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Datasheet for the decision of 12 November 2018

Case Number: T 1144/17 - 3.3.05

Application Number: 10797026.1

Publication Number: 2452918

IPC: C01B7/19, C01F11/46

Language of the proceedings: EN

Title of invention:
METHOD FOR PRODUCING HYDROGEN FLUORIDE

Patent Proprietor:
Daikin Industries, Ltd.

Opponent:
Solvay SA

Headword:
Production of HF/DAIKIN

Relevant legal provisions:
EPC Art. 83, 54, 56

Keyword:
Sufficiency of disclosure - (yes)
Novelty - multiple selection - (yes)
Inventive step - (yes)

Decisions cited:

Catchword:

Europäisches Patentamt
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Office européen des brevets

Beschwerdekammern

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Case Number: T 1144/17 - 3.3.05

D E C I S I O N
of Technical Board of Appeal 3.3.05
of 12 November 2018

Appellant: (Opponent)	Solvay SA Rue de Ransbeek, 310 1120 Brussels (BE)
Representative:	Mross, Stefan P.M. Solvay SA Intellectual Assets Management RIC Lyon 85 avenue des Frères Perret BP 62 69182 Saint-Fons (FR)
Respondent: (Patent Proprietor)	Daikin Industries, Ltd. Umeda Center Building 4-12, Nakazaki-Nishi 2-chome Kita-ku Osaka-shi Osaka 530-8323 (JP)
Representative:	Hoffmann Eitle Patent- und Rechtsanwälte PartmbB Arabellastraße 30 81925 München (DE)
Decision under appeal:	**Decision of the Opposition Division of the European Patent Office posted on 1 March 2017 rejecting the opposition filed against European patent No. 2452918 pursuant to Article 101(2) EPC.**

Composition of the Board:

Chairman	E. Bendl
Members:	T. Burkhardt
	S. Fernández de Córdoba

Summary of Facts and Submissions

I. The appeal of the opponent (hereinafter the appellant) lies from the opposition division's decision to reject the opposition against European patent EP-B-2 452 918.

II. The opposition division held that the grounds for opposition mentioned in Articles 100(b) and 100(a) in conjunction with Articles 52(1), 54 and 56 EPC did not prejudice the maintenance of the patent as granted.
In the proceedings before the opposition division the following documents were among those discussed:

D1 "Neue Erkenntnisse zum Mechanismus der Fluorwasserstoffherstellung aus Flußspat und Schwefelsäure", D Hass *et. al.*, Chem. Techn. 41, 144-147, April 1989
D2 JP 2005-132652 A / Translation into English
D3 "Anorganische Grundstoffe, Zwischenprodukte", Winnacker and Küchler, Chemische Technik, Prozesse und Produkte, Band 3, 5th ed. 2005, 612, Wiley-VCH-Verlag
D4 "Kinetsche Untersuchungen zur Reaktion zwischen Calciumfluorid und Schwefelsäure", E Kemnitz et al., Z. phys. Chemie, Leipzig, 271, 999-1007, 1990
D5 EP 1 300 362 A1
D6 DE 1 040 001 A
D7 GB 2 159 136 A
D8 US 6 355 221 B1

III. With the grounds of appeal, the appellant filed three additional documents:

D11 WO 01/85615 A1
D12 JP 2005-296888 A2

D13 JP 2006-212471 A2

IV. With the reply to the grounds of appeal dated 24 November 2017, the proprietor (respondent) maintained the claims upheld by the opposition division (claims as granted) as its main request and submitted auxiliary requests 1 to 3.

V. Independent claims 1 and 3 of the main request read as follows:

"1. A method for producing hydrogen fluoride by reacting calcium fluoride with sulfuric acid, which comprises
(a) a step for mixing and reacting calcium fluoride particles having an average particle diameter of 1-40 μm with sulfuric acid at a sulfuric acid/calcium fluoride molar ratio of 0.9-1.1 under a temperature of 0-70°C to obtain a solid-state reaction mixture; and
(b) a step for heating the solid-state reaction mixture to a temperature of 100-200°C to react with itself, and thereby producing hydrogen fluoride in a gas phase."

