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AUSTRALIA
PATENTS ACT 1990
NOTICE OF ENTITLEMENT

We, **Linde Aktiengesellschaft**, the applicant/Nominated Person in respect of Application No. 40672/93 state the following:-

The Nominated Person is entitled to the grant of the patent because the Nominated Person would, on the grant of a patent for the invention to the inventors, be entitled to have the patent assigned to the Nominated Person.

The Nominated Person is entitled to claim priority from the application listed in the declaration under Article 8 of the PCT because the Nominated Person made the application listed in the declaration under Article 8 of the PCT.

DATED this TWENTIETH day of MARCH 1995



.....
a member of the firm of
DAVIES COLLISON
CAVE for and on behalf
of the applicant(s)

(DCC ref: 1713500)



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METHOD OF PRODUCING LIGHT C₂₊-HYDROCARBONS FROM CRACKED GAS
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- (57) Claim

1. A process for the recovery of low molecular weight C₂₊ hydrocarbons, in particular ethylene and ethane, from a gas mixture obtained by the cracking of hydrocarbons and having been freed of high molecular weight hydrocarbons, characterised in that the low molecular weight C₂₊ hydrocarbons are scrubbed from the gas mixture by absorption by means of an organic, physically acting scrubbing agent in an absorption column, the scrubbing agent having a molecular weight between 50 and 75 g/mol, the scrubbing agent being passed in a circuit to be first loaded with low molecular weight hydrocarbons, being regenerated in a regenerating column and subsequently being returned into the absorption column for renewed loading.

15. A process according to any one of claims 1 to 14, characterised in that the temperature in the sump of the regenerating column does not exceed 105°C.

17. A process according to any one of claims 1 to 16, characterised in that the gas containing C₂ hydrocarbons and C₃ hydrocarbons after having been withdrawn from the head of the regenerating column and being separated from condensate after its partial condensation, is subjected to a C₂/C₃ separation and the C₂ hydrocarbons withdrawn from the C₂/C₃ separation are separated further into ethylene- and ethane-containing product gas flows.



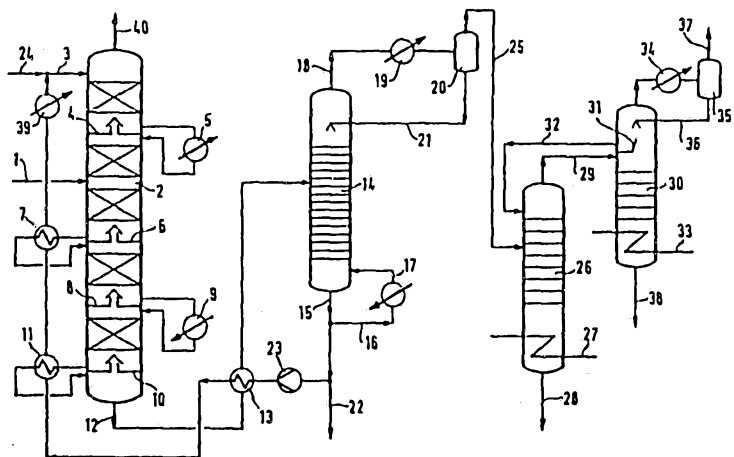
<p>(51) Internationale Patentklassifikation ⁵ : C07C 7/11</p>	<p>AI</p>	<p>(11) Internationale Veröffentlichungsnummer: WO 93/24428 (43) Internationales Veröffentlichungsdatum: 9. Dezember 1993 (09.12.93)</p>
<p>(21) Internationales Aktenzeichen: PCT/EP93/01180 (22) Internationales Anmeldedatum: 12. Mai 1993 (12.05.93) (30) Prioritätsdaten: P 42 17 611.5 27. Mai 1992 (27.05.92) DE (71) Anmelder (für alle Bestimmungsstaaten ausser US): LINDE AKTIENGESELLSCHAFT [DE/DE]; Abraham-Lincoln-Strasse 21, D-6200 Wiesbaden (DE). (72) Erfinder; und (75) Erfinder/Anmelder (nur für US): BAUER, Heinz [DE/DE]; Bertelestrasse 49, D-81479 München (DE). BECKER, Hans [DE/DE]; Zwengauer Weg 9, D-81479 München (DE). (74) Anwalt: KASSECKERT, Rainer; Linde Aktiengesellschaft, Zentrale Patentabteilung, D-82049 Höllriegelskreuth (DE).</p>	<p>(81) Bestimmungsstaaten: AU, CA, JP, KR, NO, US, europäisches Patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE). Veröffentlicht Mit internationalem Recherchenbericht.</p> <p style="font-size: 2em; text-align: center;">67 1277</p>	

