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**Datasheet for the decision
of 30 July 2015**

Case Number: T 1648/12 - 3.3.06

Application Number: 06705228.2

Publication Number: 1850947

IPC: B01D53/14

Language of the proceedings: EN

Title of invention:

AN IMPROVED CO₂ ABSORPTION SOLUTION

Applicant:

CO₂ Solutions Inc.

Headword:

CO₂ absorption/CO₂ Solutions Inc.

Relevant legal provisions:

EPC Art. 52(1), 56, 123(2)

RPBA Art. 12(4)

Keyword:

Claim requests corresponding requests not admitted by Examining Division -

admitted (no) (Main and Auxiliary Requests I and II)

Amendments - added subject-matter (yes) Auxiliary Request III

Inventive step -

(no) obvious solution (Auxiliary Requests IV and V)

Decisions cited:

G 0007/93

Catchword:



**Beschwerdekammern
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Chambres de recours**

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Case Number: T 1648/12 - 3.3.06

D E C I S I O N
of Technical Board of Appeal 3.3.06
of 30 July 2015

Appellant: CO2 Solutions Inc.
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Decision under appeal: **Decision of the Examining Division of the European Patent Office posted on 16 December 2011 refusing European patent application No. 06705228.2 pursuant to Article 97(2) EPC.**

Composition of the Board:

Chairman B. Czech
Members: G. Santavicca
C. Vallet

Summary of Facts and Submissions

- I. The appeal lies from the decision of the Examining Division to refuse European patent application n° 06705228.2.
- II. In the decision under appeal, the Examining Division *inter alia* came to the conclusion that the "*formulation for the absorption of CO₂*" according to Claim 1 of each of the then pending requests lacked an inventive step in the light of a combination of documents
- D1: WO 2004/056455, as the closest prior art, and
- D5: EP 0 558 019 A.
- III. With its statement setting out the grounds of appeal dated 25 April 2012, the Appellant (Applicant) submitted six sets of amended claims as Main Request and Auxiliary Requests I to V, as well as experimental results.
- IV. In its communication pursuant to Article 15(1) RPBA annexed to the summons to oral proceedings, the Board cited the following further documents:
- D6: US 3,622,267 (mentioned in D5),
- D7: US 4,814,104 (mentioned in the application) and
- D8: Blauwhoff, Versteeg and van Swaij, *A study on the reaction between CO₂ and alkanolamines in aqueous solutions*, Chemical Engineering Science, Vol.39, No.2, 1984, pp.207-225.

In this communication, the Board *inter alia* questioned

- under Article 12(4) RPBA, the admissibility of the Appellant's claim requests, in particular of the Main Request and of Auxiliary Requests I and II which were identical to claim requests not admitted by the Examining Division;
- under Article 123(2) EPC, the allowability of the claims of Auxiliary Request III having regard to the feature "*tertiary alkanolamines*"; as well as,
- whether the subject-matter claimed (all requests) involved an inventive step in the light of D1 (as the closest prior art), taken alone or in combination with any of D6 or D7. In this connection it referred also to D8.

- V. In its reply, dated 7 July 2015, the Appellant did not further address the substance of the case but merely announced that it would not attend the oral proceedings.

Up to the date of the oral proceedings, no further arguments were submitted in response to the detailed comments and objections expressed in the Board's communication.

- VI. Oral proceedings took place on 30 July 2015 in the announced absence of the duly summoned Appellant, pursuant to Rule 115(2) EPC.

- VII. Claim 1 according to Auxiliary Requests III, IV and V respectively reads as follows:

Auxiliary Request III

"1. Use of carbonic anhydrase as an activator to increase CO₂ absorption rate in an aqueous solution used for CO₂ absorption, wherein the aqueous solution

contains at least one CO₂ absorption compound selected from the group consisting of alkanolamines, said alkanolamines being selected from the group consisting of tertiary alkanolamines, and the carbonic anhydrase being directly added into the absorption solution."

Auxiliary request IV

Compared to Claim 1 according to Auxiliary Request III, Claim 1 according to Auxiliary Request IV includes the additional feature "*said tertiary alkanolamine being selected from the group consisting of triethanolamine and N-methyldiethanolamine (MDEA)*".

