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D E C I S I O N
of 5 October 2000

Case Number: T 0472/97 - 3.3.5

Application Number: 91116924.1

Publication Number: 0490037

IPC: B01J 20/18

Language of the proceedings: EN

Title of invention:
Adsorbent and cleaning method of waste gas containing ketonic organic solvents

Patentee:
Tosoh Corporation

Opponent:
Degussa-Hüls Aktiengesellschaft

Headword:
Zeolite/TOSOH

Relevant legal provisions:
EPC Art. 54(1), 83

Keyword:
"Novelty, main request - no, auxiliary request - yes"
"Sufficiency of disclosure - yes, skilled person can interpret and perform the invention in spite of some inconsistencies in the patent in suit"

Decisions cited:
-

Catchword:
-

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Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number: T 0472/97 - 3.3.5

D E C I S I O N
of the Technical Board of Appeal 3.3.5
of 5 October 2000

Appellant: (Opponent)
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Decision under appeal: **Interlocutory decision of the Opposition Division of the European Patent Office posted 5 March 1997 concerning maintenance of European patent No. 0 490 037 in amended form.**

Composition of the Board:

Chairman: R. K. Spangenberg
Members: G. J. Wassenaar
M. B. Günzel

Summary of Facts and Submissions

I. The appeal is from the decision of the Opposition Division to maintain European patent No. 0 490 037 in amended form with claims 1 to 6 filed during oral proceedings on 26 February 1997. Claim 1 thereof reads as follows:

"An adsorbent comprising zeolite with an amount of solid acid of not more than 0.05 mmol/g as determined by pyridine temperature programmed desorption method and with a SiO_2/Al_2O_3 molar ratio of not less than 50, provided that a zeolite having a SiO_2/Al_2O_3 molar ratio of 26,000 and a Na/Al molar ratio of 300 is excluded."

II. In the statement of the grounds of appeal, the appellant (opponent) attacked the claims as maintained by the Opposition Division on the grounds of unallowable extension (Article 123(2) EPC), insufficient disclosure (Article 83 EPC), lack of novelty (Article 54(1) EPC) and lack of inventive step (Article 56 EPC). With respect to lack of novelty, inter alia, reference was made to:

D2: ZEOLITES, 1990, Vol. 10, March, pages 205 to 206, and

D11: Materials Chemistry and Physics, 11 (1984), pages 515 to 523.

The relevant arguments can be summarized as follows:

The disclaimer in claim 1 offended Article 123(2) EPC because it was not based on the application as originally filed.

The invention as claimed was insufficiently disclosed for two reasons:

1. The description contained two incompatible methods for determining the amount of solid acid, and

2. The amount of solid acid in the claims was related to the amount of zeolite, whereas according to the description the amount of solid acid was determined for the adsorbent as such, which also comprised a binder.

The adsorbent according to claim 1 lacked novelty because D11, in particular Figures 4 and 5 revealed that, at temperatures above 300°C, H-Silicalite, with a Si/Al ratio of 10000, did not contain any substantial amount of adsorbed pyridine.

III. The respondent refuted the appellant's arguments. With respect to the novelty objection based on D11, the respondent argued that, because of its low aluminium content, H-Silicalite had indeed a low Lewis-acid content but, because of its hydrogen content a substantial amount of Brönsted-acid sites. To determine the total amount of solid acid, the pyridine TPD chromatograms as shown in Figure 3 must be integrated over the temperature range of 373 to 673 K. The amount of pyridine so determined clearly exceeded the maximum value of 0.05 mmol/g defined by present claim 1.

IV. During oral proceedings, which were held on 5 October 2000, the respondent submitted a new set of claims 1 to 3 as an auxiliary request. Claim 1 thereof reads as follows:

"A method of cleaning waste gas containing ketonic organic solvents comprising contacting the waste gas containing ketonic organic solvents with an adsorbent comprising zeolite with an amount of solid acid of not more than 0.05 mmol/g as determined by pyridine temperature programmed desorption method and with a SiO_2/Al_2O_3 molar ratio of not less than 50."

