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Bezeichnung der Erfindung: Removal of sulfur dioxide from gases

Title of invention:

Titre de l'invention :

Klassifikation / Classification / Classement : B 01 D 53/34

ENTSCHEIDUNG / DECISION

vom/of/du 21 September 1988

Anmelder / Applicant / Demandeur : PFIZER INC.

Patentinhaber / Proprietor of the patent /

Titulaire du brevet :

Einsprechender / Opponent / Opposant :

Stichwort / Headword / Référence :

EPÜ/EPC/CBE Article 56 EPC

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

Leitsatz / Headnote / Sommaire

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Office

Office européen
des brevets

Beschwerdekammern

Boards of Appeal

Chambres de recours

Case Number : T 267/85 - 3.4.1



D E C I S I O N
of the Technical Board of Appeal 3.4.1
of 21 September 1988

Appellant : PFIZER INC.
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Representative : Wood, David John
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Decision under appeal : Decision of Examining Division 031 of the
European Patent Office dated 3 July 1985
refusing European patent application
No. 82 302 305.6 pursuant to Article
97(1) EPC

Composition of the Board :

Chairman : K. Lederer

Members : J. Roscoe

R. Schulte

Summary of Facts and Submissions

- I. European patent application No. 82 302 305.6 (publication number 0 093 807) was refused by decision of the Examining Division.
- II. The decision was based on the set of claims as filed, amended according to Applicant's letter filed 18 February 1985.

The reason given for the refusal was that independent Claim 1 lacked an inventive step within the meaning of Article 56 EPC, having regard to the following prior art documents:

US-A-3 757 488 (D1); and
US-A-4 277 451 (D2).

The Examining Division held in particular that the subject-matter of Claim 1 differed from the disclosure of document D1 in that it additionally required the process to be carried out at a temperature of from 15°C to 80°C, and in that the aqueous solution was stripped of its absorbed sulfur dioxide by heating and recovered for re-use in the process. The definition of the claimed range for the operating temperatures, however, was considered to be within the normal competence of the skilled man, while regeneration of used scrubbing solution for re-use was fundamental to the economics of the gas scrubbing art that used such solutions, thermal stripping further being one of the main modes of achieving such regeneration. Document D2 also disclosed a process using an absorbent solution for the desulfurisation of similar exhaust gases, wherein the gas is contacted with the solution at a temperature

within the claimed range, and the absorbent is subsequently thermally stripped for re-use.

III. The Appellant lodged an appeal against the decision.

IV. The Appellant requests that the decision be set aside and a patent be granted on the basis of Claims 1 to 10 as filed on 2 April 1988 of which Claim 1, the sole independent claim, reads as follows:

"1. A process for the desulfurisation of gas containing from 100 ppm to 30 volume percent sulfur dioxide, which comprises contacting the gas at a temperature of from 15 to 80°C with an aqueous solution at a pH of from 3 to 9 of tripotassium citrate at a concentration of from 0.1 molar to saturation; separating the gas from the solution; heating the solution to strip sulfur dioxide therefrom; and recovering the stripped solution for re-use in the desulfurisation process."

Claim 1 is distinguished from Claim 1 on which the decision was based in that the aqueous solution which was originally defined in the claim as containing mono-, di- or tri-potassium citrate, is specified to be a solution of tripotassium citrate.

V. In support of the allowability of his request, the Appellant submits that the invention lies in the unexpected and surprising discovery that the regenerative type sulfur dioxide removal process is dramatically more effective and can be operated at a much lower energy requirement when the absorbing solution is a tripotassium citrate solution, as evidenced for instance by the data presented in the original description and the results of comparative tests included in a declaration by one of the co-inventors filed on 7 October 1986.

Such an advantage could not have been foreseen from the cited prior art, nor could the teaching of document D1 that aqueous solutions of citric acid and potassium citrate are particularly useful in a sulfur dioxide absorption process without regeneration have given any hint to the skilled person that such solutions would be especially suitable also in a recovery and regeneration process. In particular, document D2, filed after document D1 was published, and later work on such processes select different solutions such as solutions of potassium salts of tartaric acid (D2) or of sodium citrate instead of the claimed solution, and document D2 further stresses that many absorbent solutions are not useful in recovery and regeneration processes because the complex transformations which can occur chemically alter the absorbent so that it can no longer be regenerated and recycled after stripping of the sulfur dioxide.