Auxiliary Request V

Compared to Claim 1 according to Auxiliary Request III, Claim 1 according to Auxiliary Request V includes the additional feature "*said tertiary alkanolamine being N-methyldiethanolamine (MDEA)*".

VIII. The arguments of the Appellant of relevance for the present decision can be summarised as follows:

*Allowability of the amendments
(Auxiliary Requests III to V)*

The use defined in Claim 1 according to Auxiliary Request III found basis in Claims 8 and 9 of the application as filed. In particular, the limitation of the carbon dioxide absorption compound to, more specifically, "**tertiary** alkanolamines" (emphasis added) was generally disclosed in the application as filed (page 3, lines 19 to 30). Hence, the application provided a basis for claiming also the use of other tertiary alkanolamine compounds than the ones expressly

mentioned in the application as filed, including triethanolamine (TEA) and N-methyldiethanolamine (MDEA). The application as filed also disclosed that any CO₂ absorption compound could be used in combination with the carbonic anhydrase, as well as the limiting feature "*the carbonic anhydrase being directly added into the absorption solution*" were disclosed in the application as filed. Therefore, the amended claims complied with Article 123(2) EPC.

Inventive step (Auxiliary Requests IV and V)

D1 described a method for removing carbon dioxide from a gas comprising a step of contacting carbon dioxide with a solution comprising a primary, sterically hindered alkanolamine (2-amino-2-hydroxymethyl-1,3-propanediol, *vulgo* "Tris") and carbonic anhydrase (Example on page 11, line 8), the carbonic anhydrase being "used free within the Tris solution", i.e. not being immobilised. Since the primary alkanolamine Tris was structurally close to tertiary alkanolamines, and since the carbonic anhydrase was added directly into the absorption solution, as defined in Claim 1 at issue, D1 disclosed the closest prior art. The use according to Claim 1 at issue differed from the disclosure of D1 in that a tertiary alkanolamine such as TEA or MDEA was used in the CO₂ absorption solution according to Claim 1, whereas D1 only disclosed the use of the primary alkanolamine Tris.

The technical problem solved by the invention, as deducible from the application as filed, consisted in providing an alternative use of carbonic anhydrase solutions for absorption of CO₂, which, by means of the use of tertiary alkanolamines and the addition of the carbonic anhydrase directly into the absorption

solution, provided unexpected advantages, such as a synergistic enhancement of the CO₂ absorption properties of the tertiary alkanolamines. The achievement of these unexpected advantages was shown by the comparative evidence submitted with the statement setting out the grounds of appeal.

These unexpected advantages could not be inferred from D1, taken alone or in combination with D5. D5 disclosed tertiary alkanolamines, including MDEA, as CO₂ absorbing compounds, but neither mentioned nor suggested the combination of the tertiary alkanolamine MDEA with carbonic anhydrase.

Thus, it was not obvious to a person skilled in the art, even in the light of D1 and D5, that using an aqueous solution of a tertiary alkanolamine, in particular MDEA, in combination with carbonic anhydrase added to the solution, was a particularly effective way of absorbing CO₂, let alone that the combination would exhibit synergistic advantages.

Reasons for the Decision

Main Request and Auxiliary Requests I and II - Admissibility

1. The Main Request and Auxiliary Requests I and II filed together with the statement of grounds of appeal correspond to Auxiliary Requests III to V filed during the oral proceedings before the Examining Division and not admitted into the proceedings by the Examining Division pursuant to Rule 137(3) EPC (Decision under appeal, Reasons, 10ff.). This was not disputed by the Appellant.

- 1.1 In accordance with the principles developed in G 7/93 (OJ EPO 1994, 775, Reasons, 2.6) the discretionary decision of the Examining Division not to admit said claim requests may be overruled provided the Examining Division failed to exercise its discretion in an appropriate manner.

- 1.2 In the present case, the Examining decision refused to admit these claim requests because they were only filed during the oral proceedings, because the method claim thereof contravened the requirements of Article 123(2) EPC and because these claim requests would *prima facie* not overcome the inventive step objection raised previously.
 - 1.2.1 In accordance with the indications already given in the Board's communication (Point 7.1), the Board holds that the Examining Division exercised its discretion in accordance with the right principles and in a reasonable way, and thus without exceeding the proper limits of its discretionary remit.
 - 1.2.2 Neither the statement setting out the grounds of appeal, nor the Appellant's reply to the Board's communication contains any statement or argument regarding possible flaws in the discretionary decision taken by the Examining Division.

