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D E C I S I O N
of 2 February 2005

Case Number: T 0956/02 - 3.3.5

Application Number: 95941571.2

Publication Number: 0801027

IPC: C01B 39/36

Language of the proceedings: EN

Title of invention:

Rare earth-ZSM-5/ZSM-11 cocrystalline zeolite

Patentee:

CHINA PETRO-CHEMICAL CORPORATION, et al

Opponent:

Exxon Chemical Patents Inc.

Headword:

Zeolite mixture/CHINA PETRO-CHEMICAL

Relevant legal provisions:

EPC Art. 100(b)

Keyword:

"Sufficiency of disclosure (no) "

Decisions cited:

G 0004/95, T 0334/94

Catchword:

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Case Number: T 0956/02 - 3.3.5

D E C I S I O N
of the Technical Board of Appeal 3.3.5
of 2 February 2005

Appellants:

(Proprietors of the patent)

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Decision under appeal:

Decision of the Opposition Division of the
European Patent Office posted 18 July 2002
revoking European patent No. 0801027 pursuant
to Article 102(1) EPC.

Composition of the Board:

Chairman:

M. M. Eberhard

Members:

J. D. Schwaller

J. H. P. Willems

Summary of Facts and Submissions

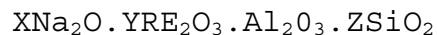
- I. This appeal lies from the decision of the opposition division revoking European patent No. 0 801 027 as granted on the grounds that it does not comply with the requirements of sufficiency of disclosure set out in Article 100(b) EPC.

Claim 1 as granted reads as follows:

"A rare earth-containing crystalline aluminosilicate zeolite, characterized by

(1) having a cocrystalline structure of ZSM-5 and ZSM-11, wherein the weight ratio of the moiety having the crystalline structure of ZSM-5 to the moiety having the crystalline structure of ZSM-11 is 0.1 to 10;

(2) having, in its originally synthesized anhydrous state, a chemical formula in terms of mole ratios of oxides as follows:



wherein $X = 0.1-10$, $Y = 0.01-1.0$, and $Z = 20-300$;

(3) having the X-ray diffraction pattern set forth in Table 1:

d(Å)	100 I/I ₀
11.2 ± 0.2	M
10.1 ± 0.2	M
6.05 ± 0.14	W
4.40 ± 0.08	W
3.86 ± 0.07	VS
3.72 ± 0.07	S
3.65 ± 0.07	W-M
2.01 ± 0.02	W
wherein: VS = 60-100 S = 40-60 M = 20-40 W < 20	

(4) having adsorption capacities towards n-hexane, cyclohexane and water of 9 to 11%, 3 to 5% and 4 to 8% by weight, respectively."

II. The opposition division held that the patent did not disclose any detailed method of determining the relative amounts of ZSM-5 and ZSM-11 in the claimed material. It further considered that, given the knowledge of the skilled person in this technical field, the X-ray diffraction data would be unable to provide such results. It concluded that, in the absence of any teaching whatsoever as to how the relative proportions of the two components were to be measured, the skilled person could not determine whether any specific product met the requirement (1) of claim 1 and therefore the requirements of Article 100(b) EPC were not satisfied.

III. In the grounds of appeal, the appellant submitted that the decision of the opposition division had relied on documents concerned with the identification of intermediate forms of ZSM-5 and ZSM-11 zeolites from a qualitative viewpoint, without taking into account the declaration by Professor Lin which, in its view, confirmed that a quantitative assessment could be done using an X-ray diffraction method.

Together with the grounds of appeal, the appellant (patentee) filed an extract (pp. 536-538 and 549-550) from a textbook published in 1974, namely "X-ray diffraction procedures for polycrystalline and amorphous materials", 2nd edition, by Harold P. Klug and Leroy E. Alexander (hereinafter D12) along with a second declaration by Professor Lin. The appellant argued that this second declaration showed the suitability of the polycrystalline X-ray diffraction phase method, a method well-known to the skilled person at the filing date of the patent as a way to quantitatively determine the amount of the respective moieties within the claimed compound.