(54) Title: METHOD OF PRODUCING LIGHT C₂₊-HYDROCARBONS FROM CRACKED GAS

(54) Bezeichnung: VERFAHREN ZUR GEWINNUNG LEICHTER C₂₊-KOHLENWASSERSTOFFE AUS EINEM SPALTGAS

(57) Abstract

The invention concerns a method for the production of light C₂₊-hydrocarbons, in particular ethylene and ethane, from cracked gas, in particular from a gas produced by a fluid catalytic cracking process. The invention proposes that the light C₂₊-hydrocarbons are scrubbed out of the cracked gas by absorption in an organic, preferably paraffinic, scrubbing agent with a physical action, the scrubbing agent having a molecular weight between 50 and 75 g/mole, preferably between 60 and 75 g/mole. Pentane, iso-pentane or mixtures thereof are particularly suitable for use as the scrubbing agent. Before the used scrubbing agent is regenerated, any methane which has also been scrubbed out is preferably stripped from the agent.



(57) Zusammenfassung

Die Erfindung betrifft ein Verfahren zur Gewinnung leichter C₂₊-Kohlenwasserstoffe, insbesondere Ethylen und Ethan, aus einem Spaltgas, insbesondere aus einem FCC-Abgas. Erfindungsgemäß werden die leichten C₂₊-Kohlenwasserstoffe aus dem Spaltgas durch Absorption mit einem organischen, vorzugsweise paraffinischen, physikalisch wirkenden Waschmittel ausgewaschen, wobei das Waschmittel ein Molekulargewicht zwischen 50 und 75 g/mol, vorzugsweise zwischen 60 und 75 g/mol, aufweist. Als Waschmittel eignen sich besonders Pentan, Isopentan oder Mischungen daraus. Vor der Regenerierung des beladenen Waschmittels wird mit Vorteil mitausgewaschenes Methan ausgestrippt.

Process for the recovery of low molecular weight
 C_2+ hydrocarbons from a cracking gas

The invention relates to a process for the recovery of low molecular weight C_2+ hydrocarbons, in particular ethylene and ethane, from a gas mixture obtained by the cracking of hydrocarbons and having been freed of high molecular weight hydrocarbons.

In various cracking gas processes, in particular in fluidized catalytic cracking (FCC) a gas mixture is obtained which comprises inter alia low molecular weight hydrocarbons, in particular ethylene and ethane, but also C_3 and C_4 hydrocarbons. In most refineries the high molecular weight hydrocarbons (C_5+ hydrocarbons) are first separated from the FCC waste gas by fractional distillation. Thereafter the C_3+ hydrocarbons are recovered by oil scrubbing, whereas the entire C_2 hydrocarbons, but also a certain proportion of C_3- and C_4 hydrocarbons are discharged into the fuel gas grid.

In the event that C_2 hydrocarbons as well are to be recovered from the FCC waste gas, this is done by partial condensation in a low temperature process. Such a process is known for example from EP-B 0 185 202. Because the FCC waste gases invariably contain traces of higher, polyunsaturated hydrocarbons, nitrogen oxides and oxygen, this process always involves a risk that explosive resins may form in the very low temperature items of equipment. A plant with partial condensation in a low temperature process accordingly always entails a certain safety risk. Thus, for example in February, 1990, such a plant exploded in France after having been in operation for eight years.



A process for removing ethane and ethylene by absorption from a gas mixture coming from a coal gasification is known from US-A-4 035 167 whereby cyclohexane, cyclohexene, 1-chlorobutane or 1,1,1-trichloroethane is used as scrubbing agent for the absorption.

A process for absorption of ethylene and ethane from a cracking gas is described in DE-B-1 088 477 whereby after separation of H₂, CO, N₂ and methane, the remaining C₂₊ hydrocarbon mixture, ~~is fed with the contaminants of an acetone wash.~~ ^{including the contaminants is fed to an acetone wash.}



We have now found C_{2+} hydrocarbons can be recovered from the input gas in a simple manner, but where the safety of the plant is warranted by a special mode of operation.

