Aug. 29, 1972 R. J. FIOCCO ET AL 3,687,837

COAL LIQUEFACTION SOLIDS REMOVAL

Filed Aug. 27, 1970 3 Sheets-Sheet 1

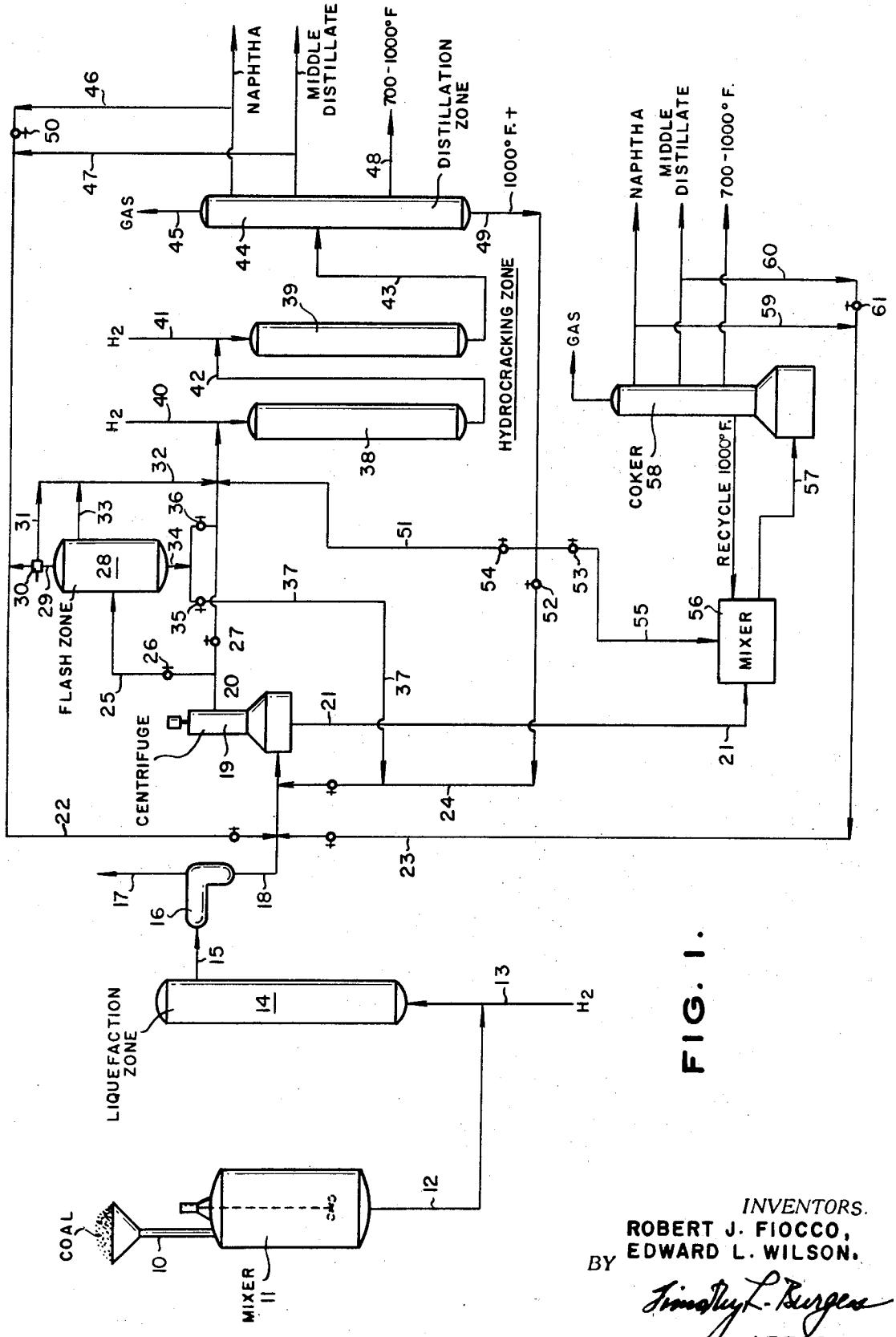


FIG. 1.

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COAL LIQUEFACTION SOLIDS REMOVAL

