

**Internal distribution code:**

- (A) [ - ] Publication in OJ  
(B) [ - ] To Chairmen and Members  
(C) [ - ] To Chairmen  
(D) [ X ] No distribution

**Datasheet for the decision  
of 29 July 2014**

**Case Number:** T 0368/11 - 3.3.10

**Application Number:** 06706456.8

**Publication Number:** 1848681

**IPC:** C07C45/52

**Language of the proceedings:** EN

**Title of invention:**

PROCESS FOR DEHYDRATING GLYCEROL TO ACROLEIN

**Patent Proprietor:**

ARKEMA FRANCE

**Opponent:**

Müller-Boré & Partner

**Headword:**

**Relevant legal provisions:**

EPC Art. 54, 56

**Keyword:**

Novelty - (yes) no direct and unambiguous disclosure  
Inventive step - (yes) improvement shown, non-obvious solution

**Decisions cited:**

**Catchword:**



**Beschwerdekammern  
Boards of Appeal  
Chambres de recours**

European Patent Office  
D-80298 MUNICH  
GERMANY  
Tel. +49 (0) 89 2399-0  
Fax +49 (0) 89 2399-4465

Case Number: T 0368/11 - 3.3.10

**D E C I S I O N**  
**of Technical Board of Appeal 3.3.10**  
**of 29 July 2014**

**Appellant:** Müller-Boré & Partner  
(Opponent) Patentanwälte PartG mbB  
Friedenheimer Brücke 21  
81639 München (DE)

**Respondent:** ARKEMA FRANCE  
(Patent Proprietor) 420, rue d'Estienne d'Orves  
92700 Colombes (FR)

**Decision under appeal:** **Decision of the Opposition Division of the European Patent Office posted on 29 December 2010 rejecting the opposition filed against European patent No. 1848681 pursuant to Article 101(2) EPC.**

**Composition of the Board:**

**Chairman** P. Gryczka  
**Members:** J. Mercey  
C. Schmidt

## Summary of Facts and Submissions

I. The Appellant (Opponent) lodged an appeal against the decision of the Opposition Division rejecting the opposition against European patent No. 1 848 681, which was granted on the basis of nine claims, claim 1 of which read as follows:

"Process for manufacturing acrolein by gas-phase dehydration of glycerol in the presence of a strongly acidic solid catalyst with a Hammett acidity  $H_0$  of between -9 and -18."

II. Notice of Opposition had been filed by the Appellant requesting revocation of the patent as granted in its entirety on the grounds of lack of novelty and lack of inventive step. *Inter alia* the following documents were submitted in the opposition proceedings:

(1) L. H. Dao, M. Haniff, A. Houle, D. Lamothe, "Reactions of Model Compounds of Biomass-Pyrolysis Oils over ZSM-5 Zeolite Catalysts" in "Pyrolysis Oils from Biomass: Producing, Analyzing and Upgrading", Chapter 27, 1988, American Chemical Society,

(3) EP-A-1 710 227 and

(4) US-A-5 387 720.

III. The Opposition Division held that the subject-matter of granted claim 1 was novel over documents (1) and (3), these documents not disclosing a process using a catalyst having a Hammett acidity of between -9 and -18. It also held that the subject-matter involved an inventive step, document (1) being considered to represent the closest prior art. Carrying out the process with more strongly acidic catalysts resulted in the catalysts deactivating less quickly and being more

active. None of the cited prior art suggested that said improvements could be achieved by using a catalyst having a Hammett acidity of between -9 and -18, document (4) in fact teaching away from the subject-matter claimed.

IV. The Appellant submitted that the subject-matter of claim 1 of the granted patent was not novel over the disclosure of document (1), since the H-ZSM-5 zeolite used in the process disclosed this document had a Hammett acidity of between -16.12 and -8.2, as shown in the "Experimental Demonstration" filed with letter dated 3 May 2011. It argued that should the subject-matter of granted claim 1 be considered novel over document (1), then it was not inventive, since the person skilled in the art when considering the implicit teaching of said document, would inevitably arrive at the claimed subject-matter without any inventive efforts. Lack of inventive step of the subject-matter of the dependent claims 2 to 9 was also challenged on the basis of a combination of the teaching of document (1) with that of various other prior art documents.