- 1.3 Therefore, the Board saw no reason for overruling the Examining Division's discretionary decision in this respect and decided not to admit the new Main Request and Auxiliary Requests I and II into the proceedings (Article 12(4) RPBA).

Auxiliary requests III to V - Admissibility

2. These amended claim requests were submitted for the first time together with the statement of grounds of grounds of appeal.

2.1 Nature of the amendments

Use Claim 1 according to Auxiliary request III is identical to the combination of use Claims 8 and 9, first alternative, of the Main Request that had been pending before the first instance. Claims 2 and 3 are identical to Claims 10 and 11 of said Main Request.

Claim 1 of Auxiliary Request IV is identical to the combination of Claims 8, 9 (first alternative) and 10 of said main request. Claim 2 thereof is identical to Claim 11 of said Main Request.

Claim 1 of Auxiliary Request V is identical to the combination of Claims 8, 9 (first alternative) and 11 of said Main Request.

2.2 The claims according to the Auxiliary Requests III to V at issue consist of use claims only, they no longer comprise claims directed to compositions or methods. Therefore, their filing contributes to the convergence of the debate, without raising new and/or complex issues not dealt with in the first instance proceedings. They concern subject-matter which was dealt with in the decision under appeal, albeit in an *obiter dictum* (Reasons, 11.2).

2.3 Therefore, despite their late filing, the Board decided to admit Auxiliary Requests III to V into the proceedings (Article 12 RPBA).

Auxiliary Request III - Requirement of Article 123(2) EPC

3. In accordance with the Board's provisional opinion as expressed in its communication (point 8.1.1 to 8.1.3) the subject-matter claimed does not find sufficient basis in the application as filed:
4. The expression "*tertiary alkanolamines*", contained in Claim 1 of Auxiliary Request III, is mentioned only in the context of the acknowledgement, in the description of the application as filed (page 1, lines 19-20 and 23-25; page 3, line 19) of the background of the invention.
 - 4.1 In the description of the actual invention, starting at page 6, "*alkanolamines*" are generally mentioned (page 7, line 2). It is also stated that "*By amines ... is meant any optionally substituted aliphatic amines*" (page 7, lines 3 and 4).
 - 4.2 However, "*tertiary alkanolamines*" are not expressly mentioned as a class of compounds in this part of the description or in the claims of the application as filed, apart from some specific examples thereof, including methyldiethanolamine (MDEA) and triethanolamine (TEA), mentioned e.g. on page 7, lines 12 and 14.
 - 4.3 Insofar as Claim 1 according to Auxiliary Request III refers to the (generic) class of "*tertiary alkanolamines*", and thus encompasses the use of other tertiary alkanolamines (such as DMMEA or DEMEA specifically invoked by the Appellant on page 2, second paragraph, of its statement of grounds) than the ones specifically mentioned in the application as filed as absorbents for the purposes of the invention, i.e. MDEA

and TEA, its subject-matter represents an intermediate generalization, unsupported by the original disclosure.

4.4 The Appellant did not file any counter arguments. Hence, the Board concludes that the subject-matter as defined by Claim 1 at issue extends beyond the content of the application as filed.

4.5 Therefore, Claim 1 of Auxiliary Request III does not comply with the requirements of Article 123(2) EPC.

4.6 Consequently, Auxiliary Request III is not allowable.

Auxiliary Requests IV and V - Inventive step

The invention

5. The invention (see claim 1; both requests) relates to the use of carbonic anhydrase as an activator for increasing the carbon dioxide absorption rate in aqueous solutions for absorbing carbon dioxide, in particular in the context of gas purification (page 1, lines 3-8, of the application as filed) .

The closest prior art

6. Document D1

6.1 In view of the similarity of the objectives addressed, respectively, in the application at issue and in document D1, and of the similarity of the chemical compositions used for CO₂ absorption, the Board accepts that D1 discloses the closest prior art. More particularly, the method illustrated in the example of D1 (pages 11 and 12) is a most appropriate starting point for the assessment of inventive step.

