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**Datasheet for the decision
of 21 January 2016**

Case Number: T 2186/12 - 3.3.05

Application Number: 01906687.7

Publication Number: 1251949

IPC: B01J8/02, B01J12/00, C01B3/38

Language of the proceedings: EN

Title of invention:
METHOD AND APPARATUS FOR OBTAINING ENHANCED PRODUCTION RATE OF
THERMAL CHEMICAL REACTIONS

Patent Proprietor:
BATTELLE MEMORIAL INSTITUTE

Opponent:
Fraunhofer-Gesellschaft zur Förderung der
angewandten Forschung e.V.

Headword:

Relevant legal provisions:
EPC Art. 123(2), 123(3), 56
RPBA Art. 13(1)

Keyword:
Late filed request - admitted (yes)
Amendments - added subject-matter (no) -
broadening of claim (no)
Inventive step - (yes)

Decisions cited:

G 0002/88, G 0010/91, T 1736/09

Catchword:



Beschwerdekammern
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Case Number: T 2186/12 - 3.3.05

D E C I S I O N
of Technical Board of Appeal 3.3.05
of 21 January 2016

Appellant:
(Patent Proprietor)

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Decision under appeal:

**Decision of the Opposition Division of the
European Patent Office posted on 9 August 2012
revoking European patent No. 1251949 pursuant to
Article 101(3) (b) EPC.**

Composition of the Board:

Chairman J.-M. Schwaller
Members: G. Glod
C. Vallet

Summary of Facts and Submissions

- I. The present appeal lies from the decision of the opposition division to revoke European patent EP-B-1 251 949 for lack of inventive step.
- II. The documents relevant to the present decision and cited during the opposition proceedings included the following:
- D2: Tonkovich et al., The Catalytic Partial Oxidation of Methane in a Microchannel Chemical Reactor, 1998
- D3: Schubert et al., Realization and Testing of Microstructure Reactors, Micro Heat Exchangers and Micromixers for Industrial Applications in Chemical Engineering, 1998
- D4: Charlesworth et al., Combustion and Steam Reforming of Methane on Thin Layer Catalysts for Use in Catalytic Plate Reactors, 1996
- D5: Cuta et al., Fabrication and Testing of Micro-Channel Heat Exchangers, SPIE, vol. 2640, September 1995
- D6: Tonkovich et al., Microchannel Heat Exchangers for Chemical Reactors, 1996
- III. With its grounds of appeal containing two annexes, the patent proprietor (hereinafter: the appellant) submitted a main request and ten auxiliary requests.
- IV. By letter of 12 July 2013, the opponent (hereinafter: the respondent) replied to the statement setting out

the grounds of appeal and, among others, filed the following document:

D10: Bier, W. et al., Gas to gas heat transfer in microheat exchangers, 1993

- V. The board issued a communication under Article 15(1) RPBA on 2 July 2015.
- VI. With a letter dated 20 November 2015 the appellant provided further arguments and submitted a new main request and auxiliary requests 1 to 8.
- VII. The respondent submitted further arguments with a letter dated 21 December 2015.
- VIII. Oral proceedings took place on 21 January 2016. The appellant made the third auxiliary request submitted with the letter of 20 November 2015 his sole request. In addition, he deleted examples 1 and 2 from the description and submitted amended pages 9 and 10 of the description.

Independent claim 1 of this request reads as follows:

"1. A process for the catalytic steam reforming of a hydrocarbon, comprising: passing at least one reactant into at least one reaction chamber; said reaction chamber comprising a catalyst that catalyzes the reaction of said at least one reactant; transferring heat to said at least one reaction chamber from at least one heat exchanger, and wherein a heat transfer fluid flows in the heat exchanger; obtaining at least one product from said reaction chamber; wherein the contact time of the reactant with the catalyst is less than 0.3 seconds and the pressure drop through the

reaction chamber is less than 15 psig (103 KPa); characterised in that said step of transferring heat, at steady state, transfers at least 0.6 W/cc of total reactor volume, where total reactor volume is defined as the sum of the volume of the reaction chamber(s) and heat exchanger chamber(s) including the volume of chamber walls, and wherein there are multiple reaction chambers and heat exchangers operating in parallel."

Dependent claims 2 to 7 represent specific embodiments of the process claim 1.

IX. The arguments of the **appellant** may be summarised as follows:

It would not consent to introducing the ground of sufficiency (Article 100(b)/Article 83 EPC) into the proceedings.