There further existed some prejudice in the art against the use of potassium citrate in a recovery and regeneration process since, in particular, potassium citrate is approximately 20% more expensive than sodium citrate. In addition, sulfur dioxide extraction from solutions containing potassium ions was known to be less effective than from solutions containing sodium ions as reported in the references

- G.T. Rochelle, "Process Synthesis and Innovation in Flue Gas desulfurisation", EPRI, FP-463-SR, Special Report, July 1977, pages 4-1 to 5-8; and
- A.V. Slack and G.A. Hollinden, "Sulfur Dioxide Removal from Waste Gases", 2nd Edition, Noyes Data Corporation, Park Ridge, New Jersey, 1975, pages 222-224.

The Appellant also submitted that the US Board of Appeal allowed the corresponding US application on the basis that the showing of unexpected results overcome any prima facie case of obviousness.

Reasons for the Decision

1. The appeal is admissible.
2. **Novelty**
 - 2.1 Document D1 discloses a process for selectively removing sulfur dioxide from flue gases by contacting the gases with an aqueous solution containing at least one alkali metal salt of an organic acid. The pH of the solution is between 4.0 and 6.0. Preferably, the alkali metal salt of an organic acid is potassium citrate, of which the concentration may be 1.0 mole in one liter of water, and the gas may originally contain 300 parts per million sulfur dioxide (Claims 1, 3 and 4. Example No. 1). The indication in the description (column 2, lines 63 to 67) that potassium citrate "may" reduce in the solution to potassium dihydrogen citrate (monopotassium citrate) or potassium monohydrogen citrate (dipotassium citrate) shows that the term potassium citrate, as used in Example No.1, refers to non-reduced form (tripotassium citrate).

This known method is used for separating sulfur dioxide from hydrogen sulfide and other gases (column 1, lines 12 to 16), which application, albeit not explicitly contemplated in the present description, is within the scope of the claims.

Thus, the subject-matter of Claim 1 is distinguished from the process disclosed in document D1 in that the contact

temperature of the gases with the aqueous solution, on which document D1 is silent, is specified to be in the range of from 15°C to 80°C and in that the solution is subsequently heated to strip sulfur dioxide therefrom and recovered for re-use in the desulfurisation process.

- 2.2 Document D2 discloses a process for the desulfurisation of gas containing sulfur dioxide in an unspecified amount, which comprises contacting the gas at a temperature of from 40 to 80°C with an aqueous solution at a pH of from 4.5 to 6.5 of a potassium salt of an organic acid (potassium tartrate) at a concentration of from 0.1 molar to saturation; separating the gas from the solution; heating the solution to strip sulfur dioxide therefrom; and recovering the stripped solution for re-use in the desulfurisation process (Claims 1, 3 and 4).

Thus, the subject-matter of Claim 1 is distinguished from the process in document D2 in that the sulfur dioxide concentration in the gas to be desulfurised is specified to be in the range from 300 ppm to 30 volume percent, and in that the potassium salt is potassium citrate instead of potassium tartrate.

- 2.3 Document US-A-4 140 751 (D3) discloses a process for the desulfurisation of gas containing sulfur dioxide in a non specified amount, which comprises contacting the gas at a temperature of from 120 to 140°F (48 to 60°C) with an aqueous solution of a citrate of an alkali metal citrate (sodium citrate) obtained by blending a 0.5 molar sodium citrate solution with 0.5 molar citric acid to a pH of 4; separating the gas from the solution; heating the solution to strip sulfur dioxide therefrom; and recovering the stripped solution for re-use in the desulfurisation process (Claim 1, column 5, lines 30 to 34 and 45 to 48).

Thus the subject-matter of Claim 1 is distinguished from the process known from document D3 in that the sulfur dioxide content of the gas to be treated is specified to be in the range from 100 ppm to 30 volume percent, and in that potassium citrate is substituted for the sodium citrate proposed in document D3.

- 2.4 The remaining documents cited in the European Search Report or referred to in the application documents are not more relevant to the subject-matter of Claim 1.
- 2.5 For these reasons, the subject-matter of Claim 1 is considered to be novel within the meaning of Article 54 EPC.