V. The Respondent (Proprietor of the patent) submitted that document (1) did not disclose the process according to claim 1 of the granted patent, since the Appellant had not proved that the H-ZSM-5 catalyst used therein had a Hammett acidity of between -9 and -18. With letter dated 27 September 2011, the Respondent provided experimental data showing that the sodium content of an H-ZSM-5 zeolite had a considerable influence on its acidity and that H-ZSM-5 zeolites having a sodium content similar to that disclosed in document (1) had a Hammett acidity  $> -8.2$  and thus outside the claimed range. The claimed subject-matter was inventive, since Examples 2 to 6 of the patent in

suit showed that catalysts having a Hammett acidity of between -9 and -18 deactivated less quickly than less strongly acidic catalysts. With letter dated 21 March 2014, the Respondent filed auxiliary requests 1 and 2.

- VI. In a communication of the Board dated 11 March 2014 accompanying a summons to oral proceedings, it was noted that the Appellant no longer objected to novelty on the basis of document (3). The Board also questioned whether the Examples in the patent in suit were indeed suitable for demonstrating that the prolonged catalyst activity in the claimed process is directly attributable to the (potentially) characterising feature of the invention, namely the Hammett acidity between -9 and -18.
- VII. With letter dated 15 April 2014 the Respondent filed auxiliary request 3 and submitted a technical report intended to show that a process according to the invention employing an H-ZSM-5 zeolite having a Hammett acidity of  $< -12.0$  was superior to the process according to document (1) employing an H-ZSM-5 zeolite having a Hammett acidity of  $> -8.2$ .
- VIII. With letter dated 15 May 2014 the Appellant indicated that it would not be attending the oral proceedings and with letter dated 5 June 2014, the Respondent requested that the oral proceedings be cancelled and that the Board issue a written decision.
- IX. By a notification dated 15 July 2014 the Board informed the parties that the oral proceedings were cancelled.
- X. The Appellant requested that the decision under appeal be set aside and the patent be revoked.

The Respondent requested that the appeal be dismissed, or subsidiarily, that the patent be maintained on the basis of either of auxiliary requests 1 and 2, filed with letter dated 21 March 2014, or on the basis of auxiliary request 3, filed with letter dated 15 April 2014.

## **Reasons for the Decision**

1. The appeal is admissible.

*Main request (patent as granted)*

2. *Novelty*

- 2.1 The appealed decision found the subject-matter of the claims to be novel over documents (1) and (3). Novelty over document (3) was no longer contested during the appeal proceedings and the Board sees no reason to take a different view to the Opposition Division in this respect. However, the Appellant maintained its objection to novelty of the claimed invention with regard to document (1).

- 2.2 Document (1) (see last full paragraph on page 331 and Table 4) discloses the dehydration of glycerol to acrolein at 400°C and thus in the gas-phase over a cation exchanged H-ZSM-5 zeolite catalyst, both parties acknowledging that document (1) did not explicitly disclose the Hammett acidity of said zeolite.

- 2.2.1 The chemical composition of the H-ZSM-5 zeolite used in the gas-phase dehydration of glycerol of document (1) is given in Table 1 and is characterised by a  $\text{Na}_2\text{O}/\text{Al}_2\text{O}_3$  ratio of 0.40 mol/mol. This means that only about a

half of the sodium cations have been exchanged with H<sup>+</sup> ions, such that a significant proportion remain in a neutralised, and thus not in a strongly acidic form.

2.2.2 The experimental data filed by the Respondent (see point V above) show that the sodium content of an H-ZSM-5 zeolite has a considerable influence on its acidity and that H-ZSM-5 zeolites having a Na<sub>2</sub>O/Al<sub>2</sub>O<sub>3</sub> ratio similar to that disclosed in document (1) have a Hammett acidity outside the claimed range of -9 to -18. More particularly, the data show that ZSM-5 zeolites having a SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratio similar to that of document (1) but containing virtually no sodium (zeolites A and B) have a Hammett acidity of < -8.2, whereas those having a sodium content similar to the zeolite disclosed in document (1) (zeolites C and D) have a Hammett acidity of > -8.2 and thus outside the claimed range. Furthermore, the Respondent's data show that mixtures of each of the zeolites A to D with 20% bentonite, such a mixture being used in the process of document (1), all have a Hammett acidity of > -8.2.

2.2.3 The Appellant submitted that the H-ZSM-5 zeolite used in the process of document (1) had a Hammett acidity of between -16.12 and -8.2, as supported by the "Experimental Demonstration" filed with letter dated 3 May 2011.

