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**Datasheet for the decision  
of 1 December 2016**

**Case Number:** T 1381/13 - 3.3.05

**Application Number:** 06255984.4

**Publication Number:** 1790614

**IPC:** C01B31/20, B01D53/60

**Language of the proceedings:** EN

**Title of invention:**

Purification of carbon dioxide

**Patent Proprietor:**

Air Products and Chemicals, Inc.

**Opponent:**

L'AIR LIQUIDE SOCIETE ANONYME POUR L'ETUDE ET L'  
EXPLOITATION DES PROCEDES GEORGES CLAUDE

**Headword:**

Carbon dioxide purification/AIR PRODUCTS

**Relevant legal provisions:**

EPC Art. 123(2), 56

**Keyword:**

Amendments - allowable (yes)  
Inventive step - (yes)

**Decisions cited:**

**Catchword:**



**Beschwerdekammern**  
**Boards of Appeal**  
**Chambres de recours**

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Case Number: T 1381/13 - 3.3.05

**D E C I S I O N**  
**of Technical Board of Appeal 3.3.05**  
**of 1 December 2016**

**Appellant:** L AIR LIQUIDE SOCIETE ANONYME POUR L ETUDE ET L  
(Opponent) EXPLOITATION DES PROCEDES GEORGES CLAUDE  
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**Decision under appeal:** Interlocutory decision of the Opposition  
Division of the European Patent Office posted on  
15 April 2013 concerning maintenance of the  
European Patent No. 1790614 in amended form.

**Composition of the Board:**

**Chairman** E. Bendl  
**Members:** G. Glod  
P. Guntz

### **Summary of Facts and Submissions**

- I. The appeal of the opponent (appellant) lies from the decision of the opposition division finding that patent EP-B-1790614 in amended form met the requirements of the EPC.
  
- II. The following documents of relevance to the present decision were cited in the decision under appeal:  
  
D11: Oryshchyn D. et al., 29<sup>th</sup> Int. Techn. Conference of Coal Utilization & Fuel Systems, April 2004  
S3: Torrente-Murciano et al., 1<sup>st</sup> Int. Oxyfuel Combustion Conference, Cottbus, Germany, 2009 (post-published)
  
- III. With the statement of grounds of appeal the appellant inter alia objected that the claims as maintained by the opposition division did not meet the requirements of Articles 123(2) and 56 EPC and submitted the following documents:  
  
D20: IPCC Special Report on Carbon Dioxide Capture and Storage, 2005, p. 106, 141, 265  
D21: Anheden M. et al., pages 1 to 6, GHGT 7, 2004
  
- IV. The board issued a communication pursuant to Article 15(1) Rules of Procedure of the Boards of Appeal (RPBA) expressing its preliminary opinion.
  
- V. The respondent filed eight auxiliary requests with a letter of 13 October 2016.
  
- VI. Oral proceedings took place on 1 December 2016, at which the respondent made the second auxiliary request filed on 13 October 2016 its sole request. The only objections raised by the appellant relating to the

substance of the claimed subject-matter were in the context of the requirements of Articles 123(2) and 56 EPC.

VII. Independent claims 1 and 2 read as follows:

*"1. A method for the removal of SO<sub>2</sub> contaminant from carbon dioxide flue gas (1, 2, 3, 4, 5, 11, 12, 13) from an oxyfuel combustion process, said method comprising:  
maintaining said carbon dioxide flue gas (5, 13) at elevated pressure(s) in the presence of molecular oxygen ("O<sub>2</sub>") and water and NO<sub>x</sub>, at a temperature of no more than about 80°C for a sufficient time to convert NO<sub>x</sub> to nitric acid (14) and all of said SO<sub>2</sub> to sulfuric acid (6); and  
separating said sulfuric acid (6) and nitric acid (14) from said carbon dioxide flue gas (5, 13) to produce SO<sub>2</sub>-free, NO<sub>x</sub>-lean carbon dioxide gas (20),  
wherein said elevated pressure(s) is at least about 0.3 MPa (about 3 bar); and wherein the concentration of NO<sub>x</sub> in the flue gas is at least 300 ppm.*

