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(54) Process for the hydrocracking of a hydrocarbonaceous feedstock.

(57) A hydrocarbonaceous feedstock (8) is hydrocracked in a plurality of reaction stages by contacting the feedstock with a first hydrocracking catalyst (2) at elevated temperature and pressure in the presence of hydrogen (9) in a first reaction stage to yield a first effluent (10), mixing at least a liquid portion of the first effluent with a second effluent that originates from a second reaction stage (4), contacting the mixture obtained in a third reaction stage with a third hydrocracking catalyst (5) at elevated temperature and pressure in the presence of hydrogen to yield a third effluent (12), passing the third effluent to a separation stage where at least one top fraction and a residual fraction (19) are obtained and passing the residual fraction to the second reaction stage where it is contacted with a second hydrocracking catalyst at elevated temperature and pressure in the presence of hydrogen (20) to yield the second effluent.

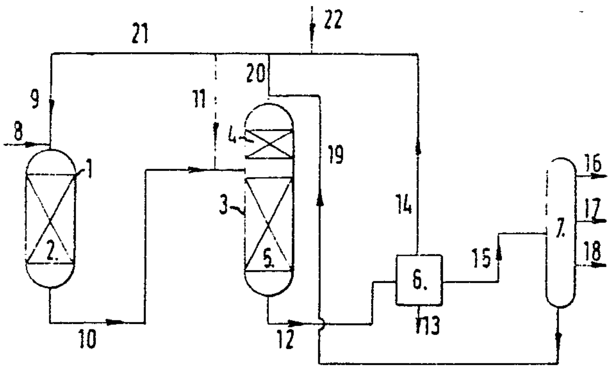

FIG.1

PROCESS FOR THE HYDROCRACKING OF A HYDROCARBONACEOUS FEEDSTOCK

The present invention relates to a process for the hydrocracking of a hydrocarbonaceous feedstock in which a plurality of reaction stages are employed.

Hydrocracking is a well-established process in which heavy hydrocarbons are contacted in the presence of hydrogen with a hydrocracking catalyst. The temperature and the pressure are relatively high, so that the heavy hydrocarbons are cracked to products with a lower boiling point. Although the process can be carried out in one stage, it has been shown to be advantageous to carry out the process in a plurality of stages. In a first stage the feedstock is subjected to denitrogenation, desulphurisation and some hydrocracking, and in a second stage most of the hydrocracking reactions occur.

The presence of nitrogen compounds in the feedstock may lead to problems as, generally, amorphous hydrocracking catalyst tend to be nitrogen-sensitive and may be deactivated by the nitrogen compounds. A solution to this problem was considered to be the use of a hydrocracking catalyst with hydrodenitrogenating activity in a first reaction stage and the use of a zeolitic hydrocracking catalyst which is less nitrogen-sensitive, in a second reaction stage. However, these zeolitic catalysts incurred the drawback that bulky molecules containing several ring structures could not be adequately cracked unless very high temperatures were employed.

U.S. Patent No. 3,702,818 describes one proposed solution to this problem in which two second stage reactors are employed, one containing a zeolite catalyst and the other an amorphous base catalyst. While bulky ring structures will be cracked by the amorphous catalyst, the recycle stream will still contain a substantial quantity of medium heavy products which will result in unduly high catalyst loading.

The present invention provides a solution to the problem of nitrogen-sensitivity and inadequate hydrocracking of bulky materials.

Accordingly, the present invention provides a process for the hydrocracking of a hydrocarbonaceous feedstock in a plurality of reaction stages, comprising contacting the feedstock with a first hydrocracking catalyst at elevated temperature and pressure in the presence of hydrogen in a first reaction stage to yield a first effluent, mixing at least a liquid portion of the first effluent with a second effluent that originates from a second reaction stage, contacting the mixture obtained in a third reaction stage with a third hydrocracking catalyst at elevated temperature and pressure in the presence of hydrogen to yield a third effluent, passing the third effluent to a separation stage where at least one top fraction and a residual fraction are obtained and passing the residual fraction to the second reaction stage where it is contacted with a second hydrocracking catalyst at elevated temperature and pressure in the presence of hydrogen to yield the second effluent.

When the first hydrocracking catalyst has a denitrogenating activity, when the third catalyst is relatively nitrogen-insensitive and the second hydrocracking catalyst is capable of hydrocracking bulky molecules, the above-mentioned problems are obviated.