The appellant concluded that it was clear, in view of this declaration, that the skilled person would have been able at the relevant date to conclude whether or not a product fell within the scope of protection of the patent in suit.

IV. In the reply to the grounds of appeal, the respondent (opponent) essentially followed two lines of argumentation. First of all, it reiterated that there was no measurement method in the patent as regards determining the weight ratio of the two zeolite

entities. Secondly, it stressed that the examples in the patent did not demonstrate that the products thus obtained were in fact mixtures of ZSM-5 and ZSM-11. They consisted of ZSM-5 only.

Along with its observations, the respondent filed a declaration by Professor Mortier. The respondent argued, *inter alia*, that the quantitative X-ray diffraction methods disclosed in D12 could be applicable to simple mixtures of components having distinct and well-defined internal structures. However, in the case of the complex zeolite system described in the patent, which included zeolites with very similar X-ray diffraction patterns, special precautions needed to be taken, especially as regards the reference compounds necessary for establishing the calibration curve. In particular, the reference samples had to have the same cations and rare earth content and the same hydration and cation exchange state as the material whose weight ratio was to be determined. The patent in suit gave no information on such reference compounds.

- V. In a letter dated 21 December 2004, the respondent requested the admission of oral submissions by a technical expert, Professor Mortier, at the oral proceedings scheduled for 2 February 2005.

- VI. On 28 January 2005, the appellant informed the parties by fax that it would not attend the oral proceedings. It furthermore gave arguments as to why the respondent's request concerning the hearing of oral submissions by a technical expert should be refused.

VII. Oral proceedings took place on 2 February 2005, in the absence of the appellant. The arguments put forward *inter alia* by the respondent during those proceedings were as follows:

- (a) Concerning the objection already raised in writing that the zeolites prepared in the examples in the patent in suit contained only ZSM-5, it submitted that, by comparing reference X-ray diffraction (XRD) diagrams of ZSM-5 and ZSM-11, no peak of ZSM-11 not coinciding with those of ZSM-5 could be found. This meant that for the sake of identification, the investigator had first to select certain "diagnostic peaks" characteristic of the ZSM-5 moiety only.

The respondent (referring to Professor Mortier's declaration) identified three "diagnostic peaks" at hkl of 232, 133 and 153 which corresponded, e.g. in Example 1, to $d(\text{\AA})$ values of 4.291, 3.647 and 2.965, respectively. He argued, on the one hand, that, when plotting the intensities of these three peaks for the four examples in the patent as a function of the amount of ZSM-5, a linear correlation should be found and, on the other hand, that by extrapolating each straight line, an intensity of 0 should be obtained for a zeolite mixture containing no ZSM-5. However, no such correlation existed as shown by the Figure on page 6 of Professor Mortier's declaration.

The respondent furthermore pointed out that the intensity variations for a particular d value in the four examples in Table 3 of the disputed

patent should in theory be lower for "non-diagnostic peaks" than for "diagnostic peaks", because the intensities of the latter were supposed to reflect the ZSM-5 amount variations in the mixture and thus should vary as a linear function of the ZSM-5 amount, contrary to the "non-diagnostic peaks". However, the variations for e.g. the "diagnostic peak" at $d(\text{\AA}) = 2.965$ were lower than for e.g. the "non-diagnostic peak" at $d(\text{\AA}) = 2.975$. The respondent concluded that the fluctuations observed at a specific d-value in Table 3 were essentially due to measurement errors and hence could not be correlated with the varying amounts of ZSM-5 in the examples. Therefore, no ZSM-11 at all was present in the zeolite mixtures disclosed in the examples.

- (b) The respondent further argued that the claimed subject-matter could not be prepared within the whole claimed range, since there was no teaching in the patent as to how the skilled person should proceed to prepare claimed ZSM-5/ZSM-11 mixtures other than those exemplified.
- (c) As regards the lack of method for determining the weight ratio between the zeolite entities, the respondent relied on the argumentation given in its written submissions and, in particular, on Professor Mortier's written declaration. As regards Professor Lin's second declaration, it emphasised that it was not explained in point 7.a thereof what kind of "pure ZSM-5" or "pure ZSM-11" should be taken to draw up the calibration curve. Furthermore the respondent contested that the

method according to steps a) to f) in point 7 of the said declaration belonged to common general knowledge.

