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DECISION
of 15 September 2004

Case Number: T 0616/03 - 3.3.6

Application Number: 95922129.2

Publication Number: 0765369

IPC: C10G 67/00

Language of the proceedings: EN

Title of invention:

Process for the removal of mercaptans and hydrogen sulfide
from hydrocarbon streams

Applicant:

CHEMICAL RESEARCH & LICENSING COMPANY

Opponent:

-

Headword:

Mercaptans and/or hydrogen sulfide removal/CHEMICAL RESEARCH &
LICENSING

Relevant legal provisions:

EPC Art. 54, 56, 83, 84

Keyword:

"Clarity and sufficiency of disclosure (main request) - no:
insufficient characterization of the invention - undue burden"
"Clarity and sufficiency of disclosure (auxiliary request) -
yes"
"Novelty and inventive step (auxiliary request) - yes"

Decisions cited:

T 0782/01

Catchword:

-



w Case Number: T 0616/03 - 3.3.6

D E C I S I O N
of the Technical Board of Appeal 3.3.6
of 15 September 2004

Appellant:

CHEMICAL RESEARCH & LICENSING COMPANY
10100 Bay Area Boulevard
Pasadena
Texas 77507 (US)

Representative:

Zumstein, Fritz, Dr.
Zumstein & Klingseisen
Patentanwälte
Bräuhausstrasse 4
D-80331 München (DE)

Decision under appeal:

Decision of the Examining Division of the
European Patent Office posted 13 December 2002
refusing European application No. 95922129.2
pursuant to Article 97(1) EPC.

Composition of the Board:

Chairman: P. Krasa
Members: L. Li Voti
J. H. Van Moer

Summary of Facts and Submissions

I. This appeal lies from the decision of the Examining Division to refuse European patent application no. 95 922 129.2, relating to a process for the removal of mercaptans and/or hydrogen sulfide from hydrocarbon streams.

II. In its decision, the Examining Division, referring to document

(1): US-A-5321163,

found *inter alia* that

- the process disclosed in document (1) differed from that claimed in the present application insofar as it required the additional step of reacting the products of the first distillation reaction zone with methanol in the same distillation column reactor in order to produce tertiary amyl methyl ether (**TAME**), whereas the claims of the present application required such products to be withdrawn from the column before being used in a further reaction, e.g. for the preparation of TAME;
- to carry out such an etherification step alternatively in a different reactor was an obvious choice for the skilled person;
- the claimed subject-matter lacked thus an inventive step in the light of the teaching of this document.

III. An appeal was filed against this decision by the Applicant (Appellant).

During the appeal the following documents were *inter alia* cited by the Board and by the Appellant, respectively:

(2): "Einführung in die thermische Verfahrenstechnik" by Prof. Dr. P. Grassmann, Walter de Gruyter & Co. ed., 1967, pages 74 to 77 and 109 to 116";

(7): Ullmann's Encyclopedia of Industrial Chemistry, 5th edition, 1988, vol. B3, page 4-85.

A new main request based on an amended claim 1 was filed by the Appellant under cover of a letter dated 18 August 2004. During the oral proceedings held before the Board on 15 September 2004 the Appellant modified said claim 1 and filed a new auxiliary request.

Claim 1 according to the main request reads as follows:

"1. A process for removing mercaptans and/or hydrogen sulfide from a hydrocarbon stream to produce a hydrocarbon stream with a reduced mercaptan and/or sulfide content, comprising the steps of: (a) feeding diolefins and a hydrocarbon stream containing mercaptans and/or hydrogen sulfide to a distillation column reactor into a feed zone in said reactor, whereby the said diolefins and the said hydrocarbon stream are together with the hydrogen of step (b) the only feeding components; (b) feeding hydrogen to said distillation column reactor at a rate to maintain the

catalyst in the active form and below that which would cause flooding of the column; (c) concurrently in said distillation column reactor (i) contacting the diolefins with said mercaptans, hydrogen sulfide or mixtures thereof contained within said hydrocarbon stream in the presence of hydrogen in a distillation reaction zone containing a catalyst bed containing a supported Group VIII metal oxide catalyst prepared in the form of a catalytic distillation structure and reacting a portion of said mercaptans and/or hydrogen sulfide with a portion of the diolefins thereby forming sulfide products and a distillate product, having a reduced mercaptan and/or hydrogen sulfide content and (ii) separating said sulfides from said distillate product by fractional distillation; (d) withdrawing distillate product from said distillation column reactor at a point above said distillation reaction zone, said distillate product having a reduced mercaptan and/or hydrogen sulfide content; and (e) withdrawing sulfide products from said distillation column reactor at a point below said distillation reaction zone, said distillation column reactor being operated under conditions to maintain froth throughout the catalyst bed by control of the bottoms and/or overheads withdrawal rate and the pressure in the distillation column reactor being such that the mixture is boiling in the catalyst bed."