Accordingly there is provided a process for the recovery of low molecular weight C_{2+} hydrocarbons, in particular ethylene and ethane, from a gas mixture obtained by the cracking of hydrocarbons and having been freed of high molecular weight hydrocarbons, characterised in that the low molecular weight C_{2+} hydrocarbons are scrubbed from the gas mixture by absorption by means of an organic, physically acting scrubbing agent in an absorption column, the scrubbing agent having a molecular weight between 50 and 75 g/mol, the scrubbing agent being passed in a circuit to be first loaded with low molecular weight hydrocarbons, being regenerated in a regenerating column and subsequently being returned into the absorption column for renewed loading. The low molecular weight C_{2+} hydrocarbons are scrubbed from the gas mixture by absorption by means of an organic, preferably paraffinic, physically acting scrubbing agent in an absorption column, the scrubbing agent having a molecular weight between 50 and 75 g/mol, preferably between 60 and 75 g/mol, the scrubbing agent being passed in a circuit to be first loaded with low molecular weight hydrocarbons, being regenerated in a regenerating column and subsequently being returned into the absorption column for renewed loading.

The process according to the invention offers the advantage that the low molecular weight hydrocarbons are removed from the gas mixture during gas scrubbing, thereby avoiding a safety risk in that the formation of explosive resins is prevented by this procedure.

Advantageously a C_{4+} hydrocarbon fraction, preferably pentane, isopentane or mixtures thereof, is employed as a scrubbing agent. Such a scrubbing agent is characterised by a very high solvent power for C_2 hydrocarbons, in particular ethylene and ethane, but also for other readily soluble components. Light gasoline comprising proportions within the above described molecular weight range, in particular pentane compounds are obtained during the pretreatment of the feed gas, for example in the fractional distillation, and are accordingly readily available.



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In a preferred embodiment of the process according to the invention the loaded scrubbing agent, prior to its being fed into the regeneration column, is stripped with a stripping gas, resulting in particular in the stripping off of co-absorbed

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methane. The methane thereby recovered can be further processed or may for example be used as a fuel gas.

Particular advantages result if the stripping of the loaded scrubbing agent is conducted in a lower section of the absorption column. This procedure, apart from simplifications in respect of apparatus requirements, offers the advantage that methane dissolved in the scrubbing agent can even be stripped off already from the scrubbing agent in the absorption region in addition to the stripping region, since the stripping gas can flow through the entire absorption column.

Further advantages result from the feature that evaporated scrubbing agent is employed as a stripping gas. For that purpose heat is introduced into the lower region of the absorption column, for example by a boiling vessel, causing a certain part of the scrubbing agent to evaporate. Due to the lower temperatures in the upper region of the absorption column the evaporated scrubbing agent is condensed once again and can be reloaded with low molecular weight hydrocarbons. Stripping with a foreign stripping gas might result in contamination of the ethylene product flow.

The gas mixture is advantageously fed into the absorption column at a pressure of 4 to 50 bar, preferably 10 to 30 bar. Although an elevated pressure during absorption entails increased energy expenditures, it also substantially decreases the amount of required scrubbing agent. Thus, for example, the scrubbing agent throughput required may be reduced by half if the pressure is increased from 4 bar to 30 bar.

Advantageously the absorption is carried out at temperatures between 0 and -50°C, preferably between -30° and -50°C, particularly preferably between -35 and -45°C. Although even lower temperatures increase the solvent power of the scrubbing agent for the low molecular weight hydrocarbons they on the other hand may cause a possible increase of NO₂ formation. Therefore, when



employing feed flows having a relatively high NO_x content a comparatively high temperature for the absorption should be selected. The cold overhead product of the absorption column can be employed for cooling the cracking gas mixture.

After the components lighter than ethylene, for example methane, have been stripped off the loaded scrubbing agent, the scrubbing agent loaded with C_2+ hydrocarbons is passed into a regenerating column. The regenerating column is operated at temperatures higher than during the absorption. The highest temperatures in the process according to the invention prevail in the lower region of the regenerating column. There the scrubbing agent is heated preferably by means of a boiling vessel in order to obtain desorption of the C_3- hydrocarbons from the scrubbing agent. The temperatures in the sump of the regenerating column are thus advantageously so selected that 120°C , preferably 105°C is not exceeded. The reason is that it was found that polymerisation for example to form butadien will then be avoided. It is difficult to remove polymerisation products from the scrubbing agent. Accordingly the temperature in the sump region of the regenerating column is predetermined at the lower limit by an inadequate desorption of the C_3- hydrocarbons from the scrubbing agent and at the upper limit by incipient polymerisation.

