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Bezeichnung der Erfindung: Process for the removal of acid gases from gaseous
Title of invention: streams
Titre de l'invention :

Klassifikation / Classification / Classement : B01D 53/14

ENTSCHEIDUNG / DECISION

vom / of / du 3 March 1988

Anmelder / Applicant / Demandeur : Shell Internationale Research
Maatschappij B.V.

Patentinhaber / Proprietor of the patent /
Titulaire du brevet :

Einsprechender / Opponent / Opposant :

Stichwort / Headword / Référence :

EPÜ / EPC / CBE Article 56 EPC

Kennwort / Keyword / Mot clé : "Inventive step (No)"

Leitsatz / Headnote / Sommaire



Case Number : T 119/86 - 3.4.1

D E C I S I O N
of the Technical Board of Appeal 3.4.1
of 3 March 1988

Appellant : Shell Internationale Research
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Decision under appeal : Decision of Examining Division 031
of the European Patent Office
dated 14 November 1985 refusing
European patent application
No. 82 200 511.2 pursuant to
Article 97(1) EPC

Composition of the Board :

Chairman : K. Lederer

Members : J. Roscoe

O. Bossung

Summary of Facts and Submissions

- I. European patent application No. 82 200 511.2 (publication number 0 066 307) was refused by decision of the Examining Division.
- II. The reason given for the refusal was that the subject-matter of Claim 1 did not involve an inventive step within the meaning of Article 56 EPC having regard to the prior art documents

US-A-4 091 073 (D1) and
GB-A-952 555 (D2).
- III. The Applicant lodged an appeal against the decision.
- IV. In a communication of the Board, the Appellant's attention was drawn to the content of the following additions prior art documents:

DE-A-3 015 739 (D3) and
US-A-3 961 015 (D4).
- V. Oral proceedings were held at which the Appellant requested that the decision of the Examining Division be set aside and a European patent be granted on the basis of amended Claims 1 to 5 presented at the old proceedings, of which the sole independent claim reads as follows:

"1. Process for the removal of H₂S, CO₂ and COS from a sour gaseous steam containing H₂S, CO₂ and COS, comprising:

(a) contacting the sour gaseous steam with a lean CO₂- and COS-selective absorbent mixture containing a reactant oxidizing H₂S to sulphur,

- (b) separating a sweet gaseous steam from the absorbent mixture containing absorbed CO_2 , absorbed COS, solid sulphur and a reduced reactant, and
- (c) stripping the absorbent mixture separated in step (b) with formation of a gaseous steam comprising CO_2 and COS and a stripped absorbent mixture containing solid sulphur and a reduced reactant, wherein
- (d) the gaseous steam separated in step (c) and comprising CO_2 and COS is contacted in the presence of water with a catalyst causing hydrolysis of COS and a CO_2 - and H_2S -containing gaseous steam is separated from the catalyst, and wherein
- (e) in order to separate H_2S from the gaseous steam separated in step (d), said gaseous steam is contacted with a lean CO_2 -selective absorbent mixture containing a reactant oxidising H_2S to sulphur and a purified gas is separated from an aqueous mixture containing absorbed CO_2 , solid sulphur and reduced reactant."

The scope of this claim is substantially the same as that of Claim 1 of the set of claims on which the appealed decision was based.

VI. In support of his request, the Appellant essentially submitted that though the claimed sequence of process steps (a), (b), and (c) for removing H_2S and CO_2 from a sour gaseous steam was already known from document D1, the step (e) was essentially the same as the combination of these known steps (d) and (b),, and step (d) for hydrolysing COS was disclosed in document D2, other processes were available in the prior art for the same purpose, so that it would not have been obvious to select the specific process

steps disclosed in documents D1 and D2 and to combine them in the way defined in Claim 1.

Furthermore, the claimed process was particularly advantageous insofar as it allowed COS removal from a concentrated gaseous steam containing CO_2 and COS only, in contrast with the COS removal operations in accordance with document D3 or D4, which had to be performed on the whole gaseous steam to be purified. The claimed process also allowed the same absorbent/reactant mixture to be employed in step (a) to remove H_2S , COS and CO_2 from the sour gaseous steam to be purified and in step (e) to remove H_2S and CO_2 formed in the COS hydrolysis step.