6.2 Indeed, said example describes a method for removing carbon dioxide from a gas in a spray absorber by absorption thereof in a solution of the primary alkanolamine 2-amino-2-hydroxymethyl-1,3-propanediol, *vulgo* "Tris", said solution also comprising carbonic anhydrase in "free" form (i.e. not bound to a substrate). The solution is sprayed in a closed loop operation until saturation of the Tris solution with carbon dioxide.

6.3 In D1 (bottom of page 8; equation (1)), carbonic anhydrase enzyme is also referred to as "biocatalyst", which catalyses the chemical reaction $\text{CO}_2 + \text{H}_2\text{O} \leftrightarrow \text{HCO}_3 + \text{H}^+$, which is also identified as Equation (6) on page 6 of the present application.

Considering also that it was generally known that alkanolamines were suitable carbon dioxide absorption compounds (see also page 1, lines 18-19 of the application as filed), the skilled person gathers from the Example of D1 the use of carbonic anhydrase as an activator to increase carbon dioxide absorption rate in an aqueous solution used for carbon dioxide absorption, wherein the aqueous solution contains a primary alkanolamine ("Tris") as carbon dioxide absorption compound, and wherein the carbonic anhydrase is used "free", i.e. is added directly into the absorption solution.

The technical problem according to the Appellant

7. According to the Appellant, the problem solved by the claimed invention consisted in providing a method for absorption of carbon dioxide achieving unexpectedly good results in terms of enhancement of the CO₂ absorption rate.

The solution

8. The application at issue, in the amended forms according to both Auxiliary Request IV (in one of two alternatives) and Auxiliary Request V, proposes to solve this problem by the use as defined in the respective Claims 1, which is characterized in particular in that the alkanolamine used is "*N-methyldiethanolamine (MDEA)*".

The alleged success of the solution

9. As regards the unexpected results allegedly achieved by the claimed use, as compared to the use according to D1 (solution of Tris comprising "free" carbonic anhydrase), the Appellant relied in particular on the comparative test results submitted with its statement setting out the grounds of appeal.
 - 9.1 These comparative tests have been carried out using an absorption solution with a specific concentration of an alkanolamine CO₂ absorption compound. The carbonic anhydrase, when present, was directly added to the solution. Carbon dioxide was injected into the reactor to reach an initial pressure level of 10 psi, and the pressure decrease over time was monitored and used to calculate the CO₂ flux across the gas-liquid interface. The comparative tests were conducted using the tertiary alkanolamines MDEA, triethanolamine (TEA) and the primary alkanolamine Tris, at concentrations of 1 M and 2 M, with and without carbonic anhydrase. The results are summarised and depicted in Figures A and B.
 - 9.1.1 According to Figure A, the best carbon dioxide flux (mmol/min.m²) is, at both concentrations of 1 and 2 M, obtained with the combination MDEA-enzyme, the second

best with Tris-enzyme and the poorest with TEA-enzyme.

Hence, Figure A shows that under the chosen conditions, MDEA performs better than Tris, but not than TEA, i.e. that not all tertiary alkanolamines perform better than the primary alkanolamine Tris.

9.1.2 Figure B shows a relative performance, namely the ratio between the carbon dioxide fluxes achieved with and without enzyme, respectively, for each tested alkanolamine, and at both tested concentrations of the alkanolamine. According to Figure B, the increase attained under the given experimental conditions in the presence of the enzyme is more pronounced for both MDEA and TEA, as compared to Tris.

9.2 These comparative tests were, however, carried out with a gas phase consisting of carbon dioxide only. Hence, as pointed out by the Board in its communication, it is questionable whether the reported results are actually comparable to those achievable in a spray absorption operation as described in the example of D1, in which the gas subjected to the absorption process contained only a low concentration of carbon dioxide ("52000ppm", i.e. 5.2%).

9.3 Thus, for the Board, it was not convincingly shown that under the conditions of the example of D1, any of the tertiary alkanolamines tested, and in particular MDEA, would actually perform better than Tris in terms of CO₂ flux and/or CO₂ Flux Ratio.

9.4 Furthermore, Claim 1 is entirely open as to the operating conditions of the claimed use. The latter thus need not necessarily be the same as those used in the comparative examples submitted by the appellant.