In principle it is possible for the regeneration of the scrubbing agent to be performed at a pressure which is lower or higher than that for the absorption. However, preferably the pressure in the regenerating column is below that of the absorption column.

From the head of the regenerating column a gas flow is withdrawn, partly condensed, passed into a separator and the condensate reintroduced from the separator as reflux into the regenerating column for reducing the losses of scrubbing agent. In addition, a product gas flow rich in C_2/C_3 hydrocarbons is recovered by the separator. C_4+ hydrocarbons passed into the regeneration are



returned from the sump of the regenerating column to the absorption column jointly with or serving as regenerated scrubbing agent.

In accordance with a further development of the process according to the invention, the head product of the regenerating column is partly condensed and after the condensate has been separated and returned into the regenerating column, the C_2 and C_3 hydrocarbon-rich gas flow is subjected to a C_2/C_3 separation. In the C_2/C_3 separation a C_2 hydrocarbon flow is recovered which in a further separating stage is separated into ethylene- and ethane-containing product gas flow.

In the following the invention will be further elucidated with reference to a working example.

There is shown in:-

Fig.1: The process according to the invention including absorption, regeneration and further separation of the C_2 and C_3 hydrocarbons by distillation.

It is to be noted that the totals of the data listed in the individual tables concerning the composition of individual flows, due to approximations may exceed 100%.

By way of duct 1 an FCC cracking waste gas, freed by fractional distillation of C_4+ hydrocarbons and dried, is fed into the absorption column 2 at a pressure of 13,3 bar and a temperature of 22°C. In this case the FCC waste gas has the following composition:-



H ₂	3,4 Weight %
N ₂	11,8 Weight %
CO	1,1 Weight %
CH ₄	31,6 Weight %
C ₂ H ₄	20,5 Weight %
C ₂ H ₆	28,4 Weight %
C ₃ H ₆	2,0 Weight %
C ₃ H ₈	1,0 Weight %
other components	< 0,1 Weight %

By way of duct 3 regenerated scrubbing agent composed to about three quarters of pentane and to about one quarter of isopentane is fed into the upper region of the absorption column 2 at a temperature of -40°C and a pressure of 13,1 bar. Above the chimney tray 4 the scrubbing agent is withdrawn, cooled in the side cooler 5 in order to withdraw the process heat formed and once again introduced into the absorption column. Above the chimney tray 6 loaded scrubbing agent is warmed up by indirect heat exchange (7) with regenerated scrubbing agent and returned again into the absorption column 2. Above the chimney tray 8 the scrubbing agent is once again withdrawn and further heated in the boiler vessel 9, before once again being fed into the absorption column 2 underneath the chimney tray 8. Above the chimney tray 10 the scrubbing agent is once again withdrawn and further heated by indirect heat exchange (11) with regenerated scrubbing agent and is fed into the lowermost section of the absorption column 2. The three column sections underneath the chimney tray 6 serve for the removal of components lighter than ethylene from the scrubbing agent. By the heating of the scrubbing agent, scrubbing agent vapour is formed which is employed as stripping gas for stripping off the components which are lighter than ethylene, in particular methane. Overhead of the absorption column 2 a gas flow is thus withdrawn which is virtually free of C₂+ hydrocarbons and which has the following composition:



H ₂	6,7 Weight %
N ₂	23,0 Weight %
O ₂	0,1 Weight %
CO	2,2 Weight %
CH ₄	61,6 Weight %

and traces of C₂₊ hydrocarbons.

From the sump of the absorption column 2 a scrubbing agent loaded with C₂₊ hydrocarbons and having a temperature of 54°C and a pressure of 13,4 bar is withdrawn:

C ₂ H ₄	3,4 Weight %
C ₂ H ₆	5,2 Weight %
C ₃ H ₆	0,8 Weight %
C ₃ H ₈	0,3 Weight %
C ₅ H ₁₂	65,6 Weight %
C ₅ H ₁₂ (Iso)	24,6 Weight %
other components	< 0,1 Weight %