The present invention can be embodied in various embodiments, which are variations of a series-flow hydrocracker and a two-stage hydrocracker. For the flow scheme of these known processes reference is made to "The Petroleum Handbook", 6th ed., Elsevier, Amsterdam, 1983, pp. 294-300.

A first embodiment is based on a series-flow hydrocracker. In a conventional series-flow hydrocracker, the effluent from a first reaction stage is passed to a second one, and the effluent from the second reaction stage is subjected to a work-up procedure including fractionation. The fractionation yields a residual fraction which is combined with the first effluent to be passed to the second reaction stage. The third stage according to the invention corresponds with the conventional second stage.

According to the present invention the combination of at least part of the first effluent and the second effluent is passed to the third reaction stage. The third effluent emanating from the third reaction stage is subjected to the work-up procedure in which the product distillation fractions are recovered and a residual fraction. The residual fraction from the fractionation is recycled to the second stage to yield the second effluent. It is possible to subject the first effluent to a gas purification step to remove ammonia and/or hydrogen sulphide that have formed in the first reaction stage, by treatment with e.g. an aqueous solution of e.g. a mineral acid. However, it is convenient to combine the complete first effluent with the second effluent. This has the advantage that little needs to be changed in the conventional series-flow process. In another preferred embodiment the first effluent is subjected to a separation in a liquid and a gaseous phase. The gaseous phase is combined with the third effluent and subjected to the work-up procedure. This has the advantage that the gaseous phase will contain most of the ammonia, hydrogen sulphide and also cracked products.

Hence there is no risk of over-cracking the desired products to undesired C_{1-4} hydrocarbon products. The liquid phase which contains only a minute proportion of the the ammonia in the first effluent, is combined with the second effluent to be hydrocracked in the third stage. It is, of course, possible to separate the first effluent at different temperature and pressure conditions. Thus one can cool the effluent to a temperature lower than the temperature prevailing in the first reaction stage. However, it is preferred to carry out this separation at substantially the same temperature and pressure as prevailing in the first reaction stage. This is accomplished advantageously by refraining from heating or cooling of the first effluent. In an even more preferred embodiment, this separation is carried out in the same reaction vessel as that in which the first reaction stage is carried out. This can be achieved by the creation of a settling area in the bottom part of the reaction vessel, from which the gaseous phase and the liquid phase are withdrawn via different openings. It will be appreciated that minor fluctuations in the first reaction stage may occur. Then it is preferred to carry out the separation at substantially the same temperature and pressure as prevailing in the exit part of the first stage.

Another embodiment of the present invention is derived from a conventional two-stage hydrocracker. In a conventional hydrocracker the effluent from a first reaction stage is combined with the effluent from a second reaction stage, and the combined effluents are subjected to a work-up procedure including fractionation which yields the desired distillate products and a residual fraction that is to be passed to the second reaction stage.

According to this above-mentioned embodiment of the present invention the residual fraction of the fractionation is mixed with the effluent of the second stage and the mixture is passed to the third reaction stage. The effluent of this third stage is separated into a gaseous top fraction and a liquid bottom fraction. Preferably, the top fraction obtained after the separation of the third effluent is combined with the first effluent and the combination is subjected to a work-up procedure from which the products are recovered by fractionation. The liquid bottom fraction from the third effluent is passed to the second reaction stage.

Accordingly, in this embodiment one or more distillate fractions are obtained from the first effluent by fractionation and recovered as products and further a residual liquid portion is obtained that is mixed with the second effluent. The top fraction obtained in the separation stage may be processed separately. Preferably, in this embodiment the top fraction obtained in the separation stage is combined with the first effluent to be fractionated.

Advantageously the separation of the third effluent is accomplished in the way as described above for the first effluent in the scheme based on a series-flow hydrocracking process. Hence, although it is possible to separate the third effluent at different temperature and pressure conditions, e.g. by cooling the effluent and to a temperature lower than the temperature prevailing in the third reaction stage, it is preferred to carry out this separation at substantially the same temperature and pressure as prevailing in(the exit part of) the third reaction stage. This is accomplished advantageously by refraining from heating or cooling of the third effluent. In an even more preferred embodiment this separation is carried out in the same reaction vessel as in which the third reaction stage is carried out. This can be achieved by the creation of a settling area in the bottom part of the reaction vessel, from which the gaseous phase and the liquid phase are withdrawn via different openings.