Claim 1 of the **auxiliary request** differs from claim 1 of the main request insofar as it specifies that **the overhead pressure in the distillation column reactor is between 96 and 1820 kPa (0 and 250 psig) and the temperature in the distillation reaction zone is between 38 and 149°C (100 to 300°F).**

Both requests contain dependent claims 2 to 11 relating to particular embodiments of the claimed process.

IV. The Appellant submitted that

- claim 1 according to both requests required that the process had to be carried out by operating below the flooding point but under conditions to maintain froth, i.e. a continuous liquid containing gas bubbles dispersed within, throughout the catalyst bed, i.e. under conditions of "artificial" flooding enabling formation of froth in the catalyst bed though maintaining a countercurrent flow of vapour and liquid in the column;
- the skilled person would have been able to recognise the occurrence of froth throughout the catalyst bed and it would have been also able to operate the distillation column reactor under the required conditions by following the teaching of the application, e.g. by adjusting the bottoms and/or overheads withdrawal rate, this process feature being contained in claim 1 of the main request, or by further selecting specific values of overhead pressure of the column and temperature in the catalytic reaction zone as specified in claim 1 of the auxiliary request;
- the requirements of Articles 83 and 84 EPC were thus complied with.

As regards novelty and inventive step the Appellant submitted *inter alia* that

- diolefins, hydrocarbon stream and hydrogen were the only components fed to the distillation column reactor according to the claims of the present application, whilst the process of document (1) required the addition of methanol to the same column in order to obtain an ether; moreover, document (1) did not specify that the distillation column had to be operated under specific conditions in order to maintain froth throughout the catalyst bed though operating under the flooding point;
- the presence of froth throughout the catalyst bed was found to improve the efficiency of the catalyst thereby allowing a reduction of the height of the catalyst used and bringing about a better separation of the reaction products; moreover, the absence of a further etherification reaction in the same distillation reactor column allowed the selection of a broader range of process conditions and increased the flexibility of the process;
- as explained in document (7), it was common general knowledge at the priority date of the present application that froth could be formed in a packed distillation column only after having reached the flooding point, i.e. under conditions which had to be avoided under any circumstance; furthermore, document (2) taught only that a spray, i.e. liquid droplets dispersed in vapour, or a

bubbles layer but not froth could be formed in a packed distillation column by operating between the loading and the flooding point;

- since according to the prior art the formation of froth throughout the catalyst bed would have been expected only at or above the flooding point, i.e. under undesirable conditions, the skilled person would not have selected such conditions enabling the formation of froth in the process of document (1);

 - therefore it was not obvious for the skilled person to modify the process of document (1), which required the addition of methanol for forming an azeotrope and reacting with the C₅ products of the first catalytic reaction zone in order to produce TAME, by leaving out the etherification step and selecting conditions enabling, below the flooding point, the maintenance of froth throughout the catalyst bed.
- V. The Appellant requests that the decision under appeal be set aside and that a patent be granted on the basis of the main request submitted as first auxiliary request on 18 August 2004 as amended at the oral proceedings (replacing "through" with "throughout" on the last but one line of claim 1) or in the alternative on the basis of claim 1 filed during the oral proceedings as auxiliary request.

Reasons for the Decision

1. *Main Request*

1.1 Article 123(2) EPC

The Board is satisfied that the amended claims according to this request comply with the requirements of Article 123(2) EPC.

1.2 Articles 83 and 84 EPC

1.2.1 According to claim 1 hydrogen is fed to the distillation column reactor at a rate below that which would cause flooding of the column and the distillation column reactor is operated under conditions to maintain froth throughout the catalyst bed by control of the bottoms and/or overheads withdrawal rate.

As explained by the Appellant during oral proceedings the claimed process requires thus to operate below the flooding point but under conditions of "artificial" flooding enabling formation of froth in the catalyst bed though maintaining a countercurrent flow of vapour and liquid in the column.