The loaded scrubbing agent from duct 12 is further heated by indirect heat exchange (13) with indirect scrubbing agent and introduced into the regenerating column 14. In the regenerating column 14 the hot regeneration of the scrubbing agent takes place. For that purpose regenerated scrubbing agent from the sump of the regenerating column (15) is partly (16) heated in the boiling vessel 17 to 105°C and returned into the lower region of the regenerating column. The scrubbing agent vapour thereby formed strips the C₂ and C₃ hydrocarbons off the scrubbing agent. The overhead product of the regenerating column (18) having a temperature of 57°C and a pressure of 7,6 bar is cooled in a condenser 19 and introduced into a separator 20. Components which have condensed out,



for the predominant part scrubbing agent which has condensed out are returned again from the separator 20 into the upper region of the regenerating column 14. The scrubbing agent flow withdrawn from the sump of the regenerating column 14 by way of duct 15 at a temperature of 102°C and a pressure of 7,8 bar and which is not returned by way of duct 16 into the regenerating column is raised by pump 23 to a pressure of 14 bar and cooled in the heat exchangers 13, 11, 7 and 39 to -40°C. The composition of the regenerated scrubbing agent withdrawn from the regenerating column is as follows:

C ₂ H ₆	0,4 Weight %
C ₃ H ₆	0,5 Weight %
C ₃ H ₈	0,2 Weight %
C ₅ H ₁₂	72,0 Weight %
C ₅ H ₁₂ (Iso)	27,0 Weight %
other components	< 0,1 Weight %

By way of duct 22 a small amount of scrubbing agent purge is separated off. A corresponding amount of scrubbing agent is added to the cooled scrubbing agent in duct 3 by way of duct 24.

By the separator 20 a C₂-/C₃ hydrocarbon gas mixture is recovered in the duct 25 having the following composition:

C ₂ H ₄	38,7 Weight %
C ₂ H ₆	55,2 Weight %
C ₃ H ₆	3,7 Weight %
C ₃ H ₈	1,9 Weight %
C ₅ H ₁₂	0,2 Weight %
C ₅ H ₁₂ (iso)	0,4 Weight %
other components	< 0,1 Weight %



This gas flow is subsequently passed to a fractional distillation and for that purpose is first introduced into column 26 for a C₂-/C₃ hydrocarbon separation. Column 26 is equipped with sump heating 27. From the sump of the column 26 a distillate (LPG) which is essentially composed of C₃ hydrocarbons is withdrawn by way of duct 28, with the following composition:

C ₃ H ₆	58,8 Weight %
C ₃ H ₈	30,9 Weight %
C ₄ H ₆	0,3 Weight %
C ₅ H ₁₂	3,2 Weight %
C ₅ H ₁₂ (Iso)	6,7 Weight %
other	
components	< 0,1 Weight %

The overhead product of C₂ hydrocarbons having the following composition:

C ₂ H ₄	36,9 Weight %
C ₂ H ₆	62,9 Weight %
C ₃ H ₆	0,1 Weight %
other	
components	< 0,1 Weight %

is fed by way of ducts 29 into the column 30. In order to keep the C₃H₆/C₃H₈+ components in ducts 29 as low as possible, the flow 32 withdrawn above the side tray 31 is returned as reflux into the upper region of the column 26. The reflux 32 has the following composition:

C ₂ H ₄	24,6 Weight %
C ₂ H ₆	75,3 Weight %
other	
components	< 0,1 Weight %



Column 30 serves for splitting the C_2 compounds. For that purpose the sump region of column 30 is heated by way of the sump heating means 33 whilst the overhead product is partly condensed (34) by column 30, and the condensate withdrawn by a separator 35 is returned as reflux into the column 30 by way of duct 36, there to attain a back-washing of the components which are heavier than ethylene. More than 99,9% of the overhead product of column 30 is represented by ethylene, so that by way of duct 37 a pure ethylene product flow with minimal contaminations is recovered. A sump product having the composition:

C_2H_4	0,9 Weight %
C_2H_6	98,8 Weight %
C_3H_6	0,3 Weight %
other	
components	< 0,1 Weight %

is withdrawn by way of duct 38 from the sump of the column 30. This ethane fraction from duct 38 may for example be mixed with the overhead product of the absorption column 2 in duct 40 and be used as a fuel gas.

The columns 26 and 30 may be combined in a single column in which case from the head of this column likewise a ethylene flow is recovered, whereas from the sump of the column a C_3 hydrocarbon/ethane mixture is withdrawn.