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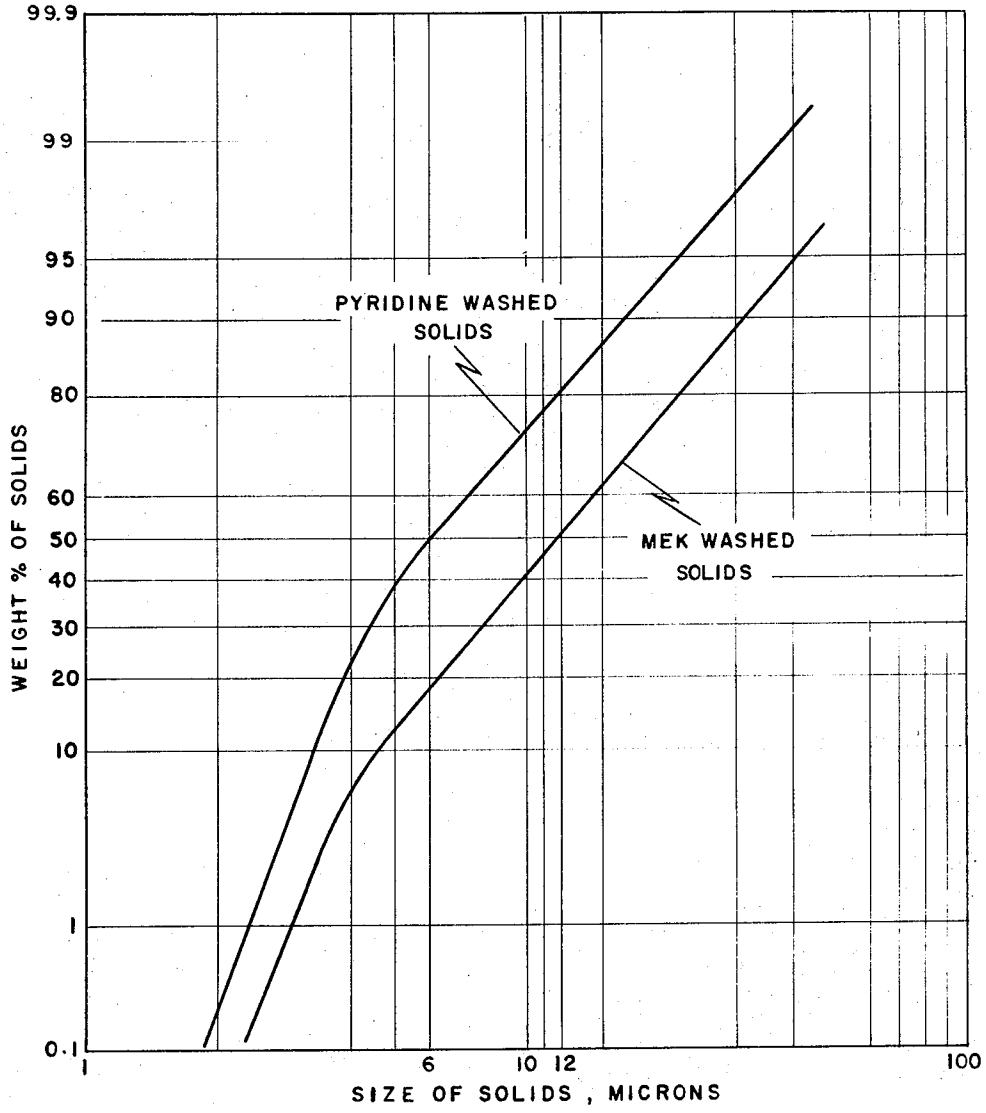


FIG. 2.

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COAL LIQUEFACTION SOLIDS REMOVAL

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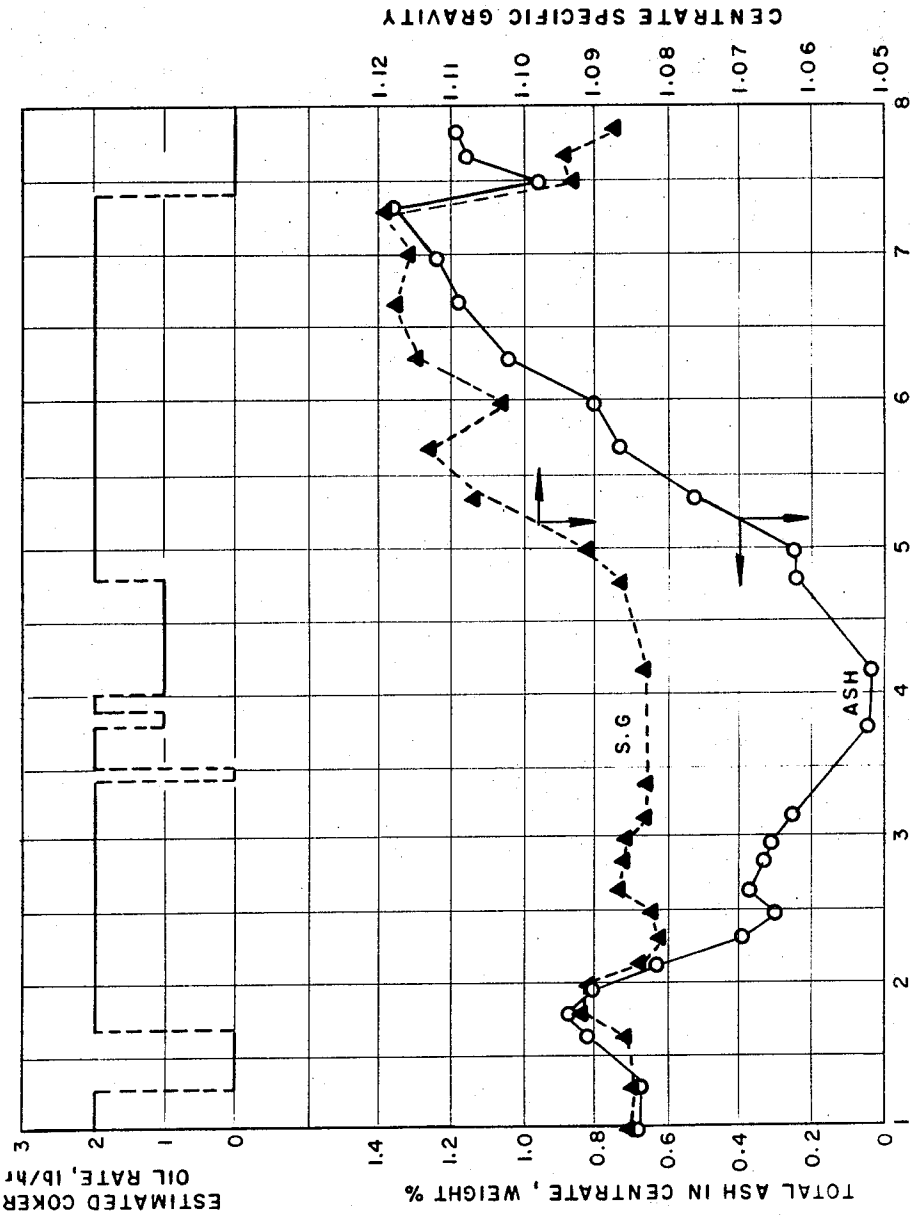