*2. A method for the removal of SO<sub>2</sub> and NO<sub>x</sub> contaminants from carbon dioxide flue gas (1, 2, 3, 4, 5, 11, 12, 13) from an oxyfuel combustion process, said method comprising:  
maintaining said carbon dioxide flue gas (5, 13) at elevated pressure(s) in the presence of molecular oxygen ("O<sub>2</sub>") and water at a temperature of no more than about 80°C for a sufficient time to convert all of said SO<sub>2</sub> to sulfuric acid (6) and 90% of said NO<sub>x</sub> to nitric acid (14); and  
separating said sulfuric acid (6) and nitric acid (14) from said carbon dioxide flue gas (5, 13) to produce SO<sub>2</sub>-free, NO<sub>x</sub>-lean carbon dioxide gas (20),*

*wherein said elevated pressure(s) is at least about 0.3 MPa (about 3 bar); and wherein the concentration of NO<sub>x</sub> in the flue gas is at least 300 ppm."*

Claims 3 to 21 relate to preferred embodiments of claim 1 and/or 2.

VIII. The arguments of the **appellant (opponent)** may be summarised as follows:

The request should not be admitted into the proceedings, since the objections leading to the submission of this request had already been raised during opposition proceedings; so it should have been submitted before.

Article 123(2) EPC

It was not derivable from the application as filed that substantially all of the sulfur dioxide was converted to sulfuric acid. Other products were possible. The complete conversion to sulfuric acid was disclosed only for a special reactor set-up.

By analogy therewith, the removal of 90% of NO<sub>x</sub> was not originally disclosed either in the claimed context.

Article 56 EPC

It was evident that the flue gas treated in D11 contained mono-nitrogen oxides. Typical ranges were given in D20 and D21. S3 (slide 11/printout page 14) showed that even amounts smaller than 300 ppm mono-nitrogen oxides catalysed the reaction of sulfur dioxide to sulfuric acid. The 300 ppm were just an arbitrary number not providing any benefit. The

addition of more mono-nitrogen oxides would merely render the process of D11 more cumbersome without leading to any advantages that would be supported by evidence. Therefore the subject-matter of claims 1 and 2 was obvious in view of D11 in combination with D20 or D21.

IX. The arguments of the **respondent (patent proprietor)** may be summarised as follows:

Article 123(2) EPC

It was unambiguously derivable that all of the sulfur dioxide was to be converted to sulfuric acid (page 4, lines 12 to 15, in combination with page 5, lines 1 and 2, of the application as filed). The "maximum" referred to on page 6, line 15, of the application as filed had to be read in combination with page 5, lines 1 and 2, of the application as filed.

The value 90% of any NO<sub>x</sub> (page 5, lines 1 to 2) was to be read in the context of the preceding paragraphs.

Article 56 EPC

D11 was the closest prior art. It was completely silent as to the amount of mono-nitrogen oxides in the flue gas. It was evident from Table 1 that, if present, their content was less than 55 ppm. The recirculation of mono-nitrogen oxides was expected to reduce their concentration in the exhaust gas. There was no disclosure of a reaction to nitric acid.

Neither D20 nor D21 contained information on the content of mono-nitrogen oxides in raw flue gas.

The problem to be solved with respect to D11 was to remove sulfur oxide and mono-nitrogen oxides to the required extent.

The solution was to have at least 300 ppm of mono-nitrogen oxides present in the flue gas and to allow sufficient contact time.

The addition of mono-nitrogen oxides enabled the production of nitric acid that reacted with mercury so that it could also be removed.

The presence of mono-nitrogen oxides facilitated the rapid conversion of sulfur dioxide to sulfur trioxide that dissolved rapidly in water, such that the rate of removal of sulfur dioxide was increased as compared to D11. D11 taught only the oxidation of sulfur dioxide to sulfur trioxide via heterogeneous catalysis.

Further, in the presence of mono-nitrogen oxides sulfuric acid was produced that might react directly with limestone to produce gypsum.

None of the prior-art documents prompted the skilled person to add mono-nitrogen oxides.

- X. The appellant (opponent) requests that the decision of the opposition division be set aside and that the patent be revoked.

The respondent (patent proprietor) requests that the patent be maintained on the basis of the claims according to the sole request, filed as auxiliary request 2 with letter of 13 October 2016.



## **Reasons for the Decision**

1. Admissibility of the request (Articles 13(1) & (3) RPBA)

The request was first filed approximately seven weeks before the oral proceedings. It overcomes clarity objections raised by the board in its communication pursuant to Article 15(1) RPBA by restricting claims 1 and 2. Although the clarity objections had already been raised during opposition proceedings and were reiterated in the statement of grounds, the request is a direct reaction to the board's communication. It does not lead to new problems, since it is a restriction of the claimed subject-matter by simple, non-surprising amendments such that the complexity of the case is reduced. In addition it helps to speed up the proceedings. Since the amendments are simple and straightforward, the appellant and the board can deal with them easily.