The second reaction stage and the third reaction stage may be carried out in different reactors. It is, however, preferred to carry out both stages in one reaction vessel. Usually that does not involve too many technically difficult adjustments, since most conventional hydrocrackers comprise a plurality of catalysts beds, and by some alterations of feed conduits and a different loading of the catalyst beds conventional hydrocrackers can be accommodated to the process of the present invention.

As described hereinabove the first hydrocracking catalyst is preferably a catalyst with hydrodenitrogenating and/or hydrodesulphurising activity. Suitable catalysts include first hydrocracking catalysts comprising at least one component of a group 8 and/or 6b metal of the Periodic Table of the Elements on an amorphous carrier. Advantageously, the carrier is selected from the group consisting of silica, alumina, thoria, titania, magnesia, zirconia and mixtures thereof. The carrier more preferably comprises alumina. Reference is made to the Periodic Table of the Elements as published in the "Handbook of Chemistry and Physics", 55th edition (1975), C R C Press, Ohio, USA.

In the second and third stage, the same or different hydrocracking catalysts may be used. Mixtures or combinations of different catalysts can be used in each stage: such mixtures or combinations may be the same or different for each stage. Preferably, however, the catalyst, catalyst mixture or catalyst combination used in the second stage is different from that used in the third stage.

The second hydrocracking catalyst is preferably a catalyst with a good performance for the cracking of bulky molecules containing a plurality of rings. A very good catalyst in this respect is a second hydrocracking catalyst comprising at least one component of a group 8 and/or group 6b metal on a silica-alumina-containing carrier. This catalyst tends to be very nitrogen-sensitive. Since the hydrocarbonaceous feed to be cracked by this silica-alumina catalyst has already twice been subjected to a hydrocracking process by which also nitrogen is removed, the drawbacks of the use of a nitrogen-sensitive catalyst do not occur here. It is emphasised that also other catalysts may be used in the second stage, e.g. catalysts based on alumina, silica, titania, magnesia, zirconia, or mixtures thereof. The use of zeolitic catalysts in the second stage is also possible, but less preferred since, though the catalysts are very active, their activity towards the conversion of bulky molecules leaves room for improvement.

The third hydrocracking catalyst is preferably a zeolitic catalyst, i.e. a catalyst comprising at least one component of a group 8 and/or group 6b metal on a zeolite-containing carrier. The zeolite in the third hydrocracking catalyst is preferably a faujasite-type zeolite, and in particular zeolite Y. The silica/alumina molar ratio in such zeolites may vary from 4 to 25, in particular 6 to 15. The unit cell size of zeolite Y may vary from 2.419 to 2.475 nm, in particular from 2.425 to 2.460 nm. Apart from the zeolite, the third hydrocracking catalyst conveniently comprises further at least one binder. The binder is suitably selected from silica, alumina, silica-alumina, thoria, titania, zirconia, magnesia and mixtures thereof. In particular alumina is preferred, optionally in combination with silica-alumina.

For all three hydrocracking catalysts the catalytically active metal from groups 8 and 6b are preferably selected from cobalt, nickel, platinum, palladium, tungsten and molybdenum. Preferably non-noble metal mixtures are used, such as Ni-W, Ni-Mo, Co-Mo and Co-W. The catalysts may further contain phosphorus and/or fluorine to boost their activity. When the catalytically active metals are non-noble they are preferably present on the hydrocracking catalysts in an amount of 1 to 16 %wt of a group 8 metal and/or from 6 to 24 %wt of a group 6 metal, the weight percentages being based on the total catalyst. Noble metals are suitably present in a lower amount, e.g. from 0.2 to 2 %wt. Noble metals can especially be used on the third and/or second hydrocracking catalyst. Especially when the catalytically active metals are non-noble they are preferably present in their oxidic or even more preferred in their sulphidic form. The preparation of the hydrocracking catalysts is known in the art.