In addition, document D1 was totally silent about COS and the possible effects of the described process on a sour gaseous steam containing COS in addition to H_2S and CO_2 , and the skilled person would not have expected that when subjecting such gaseous steam to this process, COS would also be absorbed by the absorbents used therein for extracting the CO_2 from the gaseous steam to be treated. In particular, there were no data available at the priority date of the present application on the solubility of COS in these absorbents, and from documents D3 and D4 it was even clear that several of them, such as N-methylpyrrolidone and tetraethylene glycol-dimethyl ether, which were cited also in the present application as constituting suitable COS absorbents, would not absorb COS. The claimed subject-matter was therefore a patentable application of the discovery that the known physical absorbents for CO_2 also absorbed COS.

Finally, documents D3 and D4 already disclosed processes for the removal of H_2S , CO_2 and COS from a sour gas steam while document D1 dealt with the removal of H_2S and CO_2 only, and, moreover, the process disclosed therein never

matured into a practical application. For these reasons, D1 could not be considered as a relevant prior art document from which the skilled person would be likely to start for solving the technical problem of removing all three components H_2S , CO_2 and COS from a sour gaseous steam.

Reasons for the Decision

1. The appeal is admissible.
2. There is no objection to the current set of claims as far
3. Novelty.
 - 3.1 Document D1 discloses a process for the removal of H_2S and CO_2 from a sour gaseous steam containing H_2S and CO_2 , comprising the steps of:
 - (a) contacting the sour gaseous steam with a lean CO_2 -selective absorbent mixture containing a reactant oxidizing H_2S to sulphur,
 - (b) separating a sweet gaseous steam from the absorbent mixture containing absorbed CO_2 , solid sulphur and a reduced reactant, and
 - (c) stripping the absorbent mixture separated in step (b) with formation of a gaseous steam comprising CO_2 and a stripped absorbent mixture containing solid sulphur and the reactant (Claim 1 and Figure).

This process is distinguished from the subject-matter of present Claim 1 in that COS is not mentioned in steps (a) to (c), in that the reactant contained in the stripped absorbent mixture is not in the reduced state as defined in

the claim, but is already oxidized (since stripping of the absorbent mixture is performed by means of an oxygen-containing gas), and in that it does not provide for COS hydrolysis and separation of the H₂S formed by this hydrolysis, as defined in steps (d) and (e) of the claim.

- 3.2. Document D2 discloses a process for hydrolysing COS in which a gaseous steam comprising COS is contacted in the presence of water in the form of steam with a catalyst causing hydrolysis of COS, and a CO₂- and H₂S-containing gaseous steam is separated from the catalyst, as defined in step (d) of Claim 1 (page 1, lines 32 to 40 and Claim 13).

This known process is distinguished from the subject-matter of Claim 1 in that it does not comprise the steps (a) to (c) and (e) as defined in the claim for removal of H₂S and CO₂ originally contained in the gaseous steam or produced by the COS hydrolysis. Document D2 merely states in this respect that the H₂S and CO₂ produced by the latter reaction are removed by scrubbing or other conventional means (page 2, lines 24 to 29):

- 3.3. Documents D3 and D4 both disclose a process for the removal of H₂S, CO₂ and COS from a sour gaseous steam containing them. The sour gaseous steam is first contacted with an absorbent mixture for removing H₂S and CO₂, and the remaining gaseous steam, which is thus freed of H₂S and CO₂ but still comprises COS, is contacted in the presence of water with an alkanolamine mixture which acts as a catalyst for causing hydrolysis of COS. Finally, the resulting CO₂- and H₂S-containing gaseous steam is contacted again with an absorbent mixture for removing H₂S and CO₂ (document D3, page 15, last paragraph and Claim 10; document D4, Claim 1).

Thus, the process known from document D3 or D4, comprises COS hydrolysis as defined in step (d) of present Claim 1, but said COS hydrolysis is effected on the main gaseous steam to be treated, after H₂S and CO₂ removal, not on a steam consisting of gases previously separated from such main gaseous steam. In addition, the process of document D3 or D4 neither provides for any separation from each other of the H₂S and CO₂, which are simultaneously extracted from the main gaseous steam or from said steam after COS hydrolysis, nor for oxidation of the H₂S into solid sulphur.