Therefore, the board sees no reason not to admit the request into the proceedings.

2. Article 123(2) EPC

The requirements of Article 123(2) EPC are fulfilled for the following reasons:

- 2.1 It is undisputed that the application as filed discloses that at least substantially all sulfur dioxide and 90% of mono-nitrogen oxides present in the carbon dioxide flue gas are removed by the method (page 5, lines 1 to 2, as originally filed). The method in this context is the method specified on page 4, lines 9 to 17, since that method is the one according to the invention and no other method has been described

elsewhere in the application as filed. The method is defined by the conversion of sulfur dioxide to sulfuric acid and mono-nitrogen oxide to nitric acid. The skilled person understands that at least "substantially all" sulfur dioxide should react to sulfuric acid and 90% of mono-nitrogen oxides should react to nitric acid. This is also in line with page 6, lines 12 to 26, and page 8, lines 27 to 31, which specify that sulfur dioxide is removed as sulfuric acid and mono-nitrogen oxides as nitric acid, provided that the contact time for the reactive components is sufficient and the pressure and temperature are chosen suitably.

- 2.2 The apparatus specified on page 4, lines 18 to 31, is considered as an apparatus that is suitable to implement the method identified before, but the skilled person would not understand this apparatus to be the only apparatus suitable for the method. This understanding is in line with page 6, lines 19 to 23, where counter current gas/liquid contact devices are presented as suitable for providing the required contact times for removing at least substantially all sulfur dioxide and the bulk of mono-nitrogen oxides. This means that the apparatus is just an example of a device allowing the complete removal of sulfur dioxide and the bulk of mono-nitrogen oxides.
- 2.3 There is no doubt that "at least substantially all sulfur dioxide" also means that all of the sulfur oxide may be removed. The bulk is defined as meaning "usually about 90%", whereby the skilled person understands that 90% of the mono-nitrogen oxides are removed.
- 2.4 Thus, the skilled person can derive from the cited passages on pages 4 to 6 that the features at issue

were originally disclosed in the context presently claimed.

3. Article 56 EPC

3.1 Invention

The invention concerns a method for the removal of SO<sub>2</sub> and/or NO<sub>x</sub> from carbon dioxide flue gas (paragraph 1).

3.2 Closest prior art

D11 is the closest prior art. This view has also been shared by the parties.

D11 discloses an integrated pollutant removal system with flue gas recirculation. The results presented in Table 1 and equations (1) and (2) indicate that sulfur dioxide after dissolution in water is mainly removed as sulfurous acid. Small amounts of sulfur dioxide are oxidised to sulfur trioxide, which is dissolved in water more readily than sulfur dioxide, which means that a small part is removed as sulfuric acid (page 5, lines 1 and 2). D11 also discloses that the recirculation of the flue gas reduces the presence of NO<sub>x</sub> (page 1, abstract, lines 2 and 3; page 8, lines 1 to 3). However, D11 is silent as to the concentration of NO<sub>x</sub> present in flue gas. In any case, the exhaust flue gas given in Table 1 does not mention any mono-nitrogen oxides; so the maximum concentration of mono-nitrogen oxides has to be below the degree of accuracy - 10<sup>-4</sup> = 100 ppm - given in the Table.

D20 and D21 show that NO<sub>x</sub> is present in flue gas from an oxyfuel combustion process. However, these values represent **mean** values; so it cannot be concluded that

the exhaust flue gas of D11 will **inevitably** have such a NO<sub>x</sub> content. In addition, the values of D20 and D21 do not relate to a "raw" flue gas as do the values given in Table 1 of D11 for the exhaust flue gas. D20 and D21 do not permit the conclusion that D11 implicitly discloses directly and unambiguously an exhaust gas having a NO<sub>x</sub> concentration of at least 300 ppm.

### 3.3 Claim 1

#### 3.3.1 Problem

According to the patent, the problem to be solved is to provide an improved method for the removal of sulfur dioxide from carbon dioxide flue gas (paragraph 16).