Hydrocarbonaceous feedstocks that can be used in the present process include gas oils, vacuum gas oils, deasphalted oils, long residues, short residues, catalytically cracked cycle oils thermally cracked gas oils and syncrudes, optionally originating from tar sands, shale oils, residue upgrading processes or biomass. Combinations of various hydrocarbonaceous feedstock can also be employed. The hydrocarbonaceous feedstock will generally be such that a major part, say over 50 %wt, has a boiling point above 370 °C. The present process is most advantageous when the feedstock contains nitrogen. Typical nitrogen contents are in the range of up to 5000 ppmw. Nitrogen contents may start from 50 ppmw. The feedstock will generally also comprise sulphur compounds. The sulphur content will usually be in the range from 0.2 to 6 %wt.

As to the process conditions in reaction stages, the temperature in any reaction stage is preferably from 300 to 450 °C, in particular from 350 to 420 °C, the pressure from 50 to 250 bar, in particular from 75 to 150 bar, the space velocity from 0.1 to 10 kg/l/h, in particular from 0.2 to 5 kg/l/h, and the hydrogen/oil ratio from 500 to 5000 Nl/kg, in particular from 1000 to 2500 Nl/kg.

The process will be further illustrated by means of the following Figures. For the sake of simplicity various types of equipment that is not essential to the invention are not shown. The Figures show only one catalyst bed in any reaction stage. It will be clear that instead of one catalyst bed a plurality of beds may be used.

Figure 1 shows an embodiment based on a series-flow hydrocracker in which the second and third reaction stage are carried out in a single reaction vessel.

Figure 2 shows an embodiment based on a series-flow hydrocracker in which after the first stage the first effluent is subjected to a gas-liquid separation.

Figure 3 shows an embodiment based on a two-stage hydrocracker, in which the third effluent is subjected to a gas-liquid separation which is carried out in the same reaction vessel as in which the second and third reaction stage are carried out.

In Figure 1 a reaction vessel 1 is shown, containing a catalyst bed 2. Catalyst bed 2 comprises the first hydrocracking catalyst. The figure further shows a second reaction vessel comprising a catalyst bed 4 with the second hydrocracking catalyst and a catalyst bed 5 with the third hydrocracking catalyst. Further, a gas purification section 6 is shown and a fractionator 7. During operation a hydrocarbonaceous feedstock is passed to the reaction vessel 1 via a line 8, together with a hydrogen containing gas supplied via a line 9. After having passed through the catalyst bed the mixture of hydrocarbons and hydrogen is withdrawn from the reaction vessel 1 via a line 10. To the mixture additional hydrogen is added via a line 11 and the resulting mixture is fed into the reaction vessel 3 at a position between the catalyst beds 4 and 5. The mixture then combines with an effluent emanating from catalyst bed 4, i.e. the second effluent, and together the mixtures are passed over the catalyst bed 5, yielding the third effluent that is withdrawn via a line 12. The effluent in the line 12 is passed to the gas purification section where it is freed from ammonia and/or hydrogen sulphide that is removed from the system via a line 13. The operation of the purification section is known in the art. It may make use of several gas-liquid separators at different temperatures and/or pressures to separate a hydrogen-containing gas from the desired products and of a wash section to remove ammonia and/or hydrogen sulphide. The hydrogen-containing gas (recycle hydrogen) may contain some light (C_{1-2}) hydrocarbons and/or hydrogen sulphide. Hydrogen that is recovered in the section 6 is recycled to the system via a line 14, and hydrocarbon products are passed to the fractionator 7 via a line 15. There they are separated to yield product streams 16, 17 and 18, and a residual fraction withdrawn from the fractionator 7 via a line 19. The residual fraction is recycled to the reaction vessel 3 and introduced into the vessel 3, after having been mixed with hydrogen coming from a line 20, at a position above the catalyst bed 4. The residual fraction is hydrocracked in the catalyst bed 4 to yield the second effluent. The hydrogen for the lines 9, 11 and 20 originates from a feeding line 21, that is provided with hydrogen by the recycle hydrogen in the line 14 and make-up hydrogen supplied via a line 22.