"3. A method for producing hydrogen fluoride by reacting calcium fluoride with sulfuric acid, which comprises
(c) a step for mixing and reacting calcium fluoride particles having an average particle diameter of 1-40 μm with sulfuric acid at a sulfuric acid/calcium fluoride molar ratio of 1.1-2.2 under a temperature of 0-70°C to obtain a solid-state reaction mixture; and
(d) a step for adding and mixing calcium fluoride particles having an average particle diameter of 1-40 μm to and with the solid-state reaction mixture at a sulfuric acid/calcium fluoride molar ratio of 0.9-1.1 in total of the steps (c) and (d), and then heating a

resultant mixture to a temperature of 100-200°C to react with itself, and thereby producing hydrogen fluoride in a gas phase."

Claims 2 and 4 to 5 are dependent on claims 1 and 3 respectively, and describe preferred embodiments thereof.

VI. The arguments of the **appellant** as far as relevant to the present decision may be summarised as follows:

- Articles 100(b) / 83 EPC:

Particle samples with the claimed "average [CaF_2] particle diameter" could include very large particles which would not be workable for HF manufacture. For the same reason the alleged effect would not be present over the entire claimed range.

- Articles 100(a) / 54 EPC:

The subject-matter of claims 1 and 2 was anticipated by **D5** in spite of the need to select several sub-ranges, especially since the general knowledge, as illustrated by **D3**, indicated that small particle diameters should be used.

Likewise, the subject-matter of claims 1 and 2 was anticipated by **D1**. The "average particle diameter" would be implicitly disclosed in D1 since the reaction product of the first step was solid, like the intermediate product resulting from step (a) of present claim 1.

- Articles 100(a) / 56 EPC:

D1 was the closest prior art. The problem to be solved vis-à-vis D1 was the provision of an alternative method, since the alleged effect was not present over the entire claimed range. The skilled person would choose a small particle size in the claimed range.

Even when starting from **D5** or **D7** as closest prior art, the skilled person would select the claimed particle diameter sub-range in view of **D2**, **D3** or **D8**.

Generally, the avoidance of the second pasty state was merely a "bonus-effect" of a reduced particle diameter.

VII. The arguments of the **respondent** as far as relevant to the present decision may be summarised as follows:

- Articles 100(b) / 83 EPC:

The appellant had failed to provide evidence that HF could not be manufactured using particles with the claimed average diameter.

- Articles 100(a) / 54 EPC

In order to arrive at the subject-matter of claim 1, it would be necessary to make multiple selections from the ranges disclosed in **D5**. Moreover, D5 did not disclose a solid-state reaction product of step (a).

D1 did not disclose the average particle diameter and the appellant had failed to provide any proof that this requirement was implicitly met.

- Articles 100(a)/56 EPC

Starting from **D1**, it was firstly necessary to select one of two alternatives for step (a). Even then, there was no incentive for the skilled person to select the claimed particle diameter in order to solve the problem at issue.

D5 and **D7** were also silent on the second pasty state.

VIII. The appellant requested that the impugned decision be set aside and the patent be revoked.

The respondent requested that the appeal be dismissed, or alternatively that the patent be maintained in amended form on the basis of one of auxiliary requests 1 to 3, all filed with the reply to the grounds of appeal dated 24 November 2017.

Reasons for the Decision

1. Main request: Articles 100(b) / 83 EPC

1.1 The independent claims are directed to methods for producing hydrogen fluoride involving the use of CaF_2 particles with a specific average diameter.

The appellant holds that there are CaF_2 particle distributions that, while respecting the claimed average particle diameter, contain coarse particles and do not work for the production of HF. Indeed, the common general knowledge, as illustrated by D3 (p. 612, par. 4), or D2 (par. [18]) would teach that only particles with a diameter of less than 150 µm or 100 µm respectively have to be used.

1.2 For the following reasons the board considers that the invention is disclosed in a manner sufficiently clear and complete to be carried out by a person skilled in the art:

1.3 According to par. [27] of the patent in suit, the average particle diameter may be determined by laser diffraction particle size distribution analysis, a well-known and established method. The skilled person can thus readily identify suitable particle distributions.