#### 3.3.2 Solution

As a solution to this problem a method according to claim 1 is proposed wherein the mono-nitrogen oxides are brought to 300 ppm in the flue gas and the contact time is such that the mono-nitrogen oxides are converted to nitric acid and all of the sulfur dioxide to sulfuric acid.

#### 3.3.3 Success of the solution

3.3.4 As an argument in favour of the appellant it is accepted that there is no clear evidence that the method can be considered as improved compared to D11 if mono-nitrogen oxides are not present in the flue gas, a possibility which is covered by claim 1 (see also claim 16). Table 1 of D11 shows that the process of D11 allows the complete removal of sulfur dioxide (see Table 1).

### 3.3.5 Reformulation of the problem

The problem to be solved is to provide an alternative method for the removal of sulfur dioxide.

### 3.3.6 Success of the solution

Although it appears that the addition of a further step, namely the addition of mono-nitrogen oxides, to the process of D11 can be considered a disadvantageous modification, this step also provides some benefits that compensate for this disadvantage.

The addition of mono-nitrogen oxides allows sulfur dioxide to be oxidised to sulfur trioxide which dissolves rapidly in water to provide sulfuric acid. Sulfuric acid can be reacted with limestone to produce gypsum (see also paragraph 56 of the application). The fact that all of the sulfur dioxide is removed via sulfuric acid does not require any subsequent treatment of the sulfurous acid to sulfuric acid, as would be the case in D11.

Therefore, it is accepted that the proposed solution is an alternative to the process of D11 leading to a different main product (sulphuric versus mainly sulphurous acid).

### 3.3.7 Obviousness

D11 is completely silent as to the reaction of mono-nitrogen oxides with sulfur dioxide to sulfur trioxide. Consequently it does not teach that the flue gas could be kept for a sufficient time at pressure and temperature conditions suitable to provide sulfuric acid.

D11 teaches that catalytic oxidation could be used to increase sulfur dioxide dissolution for recovery (page 5, lines 4 and 5 from bottom), but it does not indicate that this oxidation could be done by mono-nitrogen oxides present at levels above 300 ppm. Although D11 discloses that mono-nitrogen oxides are present in the flue gas, there is no indication that the mono-nitrogen oxides react to nitric acid, let alone with sulfur dioxide to provide sulfur trioxide.

D20 and D21 do not concern the removal of sulfur dioxide from flue gas and do not provide any information about the oxidation of sulfur dioxide with mono-nitrogen oxides.

The board concludes that the solution to the posed problem is non-obvious vis-à-vis the documents presented and therefore involves an inventive step.

### 3.4 Claim 2

#### 3.4.1 Problem

According to the patent, the problem to be solved is to provide an improved method for the removal of sulfur dioxide and mono-nitrogen oxides from carbon dioxide flue gas (paragraph 16).

#### 3.4.2 Solution

As a solution to this problem a method according to claim 2 is proposed wherein the flue gas contaminated with mono-nitrogen oxides contains nitrogen oxides at a concentration of at least 300 ppm and the contact time is such that 90% of the mono-nitrogen oxides are

converted to nitric acid and all of the sulfur dioxide to sulfuric acid.

#### 3.4.3 Success of the solution

It is accepted that the problem is solved, since the method allows the removal not only of all of the sulfur dioxide as sulfuric acid, but also of 90% of the mono-nitrogen oxides as nitric acid from the flue gas.

The post-published document S3 may show that the reaction between mono-nitrogen oxides and sulfur dioxide already takes place at lower concentrations than 300 ppm of mono-nitrogen oxides, but this does not question the success of the solution, but rather confirms that mono-nitrogen oxides react act as catalysts for the oxidation of sulfur dioxide to sulfur trioxide.

#### 3.4.4 Obviousness

As already indicated for claim 1 (see point 3.3.7), D11 is silent as to any reaction of mono-nitrogen oxides with sulfur dioxide.

The same applies to D20 and D21.

The board concludes that the solution to this problem also involves an inventive step.

#### 3.5 Since the subject-matter of claims 1 and 2 involves an inventive step, the same applies to claims 3 to 21, which are directly or indirectly dependent on claims 1 and/or 2.

## Order

### For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The case is remitted to the department of first instance with the order to maintain the patent in amended form on the basis of the set of claims of the sole request, submitted as "second auxiliary request" with the letter of 13 October 2016, and a description to be amended, if necessary.

The Registrar:

The Chairman:



C. Vodz

E. Bendl

Decision electronically authenticated