In Figure 2 a reaction vessel 31 is shown comprising a catalyst bed 32 in which the first reaction stage is carried out. It further shows a reaction vessel 33 with a catalyst bed 34 for the third reaction stage, and a reaction vessel 35 with a catalyst bed 36 for the second reaction stage. In addition the figure shows a gas purification section 37, a fractionator 38 and a separator 39. In operation a hydrocarbonaceous feedstock from a line 40 is mixed with a hydrogen-containing gas from a line 41 and together passed into the reaction vessel 31, where the mixture is hydrocracked over the catalyst bed 32 comprising the first hydrocracking catalyst. The first effluent is withdrawn from the reaction vessel 31 via a line 42 and passed to the separator 39. The separator 39 can be any device known in the art to accomplish a gas-liquid separation. It is preferably at the same temperature and pressure as reaction vessel 31. From the separator 39 a gaseous phase is withdrawn via a line 43 and a liquid phase via a line 44. The liquid phase is mixed with a second effluent in a line 45 coming from the reaction vessel 15, and the mixture is passed to the reaction vessel 33 via a line 46 to which hydrogen is added via a line 47. In the reaction vessel 33 the mixture from the line 46 is hydrocracked over the third hydrocracking catalyst in catalyst bed 34 to yield the third effluent withdrawn from the reaction vessel 33 via a line 48. To the third effluent the gaseous phase of the first effluent in line 43 is added, and the combined streams are fed into the gas purification section 37 to yield a waste stream of ammonia and/or hydrogen sulphide discharged via a line 49, recycle hydrogen discharged via a line 50 and the hydrocarbon product, discharged via a line 51. The line 51 debouches into the fractionator 38 where product streams 52, 53 and 54 are recovered. Distillation in the fractionator 38 further yields a residual fraction that is passed via a line 55 to the reaction vessel 35. In the presence of hydrogen coming from a line 56 the residual fraction is hydrocracked over the second hydrocracking catalyst in catalyst bed 36. The second effluent resulting from the hydrocracking over the catalyst bed 36 is withdrawn via the line 45 and mixed with the liquid phase in the line 44. Hydrogen in the process is suppied via a line 57 that is fed by make-up hydrogen from a line 58 and recycle hydrogen from the line 50.

It will be appreciated that the separator 39 can be incorporated in the reaction vessel 31 as a settling area. That would save the cost of a separate vessel. It is also possible to provide catalyst beds 36 and 34 in one reaction vessel as has been shown in Figure 1 for catalyst beds 4 and 5.

Figure 3 shows a reaction vessel 61 with a catalyst bed 62 comprising the first hydrocracking catalyst, a gas purification section 63, a fractionator 64, and a second reaction vessel 65 comprising a catalyst bed 66 for the second reaction stage and a catalyst bed 67 for the third reaction stage. The reaction vessel 65 further comprises a settling area 68 where a gas-liquid separation takes place. In operation a hydrocarbonaceous feedstock in a line 69 is combined with hydrogen from a line 70, introduced into the reaction vessel 61 and hydrocracked over the first hydrocracking catalyst in catalyst bed 62. The resulting first effluent is withdrawn from the reaction vessel 61 via a line 71. It is mixed with a gaseous top fraction in a line 72, and the mixture is fed into the purification section 63 where ammonia and/or hydrogen sulphide are withdrawn from the system via a line 74, a hydrogen-containing gas is recycled to the system via a line 75 and hydrocarbon products are separated and passed to the fractionator 64 via a line 76. In the fractionator 64 product streams 77, 78 and 79 are recovered and a residual fraction is withdrawn via a line 80. Hydrogen from a line 81 is added to the residual fraction and the mixture is fed into the reaction vessel 65 at a position between the catalyst beds 66 and 67. Between the beds 66 and 67 the residual fraction of the line 80 mixes with the effluent coming from the catalyst bed 66 and together they are passed over the catalyst bed 67 to yield a third effluent. The third effluent comes into the settling are 68, where a gas-liquid separation occurs. The gaseous top fraction is withdrawn via the line 72 and mixed with the first effluent, and the liquid residual portion is withdrawn from the reaction vessel 65 via a line 82, combined with hydrogen from a line 83 and recycled to the reaction vessel 65, but introduced at a position above the catalyst bed 66. The liquid residual portion is hydrocracked over the second hydrocracking catalyst in catalyst bed 66 to yield the second effluent. The hydrogen for the process is supplied via a line 84 that is fed by make-up hydrogen from a line 85 and the recycle hydrogen in the line 75.