However, the H-ZSM-5 zeolites tested by the Appellant were commercially available products characterised only by their SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratios. Thus it is not known whether they have a sodium content similar to the H-ZSM-5 zeolite of document (1), sodium content being crucial to the acidity of an H-ZSM-5 zeolite (see point 2.2.2 above). Nor is it known whether they were prepared by the method disclosed in document (1) (see page 329,

lines 10 to 20) and would for this reason have the chemical composition given in Table 1 of document (1). Thus, since the H-ZSM-5 zeolites tested by the Appellant are not necessarily the same as that used in document (1), and since H-ZSM-5 zeolites may have a Hammett acidity of  $> -9$  (see for example, zeolites C and D referred to in point 2.2.2 above), these experimental data cannot demonstrate that the catalyst used in the gas-phase dehydration of glycerol described in document (1) inevitably had a Hammett acidity of between  $-9$  and  $-18$ .

2.2.4 Thus, since document (1) does not directly and unambiguously disclose an H-ZSM-5 zeolite catalyst having a Hammett acidity of between  $-9$  and  $-18$  in a process for the gas-phase dehydration of glycerol, the subject-matter of claim 1 is novel over the disclosure of this document.

2.3 The Board therefore concludes that the subject-matter of claim 1 of the main request is novel within the meaning of Article 54 EPC.

### 3. *Inventive step*

3.1 The Board considers, in agreement with the Appellant and the Respondent, that the process of document (1) (see point 2.2 above) represents the closest state of the art and, hence, takes it as the starting point when assessing inventive step.

3.2 In view of this state of the art, the problem underlying the patent in suit (see paragraph [0015]), consists of providing a process for the gas-phase dehydration of glycerol wherein the catalyst



deactivates less quickly, and wherein higher acrolein yields and selectivity are achieved.

- 3.3 As the solution to this problem, the patent in suit proposes the process according to claim 1 of the granted patent, characterised by the Hammett acidity of the catalyst of between -9 and -18.
- 3.4 To demonstrate that the claimed solution achieves the alleged improvements, the Respondent relied upon the technical report filed with the letter dated 15 April 2014, upon which the Appellant did not comment. Said report shows that a process according to the invention employing an H-ZSM-5 zeolite having a Hammett acidity of  $< -12.0$  (zeolite A) was superior to processes differing therefrom only in that they employed an H-ZSM-5 zeolite having a Hammett acidity of  $> -8.2$  (zeolite C), whether it was mixed with bentonite (zeolite E) as in the process of document (1) or not, all H-ZSM-5 zeolites having the same  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio, differing only by virtue of their sodium content, and hence, Hammett acidity. More particularly, in the process using zeolite A, glycerol conversion at  $>95\%$  and acrolein yield at  $>70\%$  are steady over 6 hours, and the selectivity to acrolein is 72%. In contrast, in the processes using zeolites C and E, the zeolite deactivates and the acrolein yield drops more quickly, resulting in a glycerol conversions of 82 and 71% and acrolein yields of 52 and 39%, respectively, after 6 hours, the selectivity to acrolein also being lower than with zeolite A, namely only 63 and 55%, respectively. In view of said data, the Board is satisfied that the problem underlying the patent in suit has been successfully solved.

3.5 Finally, it remains to be decided whether or not the proposed solution to the objective problem underlying the patent in suit is obvious in view of the state of the art.

3.5.1 Neither document (1), nor any of the other cited prior art document teach that in the catalytic gas-phase dehydration of glycerol, increasing the acidity of the catalyst would lead to it deactivating more slowly, and to higher yields and selectivity of acrolein. On the contrary, document (4) (see claim 5 and col. 3, lines 66 to 67) specifically teaches to use catalysts with a Hammett acidity of between +2 and -8.2 for the gas-phase dehydration of glycerol to acrolein. The Board thus holds that contrary to the Appellant's assertion that the skilled person would inevitably arrive at the claimed subject-matter without any inventive efforts, it was not obvious for the skilled person to employ a catalyst having a Hammett acidity of between -9 and -18 in order to obtain acrolein in higher yields and selectivity.

3.6 For these reasons, the Board concludes that the subject-matter of claim 1, and by the same token that of dependent claims 2 to 9, involves an inventive step within the meaning of Articles 52(1) and 56 EPC.

*Auxiliary requests 1 to 3*

4. Since the subject-matter of the main request, namely the patent as granted, is novel and inventive for the reasons set out above, there is no need for the Board to decide on the lower ranking auxiliary requests.

**Order**

**For these reasons it is decided that:**

The appeal is dismissed.

The Registrar:

The Chairman:



C. Rodríguez Rodríguez

P. Gryczka

Decision electronically authenticated