THE CLAIMS DEFINING THE INVENTION ARE AS FOLLOWS:

1. A process for the recovery of low molecular weight C_{2+} hydrocarbons, in particular ethylene and ethane, from a gas mixture obtained by the cracking of hydrocarbons and having
5 been freed of high molecular weight hydrocarbons, characterised in that the low molecular weight C_{2+} hydrocarbons are scrubbed from the gas mixture by absorption by means of an organic, physically acting scrubbing agent in an absorption column, the scrubbing agent having a molecular weight between 50 and 75 g/mol, the scrubbing agent being passed in a circuit to be first loaded with low molecular weight hydrocarbons, being regenerated in a
10 regenerating column and subsequently being returned into the absorption column for renewed loading.
2. A process according to claim 1 wherein said organic physically acting scrubbing agent is paraffinic.
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3. A process according to either claim 1 or claim 2 wherein said organic physically acting scrubbing agent has a molecular weight between 60 and 75 g/mol.
4. A process according to any one of claims 1 to 3, characterised in that a C_{4+}
20 hydrocarbon fraction is employed as the scrubbing agent.
5. A process according to claim 4 wherein the scrubbing agent is selected from the group consisting of pentane, isopentane or mixtures thereof.
- 25 6. A process according to any one of claims 1 to 5, characterised in that the loaded scrubbing agent prior to being fed into the regenerating column is stripped by means of a stripping gas, whereby in particular co-absorbed methane is stripped off.
7. A process according to claim 6, characterised in that the stripping of the loaded
30 scrubbing agent is carried out in a lower section of the absorption column.



8. Process according to either claim 6 or 7, characterised in that evaporated scrubbing agent is employed as a stripping gas.

9. A process according to any one of claims 1 to 8, characterised in that the cracking gas is fed into the absorption column at a pressure of 4 to 50 bar.

10. A process according to any one of claims 1 to 9, characterised in that the cracking gas is fed into the absorption column at a pressure of 10 to 30 bar.

10 11. A process according to any one of claims 1 to 10, characterised in that the absorption is carried out at temperatures between 0 and -50°C.

12. A process according to any one of claims 1 to 11, characterised in that the absorption is carried out at temperatures between -30 and -50°C.

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13. A process according to any one of claims 1 to 12, characterised in that the absorption is carried out at temperatures between -35 and -45°C.

14. A process according to any one of claims 1 to 13, characterised in that the temperature in the sump of the regenerating column does not exceed 120°C.

15. A process according to any one of claims 1 to 14, characterised in that the temperature in the sump of the regenerating column does not exceed 105°C.

25 16. A process according to any one of claims 1 to 15, characterised in that the pressure in the regenerating column is below that of the absorption column.

17. A process according to any one of claims 1 to 16, characterised in that the gas containing C₂ hydrocarbons and C₃ hydrocarbons after having been withdrawn from the head of the regenerating column and being separated from condensate after its partial

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condensation, is subjected to a C₂/C₃ separation and the C₂ hydrocarbons withdrawn from the C₂/C₃ separation are separated further into ethylene- and ethane-containing product gas flows.

5 18. A process substantially as described herein with reference to the drawings.

DATED this 26th day of June 1996.