FIG. 3.

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COAL LIQUEFACTION SOLIDS REMOVAL

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U.S. Cl. 208—8 7 Claims

ABSTRACT OF THE DISCLOSURE

The clarified extract issued from a solids-liquids separation zone (in which the solids are separated from the liquids of a coal liquefaction product according to the size of the solids and concentrated in a separately discharged solids slurry) is improved by adding to the coal liquefaction product a recycle stream selected from a first fraction, boiling within the range from about 100° F. to about 700° F. and recovered from either the clarified extract or the concentrated solids slurry issued from the separation zone, and a second fraction, boiling above about 1000° F. and containing ash solids recovered from the clarified extract. The recycle stream is added in amounts of from about 1 to about 50 weight percent of the feed to the separation zone. It has been found that each of these fractions quite unexpectedly acts as an agglomerating agent for particulate solids in the liquefaction product, increasing the sizes of the particles to separable sizes, thereby providing an increase in the clarity of the coal extract issuing from the separation zone.

BACKGROUND OF THE INVENTION

This invention relates to the production of liquid fuels from solid coal, and more particularly, to the step in such processes in which undissolved solids mixed with liquefied coal extract in a coal liquefaction product are separated from the liquefied extract according to the size of the solids.

In the upgrading of coal by hydrogen transfer and cracking to obtain liquid fuels such as gasoline and kerosene, a number of processes are carried out to transfer hydrogen to the basic coal molecules, to break up the coal molecules into smaller fragments, and to remove sulfur and nitrogen from the products. The basic coal liquefaction step may be carried out in a number of ways, but it is preferred to use a hydrogen-donor solvent, such as hydrogenated creosote oil or an indigenous hydrogenated product boiling from about 300° F. to about 900° F. which is obtained in the liquefaction of coal. Extraneous molecular hydrogen may be added to the liquefaction zone, if desired. All of this, directed to the basic liquefaction of coal, is old in the art, as disclosed in various patents such as U.S. Pats. 3,018,241 and 3,117,921.

The product of coal liquefaction is a mixture of liquefied coal extract (some of which is cracked and hydrogenated), solvent, and undissolved solids, including unconverted organic solids and ash solids. The solids are conventionally separated from the liquefied extract and solvent by centrifugation, filtration, or other solids-liquids separation processes in which solids are separated from liquids according to the size of the solids. Thereafter the clarified liquid is upgraded by various catalytic processes, including catalytic hydrocracking, in order to produce liquid fuel products which are recovered by fractional distillation.

Because the clarified extract from the solids-liquids separation zone is passed into a catalytic hydrocracker to upgrade the liquid extract, and because extremely small solids (for example, 10 microns and less) can block catalyst pores and eventually produce channeling in the bed, it is of great importance that these extremely small particles be removed from the liquid extract before hydrocracking of the extract. Heretofore, in U.S. Pat. 3,018,241, it was suggested that benzene insoluble solids can be separated from the extract by the addition of a low-boiling paraffinic solvent, specifically hexane. On a commercial scale, however, the suggestion is impratical. Yields of liquid extract are so reduced that the process is economically prohibitive. It is therefore highly desirable and plainly important that more economical and more usefully effective measures be discovered for improving the removal of solids from coal extracts produced in coal liquefaction processes. This invention meets that need.

SUMMARY OF THE INVENTION

This invention provides a new method in increasing the clarity of a liquefied coal extract stream issued from a separation zone which separates solids from the liquid extract of a coal liquefaction product fed into the zone, according to the sizes of the solids, and then discharges the solids in a separate concentrated slurry. The basic feature of the invention is the recycling and addition to the coal liquefaction product of certain clarified coal extract fractions or certain fractions of the concentrated solids slurry. The recycle fractions of clarified coal extract added to the liquefaction product may be either a fraction boiling within the range from about 100° F. to about 700° F. or a fraction boiling above about 1000° F. and containing ash solids.