Claims

1. Process for the hydrocracking of a hydrocarbonaceous feedstock in a plurality of reaction stages, comprising contacting the feedstock with a first hydrocracking catalyst at elevated temperature and pressure in the presence of hydrogen in a first reaction stage to yield a first effluent, mixing at least a liquid portion of the first effluent with a second effluent that originates from a second reaction stage, contacting the mixture obtained in a third reaction stage with a third hydrocracking catalyst at elevated temperature and pressure in the presence of hydrogen to yield a third effluent, passing the third effluent to a separation stage where at least one top fraction and a residual fraction are obtained and passing the residual fraction to the second reaction stage where it is contacted with a second hydrocracking catalyst at elevated temperature and pressure in the presence of hydrogen to yield the second effluent.

2. Process according to claim 1, in which the complete first effluent is mixed with the second effluent.

3. Process according to claim 1 or 2, in which the first effluent is subjected to a separation in a liquid and a gaseous phase, the gaseous phase is combined with the third effluent and the liquid phase is combined with the second effluent to be passed to the third reaction stage.

4. Process according to claim 3, in which the first effluent is separated into a liquid and a gaseous phase at substantially the same temperature and pressure as prevailing in the first reaction stage.

5. Process according to any one of claims 1-4, in which the top fraction(s) obtained in the separation stage is(are) recovered as products.

6. Process according to claim 1, in which from the first effluent one or more distillate fractions are obtained by fractionation and recovered as products and further a residual liquid portion is obtained that is mixed with the second effluent.

7. Process according to claim 6, in which the top fraction obtained in the separation stage is combined with the first effluent to be fractionated.

8. Process according to any one of claims 1-7, in which the first hydrocracking catalyst comprises at least one component of a group 8 and/or 6b metal on an amorphous carrier.

9. Process according to claim 8, in which the carrier is selected from the groups consisting of silica, alumina, thoria, titania, magnesia, zirconia and mixtures thereof.

10. Process according to claim 9, in which the carrier comprises alumina.

11. Process according to any one of claims 1-10, in which the second hydrocracking catalyst comprising at least one component of a group 8 and/or group 6b metal on a silica-alumina-containing carrier.

12. Process according to any one of claims 1-11, in which the third hydrocracking catalyst comprises at least one component of a group 8 and/or group 6b metal on a zeolite-containing carrier.

13. Process according to claim 2, in which the zeolite is a faujasite type zeolite, in particular zeolite Y.

14. Process according to any one of claims 1-13, in which the first, second and third hydrocracking catalyst comprise at least one hydrogenating component of a metal selected from the group consisting of nickel, cobalt, platinum, palladium, tungsten and molybdenum.

15. Process according to any one of claims 1-14, in which the temperature in one or more of the reaction stages is from 300 to 450 °C, the pressure from 50 to 250 bar, the space velocity from 0.1 to 10 kg/l/h, and the hydrogen/oil ratio from 500 to 5000 Nl/kg.

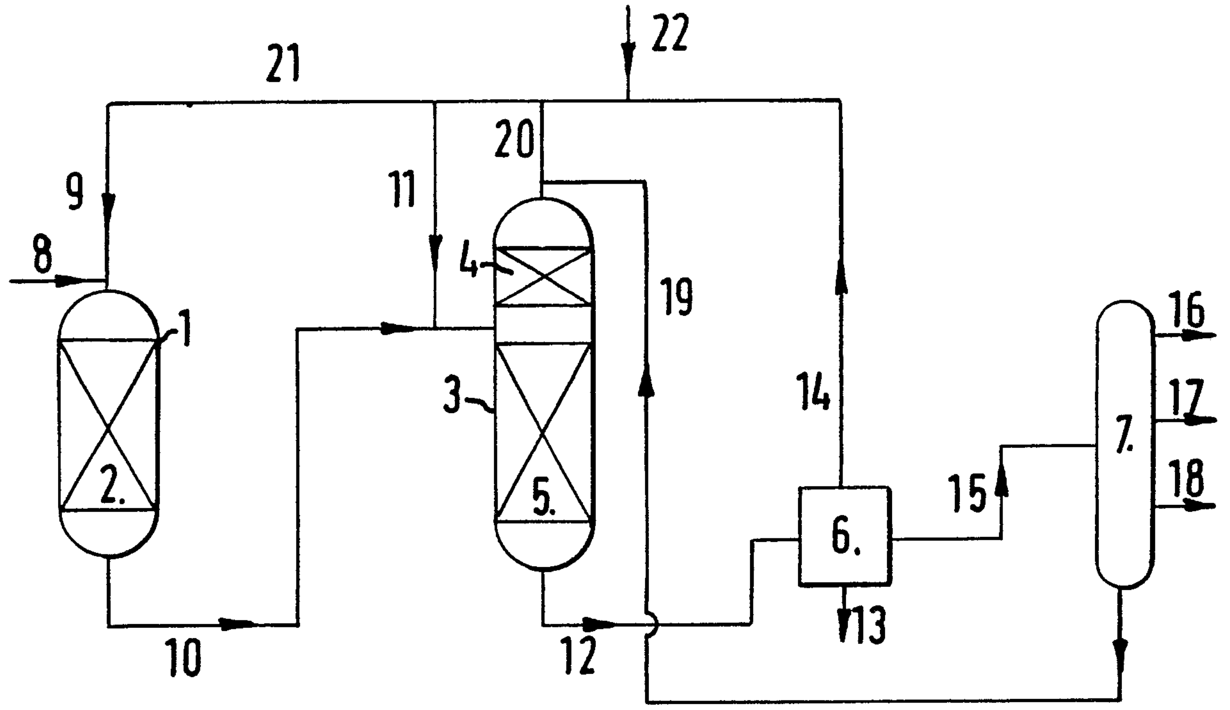
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FIG.1