The new request was limited to a process as defined in claim 10 of the patent. The further limitations were included in reaction to the preliminary opinion of the board.

In agreement with the findings of the opposition division, the requirements of Articles 123(2), 123(3) and 54 EPC were met.

The inventors were the first people to realise that high selectivities and conversion rates in steam reforming of a hydrocarbon could be obtained by operating above a threshold heat flux between a reactor chamber and a heat exchanger, provided that the residence time was sufficiently short and the pressure drop in the reactor was low.

D2 could not be considered as closest prior art, since it did not relate to steam reforming of a hydrocarbon. Moreover, the heat-exchange fluid was air which was pre-heated by a combustion plate.

D4 related to the steam reforming of a hydrocarbon and was more appropriate as closest prior art. Yet it was still not relevant, as it did not relate to microreactors and taught towards improving the catalysts in order to improve selectivity.

There was thus no reason to combine D2 with D4.

X. The arguments of the **respondent** may be summarised as follows:

The new request should not be admitted into the proceedings, since it was filed only two months before the oral proceedings and contained considerable amendments compared to the previous requests. In addition, the amendments made were based on features taken from the description.

The feature "transferring heat **to** said at least one reaction chamber **from** at least one heat exchanger" was not directly and unambiguously derivable from the application as filed.

The expression "steady state" was broader than the expression "steady rate", since the former implied that the conditions were time-invariant while the latter related only to some specific aspects (heat transfer rate or flow speed of reactants). If the system was not at steady state the heat transfer did not have to be at least 0.6 W/cc, which was different from claim 1 of the patent as granted. Therefore, the requirements of

Article 123(3) EPC were not met.

The patent did not contain enough information enabling the person skilled in the art to carry out the invention over the whole scope claimed.

D2 could be considered as the closest prior art, since it had the most features in common with claim 1. The combined volume of the reactor and of the heat exchanger plates defined the reactor volume that had to be considered when calculating the heat transfer rate, since the steps indicated in claim 1 of the patent occurred there. The calculated heat transfer of D2 was within the range claimed. D2 also related to the production of syngas, and methane steam reforming was mentioned as an alternative to the oxidation of methane.

D4 could also be considered as the closest prior art, since it disclosed the combustion and the steam reforming of methane, and it showed that the heat fluxes were similar for both reactions. Therefore it was obvious to use the reactor of D2 for steam reforming as well.

The subject-matter of claim 1 lacked inventive step in view of the combinations of D2 with D4, or D4 with D2.

- XI. The appellant requested that the decision of the opposition division be set aside and that the patent be maintained in amended form on the basis of claims 1 to 7 of the main request submitted as auxiliary request 3 with the letter of 20 November 2015 and pages 9 and 10 of the description submitted during oral proceedings before the board.

The respondent requested that the appeal be dismissed.

Reasons for the Decision

1. Admissibility of the request (Article 13(1) RPBA)
 - 1.1 Under Article 13(1) RPBA, the admission of any amendment to a party's case after it has filed its grounds of appeal or reply is at the board's discretion. When exercising such discretion, the complexity of the new subject-matter submitted, the current state of the proceedings and the need for procedural economy should be considered.
 - 1.2 Since the request was filed after oral proceedings had been arranged, it constitutes an amendment to the appellant's case within the meaning of Article 13(1) RPBA.
 - 1.3 The board observes that claim 1 at issue is limited to the specific type of reaction which was the subject-matter of dependent claim 10 as granted, namely steam reforming of a hydrocarbon. Examples 2 and 4 of the application as filed furthermore relate to steam reforming of a hydrocarbon, so the restriction of claim 1 to this type of reaction is in line with these preferred embodiments and cannot be regarded as a surprise.

This amendment also helps to clearly distinguish the claim from the most relevant prior art discussed previously in the proceedings, and as such it provides *prima facie* a basis for an allowable set of claims (see below).

The amendment relating to the transfer of heat **to** the

reaction chamber **from** the heat exchanger is directly linked to the type of reaction and does not *per se* constitute a specific complexity.

The other features added to claim 1 ("*wherein a heat transfer fluid flows in the heat exchanger*" and "*wherein there are multiple reaction chambers and heat exchangers operating in parallel*") further limit the scope of the claim but are well-known to the skilled person, and so they cannot be considered as adding complexity to the case.

- 1.4 The board thus considers that the amendments made in claim 1 are a fair reaction to the decision of the opposition division and to the preliminary opinion of the board. Since the amendments are not unexpected and are not so complex that they would notably change the scope of discussion, and since they could *prima facie* lead to an allowable set of claims, they are admitted into the proceedings.