The added recycled fraction of the concentrated solids slurry from the separation zone boils within the range from about 100° F. to about 700° F., and preferably is derived from distilled products produced by coking the concentrated solids slurry from the separation zone. The fractions of clarified coal extract and the fractions of the concentrated coal slurry which boil within the range from about 100° F. to about 700° F. may be separate or composite fractions. Quite surprisingly, it has been found that the addition of any of these particular fractions serves to agglomerate the very small particles in the liquefaction product and thereby to increase the size of these particles to a separable size, providing an increased clarity in the coal extract issued from the separation zone. The effectiveness of the clarified coal extract fraction which boils above 1000° F. and contains ash solids is especially remarkable in this regard, in that one would normally expect that the clarity of the coal extract would be poorer on addition of such a fraction, since the resultant feed to the separation zone then contains a greater percentage of very fine solids which must be processed there. In general, whichever fraction is used, suitable increases in coal extract clarity can be obtained by adding the recycle fraction to the liquefaction product in amounts of from about 1 to about 50 weight percent of the resulting feed to the separation zone.

As an aspect of the invention, it has been found that a desired clarity of the coal extract issuing from the separation zone in a continuous process can be maintained by adding to the liquefaction product more or less of a recycle fraction boiling within the range from about 100° F. to about 700° F., as needed, to keep the specific gravity of the clarified extract below a predetermined value which corresponds to the desired level of clarity.

Other aspects and advantages of the invention will be more evident from a description of preferred methods of carrying out the invention, taken in conjunction with the accompanying drawings.

DESCRIPTION OF THE DRAWINGS

FIG. 1 is a schematic flow diagram of preferred modes of carrying out the invention;

FIG. 2 is a plot of the weight percent of solids smaller than a given size versus the particle sizes of pyridine-washed solids and methyl ethyl ketone-washed solids of a liquefaction product prepared as described in Example 1, where the plot is discussed; and

FIG. 3 is a plot of the ash content and specific gravity of the overflow centrate from a centrifuge operated in accordance with invention over a 140-hour period, indicating the rate of addition of a recycle coker oil fraction derived from the centrifuge underflow. FIG. 3 is discussed in Example 2.

DESCRIPTION OF THE PREFERRED EMBODIMENT

Referring to FIG. 1, raw coal is fed by way of line **10** into a mixer **11** in which a slurry is created and withdrawn by way of line **12**. Any suitable coal-like material may be used, for example, subbituminous coal, bituminous coal, lignite or brown coal. The coal is generally ground to a particle size of about −8 mesh (Tyler screen) and finer and may be dried before it is fed into the mixer **11**. The solvent/coal weight ratio is suitably within the range from about 0.8 to about 10, and preferably from about 1 to about 2.

The slurry may be mixed with hydrogen introduced by way of line **13** and then passed into a liquefaction reactor **14**. Within the liquefaction reactor, the coal is allowed to dissolve under conditions of high temperature and pressure, such as a temperature within the range from about 650° F. to about 900° F. and a pressure from about 350 p.s.i.g. to about 2500 p.s.i.g. The hydrogen-treat rate (if hydrogen is used) may be fairly low, and may suitably range from about 100 to about 1000 s.c.f./b. of coal slurry charge. In the liquefaction reactor, the coal is depolymerized and partially thermally cracked, and a product is withdrawn, by way of line **15**, which comprises the liquefied coal extract, at least a partially depleted hydrogen-donor solvent, and undissolved solids, including ash solids. The hydrogen and noncondensable gases are separated from the liquid and solid components in a separator **16** and are removed by way of line **17**, while the slurry is carried by line **18** to a separation zone **19** which separates solids from liquids according to sizes, separately issuing a clarified coal extract and a concentrated solid slurry. Separation zone **19** is suitably a filter or, as illustrated, a centrifuge, in which case the clarified coal extract issues as a centrate overflow **20** and the concentrated solids slurry issues as an underflow **21**.

In accordance with this invention, a fraction of the clarified coal extract boiling within the range from about 100° F. to about 700° F. or above 1000° F. and containing ash solids, or a distillate fraction of products derived from coking the concentrated solids slurry from separation zone **19**, which distillate fraction boils within the range from about 100° F. to about 700° F., is added to the liquefaction product feeding to separation zone **19**. The added recycle fraction of clarified coal extract may be obtained from the clarified coal extract without subjecting the extract to further treatment after it issues from the separation zone. Thus, the clarified coal extract may be fractionated, as in a pressure reduction flash zone fractionation hereinafter described, and one of the aforesaid fractions of the clarified coal extract then recycled for addition to the liquefaction product. Alternatively, before fractionation, the clarified coal extract may be further treated to upgrade the extract. For example, the clarified coal extract may be hydrotreated to saturate the unsaturated mono and polynuclear rings of the extract to a desired level, or in a more severe treatment, the clarified coal extract may be hydrocracked to convert high-boiling aromatic compounds to lower boiling and saturated compounds. The use of a hydrocracking process is hereinafter described to illustrate this aspect of the invention.

In the drawing, reference numeral **22** indicates a recycle line of clarified coal extract boiling within the range from about 100° F. to about 700° F., reference numeral **23** indicates a recycle line of distillate fractions boiling within the 100° F. to 700° F. range and recovered from the concentrated solids slurry underflow, and reference numeral **24** indicates a recycle line of a clarified coal extract fraction boiling above about 1000° F. and containing ash solids. Whichever fraction is added, sufficient of the fraction is used to produce an increase in clarification of the clairfied overflow centrate issued from the centrifuge by line **20**. Suitably, the fraction added by line **22**, line **23** or line **24** is in amounts of from about 1 to about 50 weight percent, preferably from about 3 to about 30 weight percent of the resulting feed to the centrifuge. Relatively less of a lower boiling fraction than of a higher boiling fraction within the range from about 100° F. to about 700° F. or the fraction boiling above 1000° F. is needed to attain a desired increase in clarity. Additions of more than about 50 weight percent of a recycle fraction are impractical from a cost standpoint. When recycle fractions boiling within the 100° F.–700° F. range are added, the specific gravity of the clarified extract is monitored. A rise in specific gravity above a predetermined level corresponding to a desired centrate clarity is followed by addition of more of the recycle fraction, as needed, to reduce the specific gravity of the centrate to the predetermined level.