This condition is achieved according to the wording of the claim by regulating the withdrawal rate of bottoms and/or overheads.

These features of claim 1 find support in the description of the application reading: "A froth level may be maintained throughout the catalyst bed by control of the bottoms and/or overheads withdrawal

rate..." (page 3, lines 28 to 30) and "The present invention can be carried out in a catalytic packed column which can be appreciated to contain a vapor phase ascending and some liquid phase as in any distillation. However since the liquid is held up within the column by artificial "flooding", it will be appreciated that there is an increased density over that when the liquid is simply descending because of what would be normal internal reflux" (page 8, lines 1 to 8).

- 1.2.2 The Appellant submitted during oral proceedings that it was common general knowledge of the skilled practitioner at the priority date of the present application that froth could be formed throughout a catalyst bed in a distillation column reactor only at or above the flooding point and not below the flooding point as required by claim 1 (see document (5), left column, passage below heading "**Flooding**", reading: "The flooding boundary is an absolute boundary that cannot be crossed under any circumstance. At the flooding point the countercurrent flow of vapour and liquid in the column breaks down. The liquid builds up in the packing and is pushed upward by the vapour. A froth layer can arise above the packing after the flood point has been reached."

Therefore, even though document (2) suggested that a turbulent layer of spray or bubbles ("Sprudelschicht"), favourable to the reactants exchange between gas and liquid, could be formed by operating between the loading point, i.e. the point at which liquid starts to be held up in a packed column, and the flooding point, this could not be interpreted as suggesting the

formation of a froth layer throughout the packing below the flooding point (see document (2), page 114, lines 6 to 14 below figure 6.4.5 and page 115, lines 6 to 13).

The Board has no reason to dispute the Appellant's interpretation of the teaching of documents (2) and (7) with the only exception that the appearance of a "Sprudelschicht", i.e. a layer of bubbles, does not appear to exclude the occasional occurrence of a bubble layer contained in a continuous liquid film, i.e. of froth, in some parts of the packing. However, the Board agrees that document (2) does not suggest operating under conditions in which froth is present throughout the packing as required by claim 1.

- 1.2.3 There is no dispute that a skilled person could monitor the formation of froth within the catalyst bed and could repeat the operative conditions of the specific examples of the present application. However, it must still be evaluated if the process step of forming a froth throughout the catalyst bed is sufficiently characterized and if a skilled person, when departing from the specific conditions of these examples, could carry out the invention as claimed without any undue burden of experimentation by following the teaching of the application and using his common general knowledge.

Claim 1 requires that a level of froth throughout the catalyst bed by operating below the flooding point (the result to be achieved) is obtained by regulating the bottoms and/or overheads withdrawal rate (process features).

However, the regulation of overheads and bottoms withdrawal rate is a process step which is commonly applied to any type of distillation for obtaining the desired rectification. Therefore, the Board finds that this process step does not sufficiently define the features essential for obtaining the desired result, i.e. froth throughout the catalyst bed.

Claim 1 thus does not contain all the features essential for properly defining the claimed invention and does not furnish any hint as to the operative limits which the skilled person can follow without an undue burden of experimentation in order to obtain froth below the flooding point when departing from the specific conditions of the examples.

Since this process feature of claim 1 is the only feature reported in the description as being essential for obtaining the desired froth level throughout the catalyst bed, the present application contains in the Board's view just an invitation to perform a research program for finding out the conditions essential for obtaining the desired result. This amounts in the Board's judgement to an undue burden of experimentation for the skilled person (see also T 782/01, unpublished in OJ EPO, points 2.5, 2.6 and 2.7 of the reasons for the decision).

Claim 1 thus does not comply with the requirements of Article 84 EPC and the present application does not comply with the requirements of Article 83 EPC.

The main request has thus to be dismissed.

2. Auxiliary Request

2.1 Article 123(2) EPC

Claim 1 according to the auxiliary request differs from claim 1 of the main request insofar as it requires that the overhead pressure in the distillation column reactor is between 96 and 1820 kPa (0 and 250 psig) and the temperature in the distillation reaction zone is between 38 and 149°C (100 to 300°F).

These are preferred features of the process of the invention as specified in the description (page 3, line 37 to page 4, line 3).

The Board is thus satisfied that the amended claims according to this request comply with the requirements of Article 123(2) EPC.