During the appeal proceedings, the appellant did not make further comments with respect to the subject-matter of the claims according to the auxiliary request. In the notice of opposition, inventive step of the subject-matter of the corresponding claims as granted was denied with reference to

D4: Journal of Catalysis, 118 (1989), pages 85 to 98.

V. The appellant (opponent) requested that the decision under appeal be set aside and that the European patent No. 0 490 037 be revoked.

The respondent (patentee) requested that the appeal be dismissed. As auxiliary request, the respondent (patentee) requested that the decision under appeal be set aside and the patent be maintained with the claims 1 to 3 of the auxiliary request submitted during the oral proceedings.

Reasons for the Decision

1. The appeal is admissible.

2. *Sufficiency of disclosure*

2.1 According to the claims, the amount of solid acid is determined by pyridine temperature programmed desorption (Py-TPD). Details of Py-TPD are not mentioned in the claims but indicated in the description of the patent in suit. In the patent in suit two different Py-TPD methods are described to determine the amount of solid acid; one in the more general part of the description (page 4, lines 41 to 58) and another, according to which the amount of solid acid in the adsorbents of the examples is determined (page 7, lines 5 to 14). The examples provide concrete solutions for the execution of the invention. The Board is therefore of the opinion that a skilled person will recognise that the method used in the examples is the relevant one to determine the scope of the claims. This opinion was confirmed by the respondent (letter dated 9 January 1998, point II, and orally during the oral proceedings). The description of the method indicated on page 7, lines 5 to 14 of the patent in suit is sufficiently clear to be performed by a person skilled in the art. Said method requires a desorption in the temperature range from 300 to 950°C. If the pyridine desorption is already completed below 950°C, as pointed out by the appellant, further heating to 950°C might be superfluous but has no impact on the practicability of the method. The acknowledged inconsistency in the method described on page 4, lines 41 to 58 (at room temperature pyridine cannot be adsorbed at the indicated pressure of 50-60 Torr) is a further indication that only the method indicated on page 7, lines 5 to 14 is the relevant one.

2.2 According to the claims the amount of solid acid refers to the zeolite in the adsorbents. This is in conformity with the description of the patent in suit (see page 2,

lines 42 to 45, page 3, lines 19 to 24, page 3, line 52 to page 4, line 4). According to the examples the amount of solid acid is determined by the Py-TPD method performed with the adsorbents, which also contain a binder such as clay and silica sol. Without evidence to the contrary, it may be assumed that the binder does not adsorb pyridine at temperatures above 300°C and that it does not interfere with the adsorption and desorption of the pyridine on the zeolite. Under these assumptions the measured amount of pyridine desorbed from the adsorbents is equal to the amount of pyridine desorbed from the zeolite in the adsorbents. Since the amount of zeolite in the adsorbents is known, the amount of solid acid per gram zeolite can be calculated. The Py-TPD method used in the examples is thus suitable to determine the amount of solid acid in the zeolite. For these reasons the Board holds that the appellant's objections raised under Article 83 EPC are not founded.

3. *Main request*

3.1 D11 discloses experimental data on the chemisorption and temperature programmed desorption of pyridine on H-ZSM-5 zeolites. Among these zeolites is H-Silicalite, an ZSM-5 zeolite with an extreme low aluminium content with a Si/Al ratio of about 10000. Figure 4 discloses that at an initial pyridine concentration of about 0.02 mmol/g H-Silicalite starts to desorb at a temperature of about 550 K. It further follows from said Figure that in order to have no desorption below 573 K (300°C) the initial pyridine concentration had to be considerably lower; at least below 0.01 mmol/g. It is further indicated in Figure 3 that the amount of pyridine adsorbed on H-Silicalite at 673 K (400°C) is

0.00 mmol/g. From these figures it follows that the amount of pyridine that can be desorbed from H-Silicalite by Py-TPD in the temperature range of 300 to 950°C cannot be more than about 0.01 mmol/g; ie far below the upper limit required by claim 1. This is further confirmed by Figure 5, which shows that the amount of pyridine adsorbed on H-Silicalite at 573 K (300°C) and higher is practically zero.