The clarified centrate overflow from centrifuge **19**, as stated above, may be fractionated without further treatment, or it may be subjected to further upgrading and then fractionated. Accordingly, in the latter instance, the overflow centrate from centrifuge **19** is carried by line **25** from line **20** with the opening of valve **26** and the closure of valve **27** into a pressure reduction flash zone **28**, which is operated in accordance with known technology to recover a fraction of the centrate overflow which boils within the range from about 100° F. to about 700° F. The particular fraction selected within that range is discharged from the flash zone by way of line **29** and passes through a metering valve **30** which admits only as much of that fraction to recycle line **22** as is desired, the remaining portion of that fraction being diverted into line **31** for recycle to line **20** by way of line **32**. Fractions of the centrate fed to the flash zone which are not designed for recycle, i.e., fractions boiling above about 700° F. but below about 1000° F., or in a particular instance, a heavier fraction within the 100°–700° F. boiling range aforesaid, is discharged from flash zone **28** by line **33** for return to line **20** by line **32**. Flash zone may be operated to separate the foregoing fractions from the fraction of the overflow centrate boiling above about 1000° F., which is illustrated as discharged from flash zone **28** by way of line **34**, which, when valve **35** is opened and valve **36** is closed, passes the bottoms fraction of the centrate by way of line **37** into the centrate bottoms recycle line **24**. When valve **35** is closed and valve **36** is opened, line **34** returns the centrate bottoms to line **20** for ungrading.

Line **20** carries the clarified centrate overflow into a hydrocracking zone, suitably comprising two reactors **38** and **39**, for upgrading. The clarified centrate in line **20** may comprise all of the centrate overflow from centrifuge **19** in the instance when valve **26** is closed and valve **27** is opened, or various fractions of the centrate overflow return to line **20** by lines **36** and/or **32** when valve **27** is closed and valve **26** is opened.

In the hydrocracking zone, the clarified extract is contacted with hydrogen introduced by way of lines **40** and **41** and is passed sequentially by way of line **42** in downflow across stationary beds of catalyst granules in the reactor, suitably cobalt molybdate, nickel molybdate, nickel tungsten and palladium on various substrates, such

as kielselguhr, alumina, silica, faujasites, etc. The cobalt molybdate catalyst is preferred, and may have 3.4 weight percent cobalt oxide, 12.8 weight percent molybdenum oxide, and 8.3 weight percent alumina. The catalyst may range from 2 to 5 weight percent cobalt oxide and from 10 to 15 weight percent molybdenum oxide, all are well known in hydrocracking arts. The clarified extract passed in downflow across the catalyst beds is preferably in the liquid phase, but may be in the mixed liquid and vapor phase, hydrogen in the reactor being present in both the gas phase and dissolved in the liquid phase. Preferably, the hydrocracking reaction carried out in the reactors **38** and **39** occurs under hydrocracking conditions which include a temperature from about 650° F. to about 900° F., preferably about 750° F., a pressure of 1000 to 4000 p.s.i.g., preferably 2000 p.s.i.g., a residence time within the reactor of 30 to 300 minutes, preferably 60 minutes, and a hydrogen rate from about 3000 to about 8000 s.c.f./b., preferably about 5000 s.c.f./b., based on the total volume of liquid charged to the hydrocracking reactors.

The products of the hydrocracking reactor are removed by way of line **43** and introduced into a fractionating tower **44**, where the clarified and hydrocracking products are fractionated into a plurality of various fuel products streams, including: gas taken overhead by way of line **45**; a stream boiling within the naphtha boiling range, from about 75° F.–100° F. up to about 400° F., a selected fraction of which is removed by way of line **46** for recycle by way of line **22**; a middle distillates stream boiling over the range from about 400° F. to about 700° F., a desired fraction of which is removed by way of line **47** for introduction to recycle line **22**; a heavy distillates stream boiling above about 700° F. and up to about 1000° F., which is removed by way of line **48** for use as desired; and a clarified centrate fraction boiling above about 1000° F. and containing ash solids, which is removed from distillation tower **44** by way of ine **49** for transfer to recycle line **24**. The fractions taken from the distillation tower **44** by way of lines **46** and **47** may be singularly introduced into line **22**, or blended, by the closure or metering operation of a suitable valve **50**. The bottoms fraction boiling above about 1000° F. taken from distillation tower **44** by line **49** is routed to line **20** by way of line **51** when valves **52** and **53** are closed and valve **54** is opened. The centrate bottoms fraction is routed to recycle line **24** by line **29** on closure of valves **53** and **54** and the opening of valve **52**. When valves **54** and **52** are closed and valve **53** is opened, the bottoms fraction of the centrate is carried by way of line **55** to a mixing zone **56** where it is mixed with the underflow carried by line **21** in the mixing zone **56**. Line **55** may be closed by valve **53** to prevent mixing of the underflow from line **21** and the bottoms fraction from distillation tower **44**, if desired.