3.4 The remaining cited documents do not come closer to the subject-matter of Claim 1.

3.5 For these reasons the subject-matter of Claim 1 is novel within the meaning of Article 54 EPC.

4. Inventive step.

4.1 The nearest prior art is constituted, in the Board's view, by the process disclosed in detail in document D1, since document D1 is the sole prior art document disclosing the use of a CO₂-selective absorbent mixture containing a reactant oxidising H₂S to sulphur, for simultaneously removing CO₂ and H₂S from a gaseous steam and converting H₂S to solid sulphur. In addition, the mixture used in the specific example described in D1 comprises N-methylpyrrolidone as an absorbent and an aqueous solution of the Fe(III) chelate of N-(2-hydroxyethyl)ethylene diamine triacetic acid as a reactant (column 4, lines 42-48), which are respectively referred to in the description of the present application as constituting a suitable CO₂- and COS-selective absorbent (page 11, line 18) and reactant (page 14, lines 21 and 22, or page 8, lines 14 and 15 in conjunction with lines 9 and 10).

- 4.2. Thus, the process known from this document can immediately be seen to remove two of the three gases required to be removed, and it achieves this by the steps employed in the process claimed in Claim 1 of the present application for the same purpose. The document however does not indicate what effect the system might have on any COS present in the sour gas. The process of Claim 1 on the other hand removes any such COS and eliminates it.

Therefore the objectively assessed technical problem in relation to the sour gas steam purification process known from document D1 is to modify the process to enable it to remove from the sour gas steam not only H₂S and CO₂, but also any COS it may contain.

This technical problem is solved in accordance with claim 1 essentially by contacting the gaseous steam (comprising CO₂ and COS) stripped from the absorbent mixture, in the presence of water, with a catalyst causing hydrolysis of COS to CO₂ and H₂S (step (d)), and removing CO₂ and H₂S from the resulting gaseous steam to be purified (step (e)).

- 4.3. No contribution to inventive step can be seen in recognising the problem having regard to the fact that COS is known to be a constituent of many natural and industrial gases, such as watergas, coalgas and gases obtained from oil refining, and is commonly regarded as a most unwanted impurity (D2, page 1, lines 11 to 18; D3, page 8, first paragraph and the paragraph bridging pages 8 and 9; D4, column 2, lines 39 to 41).
- 4.4. The skilled person faced with the problem would first conduct routine tests using available gas analysis equipment to find out what happens to any COS in the sour

input gas. Having regard to above-mentioned similarity of the absorbent mixtures used in D1, and those proposed in the present application for absorbing COS, it is clear that such tests would reveal that the COS will not emerge in the "sweet" gas stream, but will leave the absorbent column with the absorbed CO₂, the converted Fe(II) chelate, and the elemental sulphur.

Therefore, starting from the process described in document D1 and having determined that the COS content is already removed from the sour gas to be treated by absorption in the absorbent mixture in the absorption column 2 the skilled person knows that it must then be stripped therefrom together with the CO₂ in the regeneration zone 5, and would seek an appropriate method for removing the COS from the gaseous stream output from the regeneration zone.

By document D2, he would be taught that COS may be removed from a gaseous mixture containing it by contacting the latter in presence of water in the form of steam with a catalyst causing hydrolysis of COS and separating a CO₂ and H₂S containing gaseous stream from the catalyst.

Having applied this known treatment to the CO₂- and COS-containing gaseous stream emerging from the regeneration zone, he would then obtain a gaseous stream containing CO₂ and H₂S but no COS. Since these are gases which were removed from the original sour gas in D1 in steps (a) and (b), the decision to use the mixture used in those steps to remove them also from the stream emerging from the regeneration zone would not in the Board's view, involve an inventive step since it has the obvious advantage of reducing the inventory of chemicals required. A similar employment of a common chemical solution for essentially the same purpose in two different stages of a multistage

gas purification process has previously been proposed in D3.