3.2 The Board cannot accept the respondent's view that the total amount of pyridine desorbed from H-Silicalite is much higher because H-Silicalite comprises hydrogen which forms Brönsted acid sites and that the total area under the curves disclosed in Figure 3 of D11 should be integrated in order to obtain the correct amount of desorbed pyridine. Integrating over the whole area would, however, imply that pyridine adsorbed at temperatures below 300°C had also to be taken into account, which would be contrary to the respondent's submission that the TPD method used to determine the acid concentration in the examples of the patent in suit has to be applied. Moreover, taking into account the fact that claim 1 does not specify the precise method according to which the pyridine desorption is to be carried out, the Board considers that, if the result of any suitable method for carrying out the pyridine desorption leads to a result below the limit stated in claim 1, the condition set out in that claim is met. It cannot be denied that the method used in the examples is such a method (see point 2.1 above). Therefore, for novelty it is only decisive how much pyridine can be desorbed from H-Silicalite at a temperature above 300°C or in other words, how much pyridine is left on a loaded H-Silicalite after heating to 300°C. Under these circumstances it is irrelevant whether or not H-

Silicalite contains a substantial amount of hydrogen. The Board agrees that in Figure 3 the area under the curve should be integrated to obtain the desorbed amount of pyridine, but for the desorption above 300°C the integration should start from 573 K. The remaining area is then minimal. The exact value cannot be derived from Figure 3 because the detector response is given in arbitrary units. From Figures 4 and 5, where the amount of pyridine is given in mmol/g, it follows unambiguously that the amount of pyridine on H-Silicalite present at temperatures above 300°C is far below 0.05 mmol/g. Thus, claim 1 lacks novelty so that the main request must be refused.

4. *Auxiliary request*

4.1 The auxiliary request comprises only method claims concerning the cleaning of waste gas containing ketonic organic solvents. The definition of the zeolite used thereby does not comprise the disclaimer contained in claim 1 of the main request. The objection under Article 123 EPC, based on that disclaimer, is thus no longer relevant. For the same reasons as given above with respect to the main request, the objections raised under Article 83 EPC are likewise not founded with respect to the subject-matter according to the auxiliary request.

4.2 During the opposition and appeal stage no document has been provided concerning problems relating to the cleaning of waste gas containing ketonic organic solvents. The only document which mentions the use of zeolites in combination with ketones is D2, concerning the base- and acid-catalysed cyclization of diketones over ZSM-5. It teaches that high silica ZSM-5

containing excess alkali ions can function as a catalyst for base-catalyst reactions. This clearly teaches away from the method of claim 1, because according to the invention it is essential that the zeolite only acts as an adsorbent and that all catalytic action should be suppressed (page 2, line 46 to page 3, line 5 of the patent in suit). There is thus no basis for a novelty or inventive step objection with respect to the claims of the auxiliary request. During oral proceedings the respondent has declared that apart from the objections raised with respect to the product claims of the main request no further objections are raised against the claims of the auxiliary request. Therefore, the finding in the decision under appeal that D4 does not relate to the problems encountered when adsorbing ketonic organic solvents, with which the Board agrees, stands uncontested. Under these circumstances there is no need to consider novelty and inventive step of the subject-matter of the claims according to the auxiliary request in any more detail.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the opposition division with the order to maintain the patent with claims 1 to 3 of the auxiliary request submitted during oral proceedings and a description to be adapted.

The Registrar: The Chairman:

S. Hue R. Spangenberg