2. Amendments - Article 123(2) EPC

Claim 1 is based on claim 1 of the application as filed in combination with the passages at page 5, lines 9 and 10; page 16, lines 15 and 16, and page 19, lines 14 and 15, of the application as filed. It is furthermore known to a skilled person that steam reforming is an endothermic process that requires heat. In this particular process it is the heat provided by the heat exchanger that needs to be transferred to the reaction chamber. This is moreover in line with example 4 of the application as filed, which relates to isooctane steam reforming, wherein the heat needed by the reaction originates from the heat exchange fluid (page 25, lines 21 to 23).

The board therefore concludes that, using common general knowledge, the subject-matter of claim 1 is directly and unambiguously disclosed in the application as filed. The requirements of Article 123(2) EPC are thus met.

3. Extension of the scope of protection (Article 123(3) EPC)

Claim 1 as granted contains the wording "*characterised in that said step of transferring heat, at steady **rate**, transfers at least 0.6 W/cc of total reactor volume, [...]*". In contrast, in claim 1 at issue the term "state" has been substituted for the term "rate".

In determining whether amendments made in opposition proceedings comply with the requirements of Article 123(3) EPC, it must be borne in mind that the protection conferred by the European patent is determined by the claims as granted, with the description and drawings of the granted patent being used to interpret the claims in accordance with Article 69(1) EPC and its Protocol (G 2/88, Reasons 4, and T 1736/09, Reasons 1.1.8).

In the present case, it observed that the expression "steady rate" is not literally disclosed in the patent, but the expression "steady state rate" is mentioned at page 4, line 37, of the description.

For the board, the skilled person would recognise from the entire content of the patent that "steady rate" is to be understood as indicated at page 4 of the description, i.e. as "steady state rate", which implies that the heat transfer rate is to be determined at steady state. This interpretation is in line with the

wording of claim 11 as granted, which relates to a reactor [...] "*configured such that, during steady state operation, at least 0.6 W of heat per cc of total reactor volume can be transferred [...]*". So in the context of the patent "at steady rate" means "at steady state", and the replacement of the one by the other does not extend the scope of protection of the patent.

The requirements of Article 123(3) EPC are therefore fulfilled.

4. Sufficiency of disclosure - Article 83 EPC

The ground of opposition under Article 100(b) EPC not being part of the opposition proceedings, it is a fresh ground of opposition. Since the patentee did not approve its consideration, sufficiency of disclosure cannot be questioned (G 10/91, Reasons 18).

5. Novelty - Article 54 EPC

The respondent did not have any objection concerning the novelty of this request. The board sees no reason to differ.

6. Inventive step - Article 56 EPC

6.1 Invention

The invention concerns a process for the catalytic steam reforming of a hydrocarbon (claim 1).

6.2 Closest prior art

It is established jurisprudence that the closest prior art is normally a prior-art document disclosing the

same purpose or aiming at the same objective as the claimed invention and having the most features in common with the claimed subject-matter.

In the present case D4 is considered the closest prior art, since it relates to steam reforming of methane on thin layers of catalysts. Flow rates were set to achieve 10 kW/m^2 at complete conversion (chapter 3.2). Experiments were performed at 650 to 850°C. The apparatus used in the process is shown in figure 2 of D4.

D2 does not seem to constitute adequate closest prior art, since it does not relate to hydrocarbon steam reforming. It is true that D2 mentions methane steam reforming in a conventional fixed-bed technology as one way of producing hydrogen, but it teaches away from using microreactors, since it indicates that such a process is not well-suited for miniaturisation (D2: page 45, Introduction, second paragraph, last sentence). D2 clearly concentrates on the partial oxidation of methane to synthesis gas.

6.3 Problem

The problem underlying the patent in suit is to improve the efficiency and selectivity of hydrocarbon steam reforming (paragraph [0008] and page 3, lines 52 to 57, of the patent in suit).

6.4 Solution

As a solution to this problem the patent in suit proposes a method according to claim 1, characterised in that the contact time of the reactant with the catalyst is less than 0.3 seconds, the pressure drop

through the reaction chamber is less than 15 psig (103 KPa), the step of transferring heat, at steady state, transfers at least 0.6 W/cc of total reactor volume, and in that in the reactor there are multiple reaction chambers and heat exchangers operating in parallel.