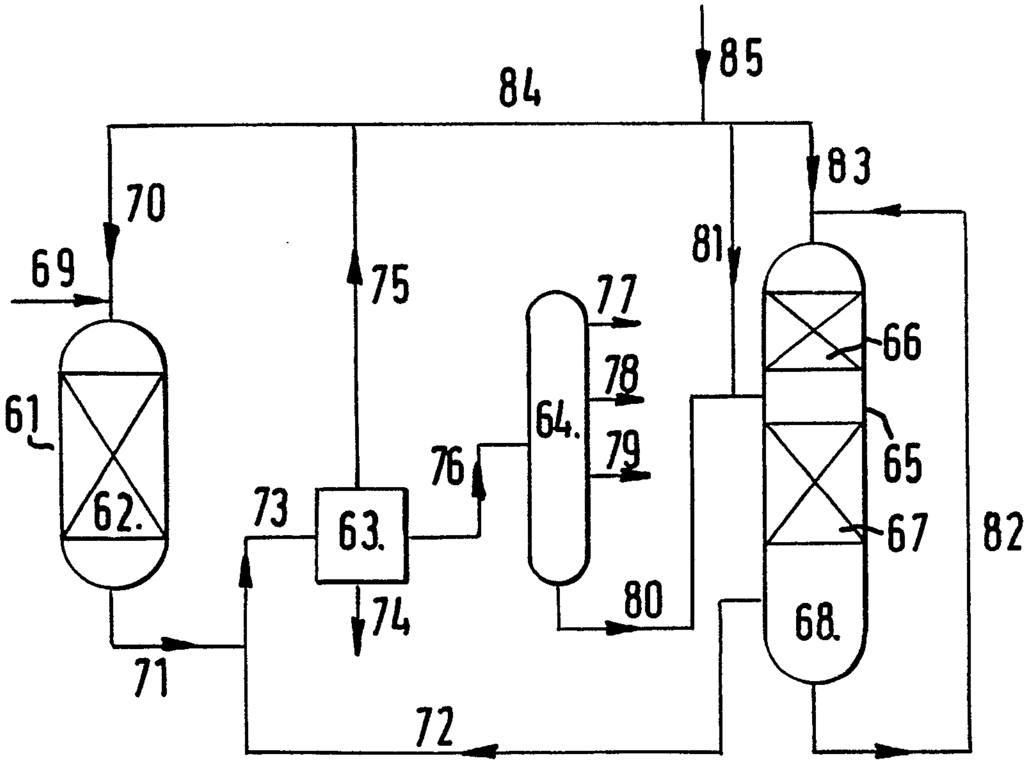
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FIG.3

