a period not to exceed 100 min. to dissolve at least a portion of any undissolved coal in said charge to said third reaction zone and to further hydrogenate a part of said coal dissolved therein to produce a third reaction mixture which includes gases, light liquids and heavy bottoms, the reaction conditions including pressure, temperature and residence time within said third reaction zone being more severe than the reaction conditions within said second reaction zone; and thereafter

i. flash separating by pressure reduction said gases and said light liquids from said heavy bottoms of said third reaction mixture.

6. A process as set forth in claim **5**, wherein is included the steps of charging a portion of said heavy bottoms of said second reaction mixture to said third reaction zone in accordance with step (g) under a pressure of from about 500 p.s.i. to approximately 3,000 p.s.i., raising the temperature of said charge to said third reaction zone to a temperature level as provided in step (h) within the range of from about 350° C. to approximately 550° C. and maintaining said charge to said third reaction zone within said temperature range for a period of about 5 to approximately 100 min.

7. A process as set forth in claim **1**, wherein is included the step of treating a part of the heavy bottoms from the last reaction zone to remove solids therefrom and returning such solids-free heavy bottoms to the charge to at least one of the reaction zones as makeup for said solvent.

8. A process as set forth in claim **1**, wherein said solvent is a solvent obtained from a previous processing of coal in accordance with said process.

9. A process as set forth in claim **1**, wherein is included the steps of directing the reaction mixture from each reaction zone into a separator, and thereafter lowering the pressure on the reaction mixtures to flash off the gases and light hydrocarbons.

10. A process as set forth in claim **1**, wherein is included the steps of continuously charging the reaction zones while simultaneously removing reaction mixtures therefrom and returning at least a part of the heavy bottoms from the last reaction zone to a preceding reaction zone while charging the heavy bottoms from the first reaction zone and each reaction zone thereafter into the next successive reaction zone.

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