6.5 Success of the solution

6.5.1 Example 4 of the patent relates to the steam reforming of isooctane conducted in the reactor configured as shown in Figure 6 and transferring 10 to 16 W/cc. The reactor was operated at 650°C, the contact time was 22 milliseconds and the pressure drop about 6.9 kPa (page 11, lines 51 to 57, of the patent). Isooctane conversions ranged from 86.5% to 95% and the hydrogen selectivity from 85 to 90% (figure 9). For the board, it can be accepted that example 4 illustrates that the steam reforming of isooctane was run efficiently.

6.5.2 D4 discloses the steam reforming of methane with a conversion of up to 40% being obtained at 650°C, while almost complete conversion was obtained at 850°C.

The conversion rate at 650°C might appear considerably higher in the reaction according to example 4 of the patent, but it should be noted that D4 and example 4 of the patent do not relate to the same reaction; so comparison is not possible. The steam reforming of isooctane is moreover much more endothermic than the steam reforming of methane.

The data presented in Annex 1 of the statement of grounds of appeal of 18 December 2012 are not relevant, since they do not relate to the steam reforming of a hydrocarbon.

Therefore, there is no convincing evidence that an improvement with respect to D4 is obtained, and the problem needs to be formulated in a less ambitious manner.

6.6 Reformulation of the problem

The problem can be seen as the provision of an alternative process for the steam reforming of hydrocarbons.

6.7 Success of the reformulated problem

Example 8 shows a specific embodiment according to claim 1 allowing the efficient steam reforming of isooctane. It is plausible that this process could also be used for other hydrocarbons. There is no proof to the contrary.

Therefore, it is accepted that this problem is successfully solved.

6.8 Obviousness

6.8.1 When establishing whether the solution is obvious in view of the state of the art, the board comes to the conclusion that the prior art does not lead to the solution of the problem as proposed in claim 1 at issue for the following reasons.

6.8.2 D4 is completely silent about the combination of contact time, pressure drop and heat transfer defined in claim 1. There is also no evidence that the combination of these process conditions is standard in the field.

As illustrated in the patent, these process conditions require a specific reactor configuration, which is not at stake in D4, which in particular discloses (chapter 1.2, first paragraph, last line) a reactor having a volume of 11 m³, while the patent teaches the use of a microreactor. D4 moreover does not provide any teaching leading towards the reaction conditions proposed in claim 1 at issue.

6.8.3 D2 discloses a micro-channel reactor for the partial oxidation of methane to synthesis gas having the configuration shown in figure 2. The combustor heat exchanger preheats an air stream, which is dedicated as heat exchanger fluid. In the configuration shown in figure 1, the reactor contains at the front a reaction zone housing active catalyst powders and at the back a heat transfer fluid. The residence time in the reactor is 49.3 milliseconds and the measured pressure drop less than 250 Pa.

For the board, the partial oxidation of methane being an exothermic reaction, it is not credible that the skilled person looking for an alternative process for an endothermic reaction would turn to an exothermic reaction.

As indicated above (point 6.2), D2 teaches away from using microreactors for methane steam reforming.

D2 moreover focuses on partial oxidation of methane to synthesis gas and does not provide any indication that the process conditions for partial oxidation of methane might also apply to methane steam reforming.

Therefore, the board is of the opinion that the skilled person trying to solve the posed problem would have no incentive to look at D2.

- 6.8.4 D3 discloses microstructure reactors with high heat transfer, but makes no mention of an endothermic reaction. In addition, D3 is completely silent about the pressure drop. The board fails to see why the skilled person trying to solve the posed problem would turn to D3.
- 6.8.5 D5 is completely silent about reaction conditions. It relates to heat exchangers for removing heat. D5 is not relevant to the solution of the problem.
- 6.8.6 D6 discloses an approach to increasing heat removal rates in chemical reactors by integrating a novel micro-channel heat exchanger. However, D6 does not mention endothermic reactions and does not indicate reaction temperatures. It discloses a maximum pressure drop of 23 psi (page 122, left-hand column, third paragraph, last line). The skilled person would not have taken this document into consideration when trying to solve the posed problem.
- 6.8.7 D10 discloses micro heat exchangers, but does not relate to the steam reforming of a hydrocarbon.
- 6.9 The requirements of Article 56 EPC are therefore fulfilled.

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 claims 1 to 7 of the main request, which corresponds to the former third auxiliary request filed with the letter dated 20 November 2015, pages 9 and 10 of the description as amended during the oral proceedings, with the entire description and the figures to be adapted as necessary.

The Registrar:

The Chairman:



C. Vodz

J.-M. Schwaller

Decision electronically authenticated