The effluent from mixing zone **56** discharges by way of line **57** for introduction into coker **58**, which preferably is operated to maintain a dense phase fluidized bed of coke particles in the lower portion thereof. Within coker **58**, the liquid hydrocarbons in the underflow undergo thermal cracking, and vaporous hydrocarbon products are passed upwardly into a distillation tower suitably mounted above the coker vessel as schematically illustrated. The fractionator is operated to produce gas, naphtha, middle distillates and heavy distillates streams, as in the case of distillation tower **44**, the desired fractions within the naphtha and middle distillates streams being removed to recycle line **23** by way of lines **59** and **60** respectively. The use of single portions of such fractions or blends thereof being controlled by a valve **61**, as in the case of valve **50** for the fractions produced from distillation tower **44**. Although not illustrated, the vaporous product from coker **58** may be routed directly to distillation tower **44** and recovered therefrom so that the fraction recycled by line **22** is a blend of centrate overflow and underflow products.

The following examples will further illustrate the invention. In the examples, increase in liquid extract clarity is quantified by the decrease in ash content of solids in the clarified extract. In this regard, ash content of solids, or reference elsewhere herein to "ash solids," is not the same as the total mineral matter content of the solids, which also contain sulfur and carbonates, for example.

Example 1

Using a disc-nozzle type centrifuge operating at 400° F., solids (including ash solids were separated, in three runs, from samples of the liquid extract of a coal liquefaction product resulting from heating a slurry of two parts hydrogenated creosote oil and one part —100 mesh Illinois #6 coal at 730° F. under 350 p.s.i.g. for about 45 minutes. In the first run, the liquefaction product feed to the centrifuge was unmodified. In the second and third runs, the feed to the centrifuge was modified by inclusion of 10 weight percent (Rune 2) and 20 weight percent (Run 3) of a hydrocracked centrate fraction boiling in the naphtha distillation range, and produced as described above in connection with FIG. 1. Solids in the feeds and ash solids in the centrates of Runs 1–3 were measured. The results of these runs are set out below in Table I.

TABLE I.—ADDITION OF HYDROCRACKED CENTRATE FRACTION (HCF) TO LIQUEFACTION PRODUCT (L.P.)

	Run number		
	1	2	3
Feedstock	L.P.	L.P.[1]	L.P.[2]
Feed:			
Rate, lb./min.	30.7	27.0	27.1
Benzene insolubles, wt. percent[3]	20.94	21.23	20.95
MEK insolubles, wt. percent	6.14	6.83	7.65
Ash, wt. percent	3.28	3.34	3.49
Organic MEK insolubles, wt. percent[4]	2.86	3.49	4.16
Quasi-solids, wt. percent[5]	14.8	14.4	13.3
Overflow/underflow splits:			
Quasi-solids	2.97	4.58	3.24
Benzene solubles	3.02	3.88	3.24
Centrate:			
Rate, lb/min	21.8	20.0	19.6
Specific gravity	1.0901	1.0652	1.0540
Viscosity at 400° F., cp.	3.7	3.0	2.9
Ash in centrate, wt. percent[3]	0.41	0.16	0.10
Ash removed from centrate, percent	87.6	94.7	96.6
Ash balance, percent	103	101	92
Improvement in clarity, percent		61	75.5

[1] Plus 10% HCF.
[2] Plus 20% HCF.
[3] Weight percents are based on L.P. in feed.
[4] Organic MEK insolubles are MEK insolubles less ash.
[5] Quasi-solids are benzene insolubles less MEK insolubles.

As may be seen by reference to Table I, in Run 1, the ash content of the centrifuge feed was reduced from 3.28 to 0.41 percent in the centrate, a reduction of 87.6 percent of the ash in the feed. In Run 2, in which 10 weight percent of hydrogenated centrate fraction was added to the feed, ash content was reduced from 3.34 weight percent to 0.16 weight percent in the centrate, an improvement in centrate clarity of 61 percent over that obtained in Run 1. In Run 3, in which 20 weight percent of hydrogenated centrate fraction was added to the centrifuge feed, ash content was reduced from 3.49 weight percent in the feed to 0.10 weight percent in the centrate, an improvement in centrate clarity of 75.5 percent over Run 1.

The content of solids in the feed to the centrifuge was determined by using benzene and methyl ethyl ketone (MEK), at room temperature, as solvents to dilute samples of the liquefaction product, at least in equal volumes, and to wash the solids which separated in a laboratory analytical centrifuge. The washed solids were pyrolyzed to determine ash contents. As set forth in Table I, the benzene insoluble solids comprised solids soluble and insoluble in MEK. The solids which were insoluble in MEK had both ash constituents and organic constituents.