LINDE AKTIENGESELLSCHAFT

10 by their Patent Attorneys

DAVIES COLLISON CAVE

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I. KLASSIFIKATION DES ANMELDUNGSGEGENSTANDS (bei mehreren Klassifikationssymbolen sind alle anzugeben) ⁶		
Nach der Internationalen Patentklassifikation (IPC) oder nach der nationalen Klassifikation und der IPC		
Int.Kl. 5 C07C7/11		
II. RECHERCHIERTE SACHGEBIETE		
Recherchierter Mindestprüfstoff ⁷		
Klassifikationssystem	Klassifikationssymbole	
Int.Kl. 5	C07C ; C10G	
Recherchierte nicht zum Mindestprüfstoff gehörende Veröffentlichungen, soweit diese unter die recherchierten Sachgebiete fallen ⁸		
III. EINSCHLAGIGE VERÖFFENTLICHUNGEN ⁹		
Art. ^o	Kennzeichnung der Veröffentlichung ¹¹ , soweit erforderlich unter Angabe der maßgeblichen Teile ¹²	Betr. Anspruch Nr. ¹³
A	US,A,4 035 167 (CONTINENTAL OIL COMPANY) 12. Juli 1977	
A	DE,B,1 088 477 (LINDE) 8. September 1960	
<p>^o Besondere Kategorien von angegebenen Veröffentlichungen ¹⁰ :</p> <p>"A" Veröffentlichung, die den allgemeinen Stand der Technik definiert, aber nicht als besonders bedeutsam anzusehen ist</p> <p>"E" älteres Dokument, das jedoch erst am oder nach dem internationalen Anmeldedatum veröffentlicht worden ist</p> <p>"L" Veröffentlichung, die geeignet ist, einen Prioritätsanspruch zweifelhaft erscheinen zu lassen, oder durch die das Veröffentlichungsdatum einer anderen im Recherchenbericht genannten Veröffentlichung belegt werden soll oder die aus einem anderen besonderen Grund angegeben ist (wie ausgeführt)</p> <p>"O" Veröffentlichung, die sich auf eine mündliche Offenbarung, eine Benutzung, eine Ausstellung oder andere Maßnahmen bezieht</p> <p>"P" Veröffentlichung, die vor dem internationalen Anmeldedatum, aber nach dem beanspruchten Prioritätsdatum veröffentlicht worden ist</p> <p>"T" Spätere Veröffentlichung, die nach dem internationalen Anmeldedatum oder dem Prioritätsdatum veröffentlicht worden ist und mit der Anmeldung nicht kollidiert, sondern nur zum Verständnis des der Erfindung zugrundeliegenden Prinzips oder der ihr zugrundeliegenden Theorie angegeben ist</p> <p>"X" Veröffentlichung von besonderer Bedeutung; die beanspruchte Erfindung kann nicht als neu oder auf erfinderischer Tätigkeit beruhend betrachtet werden</p> <p>"Y" Veröffentlichung von besonderer Bedeutung; die beanspruchte Erfindung kann nicht als auf erfinderischer Tätigkeit beruhend betrachtet werden, wenn die Veröffentlichung mit einer oder mehreren anderen Veröffentlichungen dieser Kategorie in Verbindung gebracht wird und diese Verbindung für einen Fachmann naheliegend ist</p> <p>"&" Veröffentlichung, die Mitglied derselben Patentfamilie ist</p>		
IV. BESCHEINIGUNG		
Datum des Abschlusses der Internationalen Recherche	Absenddatum des Internationalen Recherchenberichts	
19.AUGUST 1993	02.09.93	
Internationale Recherchenbehörde	Unterschrift des bevollmächtigten Bediensteten	
EUROPAISCHES PATENTAMT	Oswald De Herdt	

**ANHANG ZUM INTERNATIONALEN RECHERCHENBERICHT
 ÜBER DIE INTERNATIONALE PATENTANMELDUNG NR.**

EP 9301180
 SA 74026

In diesem Anhang sind die Mitglieder der Patentfamilien der im obengenannten internationalen Recherchenbericht angeführten Patentdokumente angegeben.
 Die Angaben über die Familienmitglieder entsprechen dem Stand der Datei des Europäischen Patentamts am
 Diese Angaben dienen nur zur Unterrichtung und erfolgen ohne Gewähr.

19/08/93

Im Recherchenbericht angeführtes Patentdokument	Datum der Veröffentlichung	Mitglied(er) der Patentfamilie	Datum der Veröffentlichung
US-A-4035167	12-07-77	Keine	
DE-B-1088477		Keine	

EPO FORM P043

Für nähere Einzelheiten zu diesem Anhang : siehe Amtsblatt des Europäischen Patentamts, Nr.12/82

INTERNATIONAL SEARCH REPORT

International application No.
PCT/EP93/01180

A. CLASSIFICATION OF SUBJECT MATTER

Int.Cl.5 C07C 7/11

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

Int.Cl.5 C07C; C10G

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US, A, 4 035 167 (CONTINENTAL OIL COMPANY) 12 July 1977	
A	DE, B, 1 088 477 (LINDE) 8 September 1960	

 Further documents are listed in the continuation of Box C. See patent family annex.

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Date of the actual completion of the international search

19 August 1993 (19.08.93)

Date of mailing of the international search report

2 September 1993 (02.09.93)

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**ANNEX TO THE INTERNATIONAL SEARCH REPORT
ON INTERNATIONAL PATENT APPLICATION NO.**

EP 9301180
SA 74026

This annex lists the patent family members relating to the patent documents cited in the above-mentioned international search report. The members are as contained in the European Patent Office EDP file on
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Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US-A-4035167	12-07-77	None	
DE-B-1088477		None	

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For more details about this annex : see Official Journal of the European Patent Office, No. 12/82