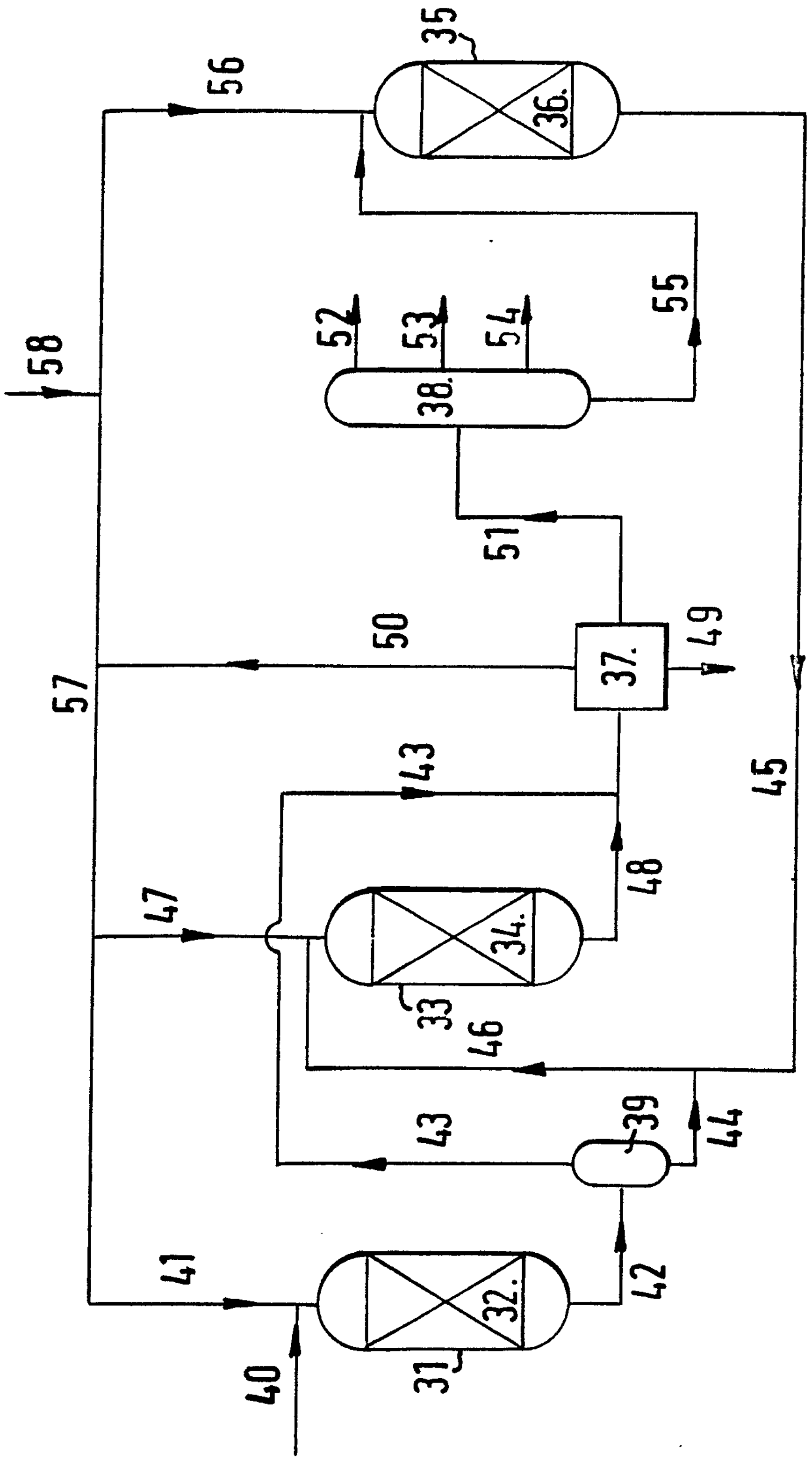
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FIG. 2

EUROPEAN SEARCH REPORT

Application Number

EP 89 20 2073

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.5)
Y	US-A-3 385 781 (HAMNER et al.) * Figure 1 *	1-15	C 10 G 65/10
Y	US-A-3 203 890 (HAENSEL) * Figure *	1-15	
A	US-A-4 404 088 (BACHTEL et al.) * Figure 2; claim 1; column 3, line 23, column 4, line 11 *	2,11,13	
A	GB-A-1 191 958 (SHELL) * Figure *	1	

TECHNICAL FIELDS SEARCHED (Int. Cl.5)

C 10 G

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
THE HAGUE	24-10-1989	MICHIELS P.

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
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L : document cited for other reasons

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