1.4 However, while such particle distributions may indeed include coarser particles, i.e. individual particles having a diameter of more than 40 µm, the appellant failed to submit evidence, e.g. in the form of experimental data, or convincing arguments for its allegation that such distributions meeting the requirements as defined in claim 1 would not work at all.

1.5 Moreover, during the oral proceedings, the appellant submitted that the skilled person was at a loss when attempting to verify the absence of the second pasty state in apparatuses of industrial scale. In other words, the effect that the second pasty state does not occur could be neither observed nor achieved over the entire claimed range when scaling-up the experiments described in the examples of the patent in suit.

1.6 Notwithstanding the question of the admissibility of this late-filed objection, the board notes that the effect that the second pasty state does not occur is not an issue of sufficiency of disclosure, especially since it does not figure in the claims. Hence this objection relates to the requirements of Art. 56 rather

than to those of Art. 83 EPC. Furthermore, again no proof for these allegations has been provided.

1.7 The board therefore concludes that the requirements of Article 83 EPC have been met.

2. **Main request: Articles 100(a) / 54 EPC**

The appellant holds that the subject-matter of independent claim 1 is anticipated by either of D1 or D5.

2.1 While admitting that **D1** does not *explicitly* disclose the claimed average CaF_2 particle diameter, the appellant holds that it is *implicitly* disclosed: Table 1 of the patent in suit showed that a larger particle diameter would result in a non-solid reaction product of step a). Conversely, the presence of a solid reaction product of step a) in D1 (p.147 col.1 par.2) would imply that the particle diameter was in the claimed range.

2.2 The board does not share the view that a particle diameter in the claimed range can be directly and unambiguously derived from the solid state of the reaction product of step a).

The appellant did not provide experimental results reproducing the set-up of D1 with varying average particle sizes showing that only for average particle diameters in the claimed range is the state of the reaction product of step a) solid. Yet it cannot be excluded that average particle diameters of between the claimed upper limit of 40 µm and below the 56 µm disclosed in Table 1 of D1 (where the product of step a) is no longer solid), on the one hand, or less than

1 µm, on the other hand, result in a solid product of step a) too.

Moreover, D1 describes on page 146, right-hand column, last full paragraph, that CaF_2 and H_2SO_4 are used in approximately stoichiometric amounts ("etwa stöchiometrische Mengen"). It cannot be derived directly and unambiguously whether this indication of approximate amounts necessarily corresponds to a H_2SO_4/ CaF_2 molar ratio of 0,9 to 1,1 as presently claimed, or possibly lies somewhat outside this range.

Thus, in summary it cannot be concluded directly and unambiguously that D1 is novelty-destroying.

2.3 With regard to **D5**, the appellant held that par. [39, 40] disclosed the claimed average particle diameter and the temperatures of steps a) and b). The appellant was further of the opinion that par. [39] disclosed a solid reaction product of step a).

2.4 The board does not agree with this view, since selections from several lists are necessary to arrive at the claimed subject-matter:

- While claim 1 of the contested patent stipulates an average particle diameter of between 1 and 40 µm, D5 discloses in par. [39] a much broader range, namely between 0,1 and 2000 µm.
- A temperature in step a) in the sub-range of 40 to 70°C has to be selected from the range 40 to 200°C disclosed in D5 (par. [39]), and
- a temperature in step b) in the sub-range of 150 to 200°C has to be selected from the range 150 to 350°C disclosed in D5 (par. [40]).

Moreover, while it is true that line 7 of par. [39] discloses the word "powder", the board considers that it is only the calcium sulfate that is produced in the form of powder, whereas the entire reaction product is rather in the form of a "slurry or paste", as indicated in line 17 of col. 13.

2.5 Therefore, the claimed subject-matter is novel.

3. Main request: Articles 100(a) / 56 EPC

3.1 The invention relates to a two-step method for producing hydrogen fluoride from CaF_2 particles and sulphuric acid.

3.2 In the board's view, **D5** is to be considered as the closest prior art, since it relates to the production of hydrogen fluoride and also addresses the problem of the corrosive nature of the reaction mixture, which is actually a "slurry or paste" (par. [3]).