2.2 Articles 83 and 84 EPC

The Appellant explained during oral proceedings that the additional features mentioned hereinabove were useful for influencing the overheads and bottoms withdrawal rate. Moreover, even though they could not guarantee by themselves the formation of the froth level required by claim 1, they indicated operative conditions to be further selected for achieving the desired result.

The Board notes that these features indeed must be deemed to affect the withdrawal rate of bottoms and overheads as well as the reaction conditions within the column. Therefore these features further limit

implicitly the overheads and bottoms withdrawal rate which can be used according to the claimed process for obtaining a froth throughout the catalyst bed.

Moreover, even though these features cannot guarantee by themselves the formation of froth, they amount in combination with the other features of claim 1 to a clear teaching of the operative limits which the skilled person has to comply with when departing from the conditions used in the illustrative examples of the application for obtaining the desired result.

The Board thus cannot conclude, in the absence of any evidence, that it would not be possible for the skilled person to perform the invention or that it would be an undue burden of experimentation for the skilled person to find suitable operative conditions within the limits indicated in the claim in order to carry out the invention throughout the whole claimed scope.

Therefore, the Board concludes that Articles 84 and 83 EPC are in this case complied with.

2.3 Novelty

Since the claimed process requires that diolefins, hydrocarbon stream and hydrogen are the only components fed to the distillation column reactor whilst the process of document (1) requires the addition of methanol (column 2, lines 4 to 6 and 24 to 33) to the same column in order to obtain an ether and document (1) does not specify any operative condition which would result in froth being maintained throughout the catalyst bed though operating under the flooding point,

the Board concludes that the claimed subject-matter is novel over the process disclosed in document (1).

2.4 Inventive Step

2.4.1 The present application and, in particular, the subject-matter of claim 1 relates to a process for the removal of mercaptans and/or hydrogen sulfide from hydrocarbons streams (page 1, lines 8 to 10).

2.4.2 The process of document (1), aiming also *inter alia* at the removal of mercaptans from hydrocarbons streams (column 1, lines 15 to 16), represents the most suitable starting point for evaluating inventive step of the claimed subject-matter as also found in the appealed decision.

2.4.3 The Appellant defined the technical problem underlying the present invention in the light of the teaching of document (1) as the provision of an alternative, more effective and flexible process for the removal of mercaptans and/or hydrogen sulfide from hydrocarbon streams.

According to the description of the application the maintenance of froth throughout the catalyst bed improves the efficiency of the catalyst thereby allowing the reduction of the height of the catalyst used and a better separation of the reaction products (page 3, lines 28 to 32).

Moreover, the Appellant stated during oral proceedings that the absence of a further etherification reaction in the same distillation reactor column allowed the selection of a broader range of process conditions and increased the flexibility of the process.

The Board accepts the underlying technical problem defined by the Appellant and has no reason to doubt that this underlying technical problem has been solved by a process having all the features of claim 1.

- 2.4.4 The Board notes that it was common general knowledge of the skilled practitioner at the priority date of the present application that froth could be formed throughout a catalyst bed in a distillation column reactor only at or above the flooding point, i.e. under undesirable conditions, and not below the flooding point as required by claim 1 (see point 1.2.2 above).

The skilled person thus, applying his common general knowledge to the teaching of document (1), requiring the formation of a methanol/C₅ azeotrope, would have not operated under conditions leading to the formation of froth throughout the catalyst bed and possibly impairing the formation of the azeotrope.

Moreover, the prior art did not contain any suggestion that froth could be formed throughout the catalyst bed below the flooding point and that this could be useful for improving the efficiency of the catalyst in such a reaction process.

The Board concludes therefore that it was not obvious for the skilled person to modify the process of document (1) by leaving out the etherification step in the same distillation column reactor and applying conditions of "artificial" flooding as required by the process of claim 1.

The subject-matter of claim 1 of the auxiliary request involves thus an inventive step.

The dependent claims 2 to 11 also involve an inventive step for the same reasons.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The case is remitted to the first instance with the order to grant a patent with the following documents:
 - Claim 1 submitted as auxiliary request during oral proceedings.
 - Claims 2 to 11 filed with letter of 21 February 2002.
 - Description to be adapted if necessary.
 - Figure 1 as originally filed.

The Registrar:

The Chairman:

G. Rauh

P. Krasa