The following experiment was undertaken to ascertain the nature of the organic constituents. A slurry of two parts by weight by hydrogenated creosote oil to one part by weight of −100 mesh Illinois #6 coal was liquefied at 730° F. and 350 p.s.i.g. for about 45 minutes. The liquefaction product was diluted with an equal volume of methyl ethyl ketone, the solids in the diluted liquefaction product were separated in a laboratory analytical centrifuge, and the solids recovered from the centrifuge were washed with MEK. Pyridine was then used as a solvent to dilute samples of the MEK washed solids and to wash the solids which separated by filtration through Whatman No. 42 fine filter paper. Coulter counter measurements were made on samples of the MEK washed solids and on samples of the pyridine-washed solids. Solids distribution curves of the weight percent of solids less than a given size were plotted against particle sizes for the pyridine-washed solids and for the MEK washed solids. These curves are illustrated in FIG. 2. The curves show that 50 weight percent of the pyridine-washed solids were less than 6 microns in diameter and 50 weight percent of the MEK washed solids were less than 12 microns in diameter. The pyridine-washed solids appear to closely approximate the "true" solids content of the solids in the liquefaction extract, i.e., pyridine-washed solids are composed of the mineral matter and unconvertible organics in the liquefaction extract. The MEK washed solids, which are larger, are apparently composed of the "true" solids plus solid substances which had been dissolved in the liquid extract but solidified on addition of the MEK to the liquefaction extract to agglomerate the "true" solids into the larger particles. Thus, the organic constituents in the MEK insolubles in Run 1 apparently contain a slight amount of converted organic material. However, because identical solvent separation techniques were used in Runs 1–3, valid comparisons can be made between Run 1 and Runs 2 and 3.

Referring to Table I, the solids which were insoluble in benzene but soluble in methyl ethyl ketone are termed "quasi-solids." Benzene solubles are, of course, liquids. The distribution in the centrifuge of liquids of the density of benzene solubles is given by the ratio of benzene solubles appearing in the overflow to those appearing in the underflow. As Runs 1 and 3 indicate, the ratio of quasi-solids in the centrate to quasi-solids in the underflow is essentially the same as the overflow/underflow ratio for benzene solubles. (Run 2 had poorer balances of benzene and MEK insolubles not shown in Table I and is not illustrative in this regard.) Hence, the quasi-solids can be taken to be dissolved in the liquid extract solvent at the 400° F. temperature.

As Runs 1 and 3 of Table I show, the percent of organic MEK insoluble solids in the feed increased about 1.3 weight percent on addition of the hydrocracked centrate fraction, the percent of ash staying fairly consistent. On the other hand, the weight percent of quasi-solids in the feed decreased by about the same amount. This suggests that the addition of the hydrocracked centrate fraction to the liquefaction product was instrumental in resolidifying slightly more than 1 weight percent of the liquid quasi-solids into organic MEK insoluble solids. Taken with the fact that the clarity of the centrate recovered from the centrifuge increased 75.5 percent on addition of the hydrocracked centrate fraction, the evidence suggests that the resolidification of the quasi-solids was instrumental in increasing the centrate clarity.

Generally speaking, the solvation power of the liquid extract of the liquefaction product at 400° F. is approximately the same as the solvation power of pyridine at room temperature, and the solvation power of the liquid extract plus the hydrocracked centrate fraction at 400° F. is approximately the same as that of MEK at room temperature. Accordingly, the solids distribution curves for pyridine insoluble solids and MEK insoluble solids in FIG. 2 may be taken as representative, respectively, of the sizes of solids in the liquefaction product in Run 1 and solids in the liquefaction product containing the hydrocracked centrate fraction in Runs 2 and 3. This indicates that the improved clarity which accompanied the indicated resolidification of quasi-solids in Runs 2 and 3 occurs, apparently, because the resolidification increases the less than 10 micron particle sizes of at least some of the already solid particles in the liquid extract to more than 10 microns, shifting these particles into a separable size range. The marked nature of the clarity improvement suggests that the addition of the hydrocracked centrate fractions in Runs 2 and 3 served to agglomerate the smaller than 10 micron particles in the liquefaction product fed to the centrifuge.

Undoubtedly, in addition to the foregoing, the reduction in specific gravity and viscosity evidenced by Table I also contributed to the increased resolution in the separation process. The viscosity figures, which were obtained in a Brookfield viscometer, cannot be regarded as quantitatively accurate at the 400° F. operating temperature. However, they can be taken as qualitatively indicative of the relative reduction in viscosity occurring with the addition of the hydrogenated centrate fraction. Example 2, which is set out below, sheds further light on the relationship between specific gravity, ash reduction, and addition of a hydrogenated centrate fraction.

Example 2

In a continuous seven-day run, coal was liquefied and centrifuged in a solid bowl scroll discharge centrifuge operating at about 400° F., essentially under atmospheric pressure, while a coker oil fraction derived from a centrifuge underflow (as described in connection with FIG. 1) and boiling between about 400° F. and about 700° F. was added to the centrifuge feed. In the liquefaction reactor, the conditions which produced the feed to the centrifuge included a temperature of 770° F., a pressure of 350 p.s.i.g., a 7:1 solvent/coal weight ratio of −100 mesh Illinois #6 coal in hydrogenated creosote oil boiling from about 300° F. to over 1000° F., a coal feed rate to the liquefaction reactor of 8 lbs./hr. and a 1 hour residence time. Except as indicated in FIG. 2, the feed rate of centrate coker oil fraction was about 2 lbs./hr., an addition of about 3 weight percent of centrate fraction to the centrifuge feed. The centrate rate was about 65 lbs./hr. The results of this seven-day run are displayed in FIG. 3.