3. Inventive Step

- 3.1 Document D1 fails to specify the temperature range at which the gas to be treated is contacted with the absorbent solution. The range from 15 to 80°C set out in Claim 1 however encompasses the temperature values normally encountered with flue gases and used also in the similar absorption steps of the processes known from documents D2 (40 to 80°C) or D3 (48 to 60°C) and is a range which the skilled man wishing to employ the method disclosed in D1 could determine by routine experiment as that in which optimum absorption occurs. Moreover the Appellant has not argued that the temperature range defined in Claim 1 provides any unforeseeable effect either alone or in combination with other features of the Claim.

For these reasons, this feature is considered to be self evident and it need not therefore be considered further when assessing the patentability of the claimed subject-matter.

- 3.2 Thus, starting from the nearest prior art as disclosed in document D1, and considering the remaining distinguishing features as set out in paragraph 2.1 above the objectively assessed technical problem to which the invention defined in Claim 1 affords a solution is to recover the absorbed sulfur dioxide from the potassium citrate solution so as to enable it to be re-used.
- 3.3 No contribution to inventive step can be seen in posing of the problem since it is common practice in sulfur dioxide and other absorption processes to remove the absorbed gas and then recycle the absorbent solution, as evidenced for instance by documents D2 and D3. The Appellant did not put forward any argument in favour of an inventive step being involved in recognition of the above defined technical problem either.
- 3.4 In the processes of both documents D2 and D3 recovery of the absorbed sulfur dioxide from a solution containing an alkali metal salt of a weak organic acid is achieved by heating the solution, e.g. by steam stripping, and the absorbent is subsequently re-used in the desulfurisation stage.

The skilled person confronted with the above defined technical problem would have no reasonable ground to doubt that such thermal recovery of sulfur dioxide and recycling of the stripped solution may operate as well in connection with an absorbent solution comprising potassium citrate, the more so since neither the claims nor the description of document D3 put any emphasis on the type of citrate to be selected for use in the process, sodium citrate being referred to once only in the description (column 5, lines 48) as a component of the citrate solution, without further comments.

The skilled person would therefore hardly fail to implement such additional steps also in the process known from document D1, and thus immediately arrive at the subject-matter of Claim 1.

- 3.5 The Appellant's submissions could not convince the Board of the patentability of the claimed subject-matter.

In particular, the arguments relying on the advantages of using potassium citrate instead of sodium citrate in terms of energy requirement and on the cost comparison between both citrates miss the point in so far as the Board's (and the Examining Division's) reasoning starts from document D1 as the nearest prior art, which is already committed to the use of potassium citrate. Subsequent discovery of unexpected advantages of the latter citrate cannot therefore by itself justify the grant of a patent.

Having regard to the alleged existence of a prejudice in the art against the capability of the potassium ion to permit sulfur dioxide regeneration, it is noticed that the passages from the Rochelle Report quoted on page 3 of Appellant's response dated 2 October 1986 and from the Slack and Hollinden reference referred to in paragraph V above both relate to processes in which the absorbent solution comprises sodium or potassium sulfite, and is normally regenerated by evaporative crystallization of the sulfite. The reported difficulties with potassium are specially linked to the formation of pyrsulfite $K_2S_2O_5$ in these processes, and it is not seen why the skilled person would expect similar problems when using citrate solutions, from which sulfur dioxide is stripped without phase separation. In this respect, in a subsequent portion of the Rochelle Report directed to the stripping of

solutions comprising weak acids without phase separation (page 4-38), the potassium citrate system is explicitly stated to meet the criteria which characterise an ideal buffer system.

Also the fact that a patent has been granted in the US for the present invention is unpersuasive, particularly since it has not been shown that the US patent office advanced but ultimately withdrew the arguments on inventive step set out above.

- 3.6 For these reasons, the subject-matter of Claim 1 is not considered to involve an inventive step within the meaning of Article 56 EPC.
4. Claim 1, is therefore not allowable under Article 52 EPC and Claims 2 to 10 are not allowable either by virtue of their dependence on it.

Accordingly, Appellant's request cannot be allowed.

Order

For these reasons, it is decided that:

The appeal is dismissed.

The Registrar

The Chairman

F. Klein

K. Lederer