3.3 According to the patent in suit, the technical problem to be solved is the provision of a method that avoids the occurrence of the "second pasty state", i.e. a pasty state in step b) of the reaction.

3.4 As a solution to this technical problem, the patent in suit proposes the method according to claim 1, stipulating in particular the use of CaF_2 particles with an average particle diameter in the range of 1 to 40 µm, a selected sulfuric acid/calcium fluoride molar ratio and specific processing temperatures.

3.5 Table 1 of the contested patent shows in lines 1 to 3 that no second pasty state is observed when an average particle diameter in the claimed range is used while

applying the remaining claimed processing conditions, whereas under comparable conditions a second pasty state is observed when an average particle diameter above the upper average particle size diameter threshold is used (line 5). In the absence of evidence/ experimental results to the contrary, the board considers that the problem is solved over the entire claimed range.

3.6 For the following reasons the board considers that it was not obvious for the skilled person, having regard to the state of the art and the common general knowledge, to solve the technical problem posed in the claimed manner.

D5 mentions in par. [3] that the reaction mixture is a corrosive "paste or slurry", but is entirely silent on any possibility to avoid the occurrence of such a corrosive second pasty state, let alone to achieve this by means of a specific average particle diameter. D5 overcomes the problem of corrosion by using a specifically designed reactor.

3.7 When starting from the closest prior art, the remaining documents cited by the appellant do not hint at a solution to the posed problem in the claimed manner either:

- **D3** discloses on page 612 that the CaF_2 particles should, for unspecified reasons, be smaller than 150 µm. However, a diameter of 150 µm is still remote from the claimed upper limit of 40 µm. So even if the skilled person contemplated the teaching of D3, he would not get any teaching towards the claimed range and the avoidance of the second pasty state.

- **D8** discloses in col.5 lines 26-54 an exemplary CaF_2 particle distribution with a range of between 20 and 150 µm, with a narrow particle size distribution being preferred. However, D8 is silent on both the second pasty state and the possibility of avoiding it by choosing a specific particle diameter. The skilled person has consequently no incentive to contemplate the teaching of D8 when seeking to solve the posed technical problem.

- While scientific papers **D1** and **D4** disclose the two-step reaction mechanism, they are silent on the average particle diameter and teach even less on the possibility to avoid the second pasty state by choosing particle diameters in the claimed range.

- **D6** discloses in col.3 lines 1-6 the formation of a solid reaction product in the first step that allows a reaction in the second step without the formation of a phase without liquid, but is silent on particle diameter and its role in the formation of the liquid.

The question of the admissibility of **D11-D13** notwithstanding, the board notes that, while at least D12 and D13 disclose particle diameters in the claimed range, none of these documents mentions the second pasty state or an incentive to address corrosion by choosing a specific particle diameter.

3.8 Hence the skilled person would not arrive at the claimed subject-matter when starting from **D5**, either alone or in combination with the cited documents of the prior art.

3.9 In addition to the findings above, the appellant holds that **D1** should be considered as the closest prior art, since D1 deals with the chemistry underlying the process, whereas D5 would seek to address the corrosion issue by means of a mechanical solution. It also referred to **D7**, but admitted that this document fails to indicate an average particle diameter in the claimed range.

However, since, in contrast to **D5**, **D1** and **D7** fail to address the problem of the corrosive nature of the reaction mixture, this view cannot be shared by the board. Thus, due to being more remote from the claimed subject-matter than D5, their combinations with the state of the art referred to in the appeal procedure would also not have led to the claimed invention in an obvious manner either.

3.10 Based on the above considerations, the board concludes that the subject-matter of the claims as granted involves an inventive step (Articles 52(1) and 56 EPC).

4. In conclusion, none of the grounds invoked by the appellant prejudices the maintenance of the patent as granted.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar: The Chairman:

C. Vodz E. Bendl

Decision electronically authenticated