FIG. 3 indicates that a monitoring of the specific gravity of the centrate provides a good measure of the how much more, or less, of centrate fraction needs to be added to the liquefaction product introduced to a centrifuge separation zone, in a continuous operation, in order to get and keep a desired increase in centrate clarity (reduction in ash content) as the makeup of liquefaction product varies. By controlling the addition of coker oil centrate fraction to keep the specific gravity of the centrate, corrected to 60° F., below about 1.085 in this example, it was possible to maintain the ash content below a maximum level of about 0.3 weight percent, and, at steady state conditions, below about 0.10 weight percent, as occurred after about 60 hours of operation. As illustrated by FIG. 4, at about 90 hours the specific gravity of the centrate, corrected to 60° F., was permitted to creep over the 1.085 mark selected for the separation control in the processing of the particular slurry of this example. Addition of sufficient centrate fraction to push the specific gravity back to or below the selected mark was not made, and as illustrated, the maintenance of a uniform centrate fraction feed rate without regard to the rising centrate specific gravity permitted the ash content of the centrate to rise.

Because of the relatively higher specific gravity of the clarified liquid extract bottoms fraction boiling above about 1000° F. and containing ash solids, the foregoing use of specific gravity measurements is not believed applicable to the bottoms fraction. The bottoms fraction with the ash solids contaminant is surprisingly effective,

however, in clarifying the liquid extract, as shown by Example 3.

Example 3

A solid bowl scroll discharge centrifuge was used to clarify the coal extract of a liquefaction product at a temperature of about 400° F. The ash content in the clarified coal extract was reduced from approximately 3 weight percent to about 0.15 weight percent. As illustrated in FIG. 1, the clarified centrate was then hydrocracked and fractionated. Upon commencing a recycle to the centrifuge feed of fractionator bottoms boiling above about 1000° F. and containing fine solids which had already passed through the centrifuge, approximately 25 pounds of recycle fraction being added to 81 pounds of the normal feed (about 31 weight percent), the clarity of the centrate did not deteriorate, as one would normally expect. Instead, the ash content of the centrate surprisingly decreased to approximately 0.03 weight percent.

Having now fully disclosed with particularity preferred modes by which our invention may be carried out various modifications and alterations within the spirit and scope of our invention, as claimed, will occur to those in the art.

We claim:

1. In the process of clarifying a coal liquefaction product comprised of coal extract liquids and undissolved solids, wherein the liquefied product is introduced into a solids-liquid separation zone which separates solids from liquid according to sizes of the solids and discharges as one issue a clarified coal extract and as another issue a concentrated solids slurry, the improvement which comprises:

fractionating at least one of said issues from said separation zone to recover a first recycle stream boiling within the range from about 100° F. to about 700° F.,

fractionating said clarified coal extract to recover a second recycle stream boiling above about 1000° F. and containing ash solids, and

adding sufficient of a recycle stream selected from said first and second recycle streams to said liquefaction product effective to cause at least a portion of the solids in the liquefaction product to agglomerate, whereby the clarity of the clarified coal extract issued from the separation zone is increased.

2. In the process of clarifying a coal liquefaction product comprised of coal extract liquids and undissolved solids, wherein the liquefied product is fed into a solids-liquid separation zone which separates solids from liquid according to sizes of the solids and separately issues a clarified coal extract and a concentrated solids slurry therefrom, the improvement which comprises:

passing the concentrated solids slurry into a coking zone operated to produce distillable products therefrom,

distilling the products from the coking zone to recover a first stream of a coker oil fraction boiling within the range from about 100° F. to about 700° F.,

fractionating the clarified coal extract to recover therefrom at least one of a second stream boiling within the range from about 100° F. to about 700° F. and a third stream boiling above about 1000° F. which contains ash solids, and

adding sufficient of a recycle stream selected from said first stream, said second stream, a mixture of said first and second streams, and said third stream, to said liquefaction product effectively to cause at least a portion of the solids in the liquefaction product to agglomerate, whereby the clarity of the coal extract issued from the separation zone is increased.

3. The process of claim 2 in which said recycle stream is added in amounts of from about 1 to about 50 weight percent of the resultant feed to the solids-liquid separation zone.

4. The process of claim 2 in which said first stream, said second stream or a mixture thereof is added to said liquefaction product as necessary to keep the specific gravity of the clarified extract below a predetermined value within the range from about 1.08 to about 1.12.

5. A method of producing liquid fuels from a coal liquefaction product of coal extract and undissolved solids, which comprises:

feeding said coal liquefaction product into a centifuge at a temperature between about 300° F. and about 550° F.,

receiving the clarified overflow and the concentrated solids underflow issuing from the centrifuge,

passing the concentrated solids underflow into a coking zone operated to produce distillable products therefrom,

distilling the products from the coking zone and the clarified overflow to recover a recycle stream selected from a fraction boiling within the range from about 300° F. to about 700° F. and a fraction boiling above about 1000° F. which contains ash solids, and

adding said recycle stream to said liquefaction product in amounts from about 1 to about 50 weight percent of the resulting feed to the centrifuge, effective to cause the clarity of the concentrate issued from the centrifuge to increase.

6. The method of claim 5 in which said recycle stream boiling within the range from about 300° F. to about 700° F. is added to said liquefaction product as needed to keep the specific gravity of the clarified overflow below a predetermined value within the range from about 1.08 to about 1.12.

7. The method of claim 5 in which said clarified overflow is hydrocracked before distillation.

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