The resulting process for removing H_2S , CO_2 and COS from a sour gaseous steam still differs from the subject-matter of Claim 1 in that the aqueous mixture from which a purified gas is separated after oxidation of H_2S into solid sulphur contains, in addition to absorbed CO_2 and solid sulphur, a reactant which is not reduced as defined in the claim but already oxidised, an oxygen-containing gas being used for simultaneously separating the purified gas and regenerating the H_2S oxidizing reactant. Immediate regeneration of the reactant during the separation step, however, is clearly not essential to the working of the gas purification method disclosed in D1, and regeneration of the reactant, which is performed for reasons of economy may obviously be performed at a later stage of the process as well, or even be dispensed with if the absorbent mixture is to be discarded, which uneconomical alternative is contemplated also in the description of the present application as filed (page 4, lines 10-11). Therefore the Board is unable to recognise, and the Appellant has not sought to convince it, that there is anything inventive in dispensing with the feature of regenerating the reactant during stripping of the absorbent mixture.

- 4.5. The arguments submitted by the Appellant in support of his request could not convince the Board of the patentability of the claimed subject-matter.

In particular, while it is not denied that other equivalent methods were available for removing H_2S , CO_2 and/or COS from a gaseous steam than those used in accordance with Claim 1, a specific combination of known features must a priori be considered as being obvious, unless it is demonstrated, for example, that such combination leads to

an unexpected advantage or that there has been some technical prejudice against its practicability. The Appellant, however, failed to establish the existence of such unexpected advantage or technical prejudice.

In particular, the first alleged advantage of the process in accordance with Claim 1 invoked by the Appellant that the latter allowed removal of COS from a concentrated gaseous steam essentially comprising CO₂ and COS only, which could therefore be performed more efficiently than the purifying operations taught for instance by documents D3 and D4 does not result from that process since feature (c) does not require the stripping to be performed in any particular way and therefore does not ensure that the gaseous steam exiting the stripper contains only, or even primarily CO₂ and COS. Moreover it has not been established to the Board's satisfaction that a higher concentration of COS in the stripper effluent automatically leads to a more efficient removal of COS from the system. The other advantage invoked, that the same absorbent/reactant mixture could be used for H₂S and CO₂ removal throughout the process as clearly foreseeable by the skilled person and would therefore rather have encouraged him to consider the claimed combination of process steps.

The statements in documents D3 and D4 that the process disclosed therein could also be used in the presence of a physical absorbent such as, among a number of other examples, propyleneglycoldimethyl ether (D3, page 14, third paragraph) or N-methyl-2-pyrrolidone (D4, column 4, lines 39 to 42) might indeed suggest that these specific compositions would not absorb COS (otherwise no further COS hydrolysis would be necessary or possible in the gaseous steam previously contacted with them) and that, accordingly, the process disclosed in document when using

the same or similar compositions as CO₂-selective absorbents (column 4, lines 25 to 30) also could not lead to COS-selective absorption.

However the Board's reasoning in item 4.3. above does not rely on the skilled man already having been aware that all, or any of, the absorbents proposed in D1 does in fact absorb COS but rather on his ability to determine by experiment whether it is or whether it emerges from absorber 2 with the CO₂ and H₂S-free gas. If the information on which the present application is based is correct, which the Board has no reason to doubt, he would have found or that it was absorbed.

The Appellant's argument that the process disclosed in document D1 has never been put into practice and that the skilled person therefore would not have considered it as a proper basis for his search for an effective process for removing H₂S, CO₂ and COS from sour gaseous steams cannot be accepted either, for the process may also have been used without the Appellant's knowledge and, furthermore, the relevant prior art is not restricted to those devices or processes which have found a practical application, but encompasses any teaching available to the public at the filing date.

Finally, the fact that document D1 does not explicitly indicate that the process disclosed therein may remove H₂S, CO₂ and COS from a sour gaseous steam does not mean that D1 is not suitable starting point for the skilled person, for, as indicated above, obvious considerations would lead the skilled person to investigate the effect of the known process from D1 on a gaseous steam containing COS as well.

4.6 Thus, the subject-matter of Claim 1 is not considered to involve an inventive step within the meaning of Article 56 EPC and the claim is, accordingly, not allowable under Article 52(1) EPC.

4.7 Since this claim is an essential part of the Appellant's sole request it follows that the request as a whole cannot be allowed.

Order

For these reasons, it is decided that:

The appeal is dismissed.

The Registrar:

The Chairman:

F. Klein

K. Lederer