Hence, there is no evidence on file demonstrating the alleged superiority of MDEA across the full ambit of Claim 1 in terms of operating conditions.

9.5 Moreover, even considering, for the sake of argument only, but in favour of the Appellant, that the tertiary alkanolamine MDEA performed better than primary alkanolamines such as Tris, the Board holds that this would not come as an unexpected effect for a skilled person, i.e. in the light of common general knowledge concerning the reaction mechanism involved.

9.5.1 The known reaction mechanism acknowledged in the application as filed (page 3, lines 19-30) but also, for instance in D7 (column 2, lines 40 *et seq.*) appears to be based on a base catalysis of the carbon dioxide hydration reaction, as confirmed for instance by D8 (page 207, right column, fourth full paragraph; page 221, in particular left column, first two paragraphs; page 224, left column, "Conclusions", third paragraph). According to D8, MDEA performed better than TEA in the absorption experiments carried out because in view of the mechanism according to Reaction (26) and of the higher pKa value of MDEA compared to TEA, there was a positive effect of the amine basicity on the catalytic activity observed (Page 221, paragraph bridging left and right column).

9.5.2 Hence, the comparatively better results illustrated by e.g. Figure A (with and without enzyme), of the comparative experiments submitted by the Appellant with its statement, appear to merely confirm that the skilled person would have expected a better performance of MDEA compared to TEA. According to D8, this better performance results from the pKa value of MDEA being

higher than that of TEA (MDEA being a stronger base than TEA).

- 9.5.3 The results obtained over Tris do not appear to be surprising either. Firstly, Tris is not a tertiary alkanolamine. Further, Tris reacts on its own with carbon dioxide, and its pKa lies between those of TEA and MDEA. Hence, Tris has a stronger amine basicity than TEA but a weaker one than MDEA.

Based on these known properties, it could be expected that MDEA performs better than Tris.

- 9.5.4 Hence, the Board concludes that the comparative data filed by the Appellant are not, on the one hand, suitable to demonstrate that an effect (in terms an improved absorption rate as compared to the use of Tris) as invoked by the Appellant can actually be reliably attained across the whole breadth of Claim 1 at issue. On the other hand, no unexpected effect, let alone synergy (in terms of the performance of MDEA as compared to Tris) can be gleaned therefrom.

Reformulation of the technical problem

10. Since it is apparent from the foregoing that the Appellant invoked an improvement over the closest prior art D1 which is neither addressed or foreshadowed in the application as filed nor proven across the whole breadth of Claim 1, the problem solved over D1 must be reformulated in a less ambitious way.

The technical problem in the light of D1 may thus be merely be seen in providing of a further use of carbonic anhydrase as an activator to promote carbon

dioxide absorption rate in an aqueous solution used for carbon dioxide absorption.

Success of the solution

11. Considering in particular the Appellant's comparative data as summarised in Figures A and B, the Board accepts as plausible that this less ambitious problem is effectively solved by the use wherein the CO₂ absorption compound is MDEA.

Obviousness

12. The use according to the respective Claim 1 differs from the use disclosed in D1 in that the tertiary alkanolamine MDEA is used instead of Tris.
 - 12.1 The suitability of the tertiary alkanolamine MDEA absorption compound for absorbing and, hence, separating carbon dioxide from gases was well known, see for instance documents D6 (Column 1, lines 33-37); D7 (column 2, lines 40-58, and column 3, lines 38-41), and D8 (*supra*).
 - 12.2 Thus, for the skilled person seeking to solve the less ambitious technical problem posed (Point 10, *supra*), the use of the tertiary alkanolamine MDEA in replacement of, or in addition to, Tris used according to the closest prior art D1 represents a known possibility readily available to the skilled person.
 - 12.3 The Appellant neither disputed the Board's detailed considerations regarding inventive step, expressed as a provisional opinion in the communication (Points 14.1 to 14.6), nor provided counter evidence.

12.4 Consequently, in the Board's judgement, the use involving MDEA as CO₂ absorbing compound according to the respective claim 1 of Auxiliary Requests IV and V does not involve an inventive step (Articles 52(1) and 56 EPC).

Conclusion

13. None of the claim requests at issue is both admissible and allowable.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:

The Chairman:



D. Magliano

B. Czech

Decision electronically authenticated