VIII. The appellant requested that the decision of the opposition division be set aside and that the patent be maintained as granted or that the matter be remitted to the opposition division for further prosecution. The respondent requested that the appeal be dismissed.

Reasons for the Decision

1. The appeal is admissible.
2. *Admissibility of an oral presentation by the respondent's expert*

The appellant argued that the respondent's letter of 21 December 2004 concerning an oral presentation reached the appellant's representative's office too late, namely less than one month prior to the oral proceedings (it received the letter on 3 January 2005, the proceedings being scheduled for 2 February 2005). Therefore, according to the conditions established in decisions G 04/95 (OJ EPO 1996, 406) and T 334/94 (decision of 25 September 1997), the request to allow the expert's oral presentation should be refused.

The board cannot accept the appellant's arguments for the following reasons. As stipulated in Enlarged Board of Appeal decision G 04/95, point 10, such a request should be made "sufficiently in advance of the day appointed for oral proceedings so that all opposing

parties are able properly to prepare themselves in relation to the proposed oral submissions". According to decision T 334/94, such a request should, at a minimum, be presented one month before the oral proceedings, i.e. the last day by which further written submissions or sets of amended claims are generally admitted.

In the present case, since the appellant decided not to attend said proceedings, the question whether or not it had sufficient time to properly prepare itself in relation to the oral presentation of the expert is irrelevant. Furthermore, the respondent's request was made more than one month before the date of the oral proceedings and thus in accordance with the minimum time limit stated in decision T 334/94. In these circumstances, the board did not see any reason to refuse the hearing of the respondent's expert.

3. *Sufficiency of disclosure*

3.1 With respect to the point raised by the respondent that the examples in the disputed patent disclosed compounds containing only the ZSM-5 entity, the question arises whether a mixture of both zeolite moieties was actually obtained in the examples. However, this question can remain open, since the requirements of Article 100(b) EPC are not satisfied for other reasons (see below).

3.2 For the sake of argument, it is assumed in the present decision that the two zeolite entities are present in the material under investigation.

- 3.3 Claim 1 of the disputed patent is defined by, *inter alia*, the technical feature: **"wherein the weight ratio of the moiety having the crystalline structure of ZSM-5 to the moiety having the crystalline structure of ZSM-11 is 0.1 to 10"** (emphasis added).

Since the skilled person reading the specification should be able to carry out the invention in all its essential aspects, and should in particular be able to verify whether he has succeeded in producing the claimed product, the board has to examine whether a method for measuring this ratio is indicated in the patent or, if this is not the case, whether the missing information could be supplied from common general knowledge.

- 3.4 Apart from the statement at paragraph [0019] that "the unit cell parameters and the weight ratio of the moiety having the crystalline structure of ZSM-5 to the moiety having the crystalline structure of ZSM-11 of the zeolites product were determined by X-ray diffraction method", the disputed patent discloses no further information as to how the quantitative determination of the above ratio may be carried out.

Textbooks or general technical literature form part of common general knowledge (Case Law of the Boards of Appeal, 4th edition 2001, p. 145). As the book by Klug and Alexander concerning X-ray diffraction procedures for polycrystalline and amorphous materials quoted in the appellant's expert declarations is regarded as such a reference book, D12 is considered to illustrate common general knowledge. D12 shows at page 549 and Figure 7-5 that XRD diagrams may be used to

quantitatively determine the amounts of individual crystalline components present in a mixture by drawing up a calibration curve and measuring the height or the integrated area of selected diffraction peaks, the said measured data being proportional to the amount of crystalline substance present in the mixture.

It is true that D12 (pp. 549-550) teaches, on the basis of an indirect analysis making use of an internal standard and of a calibration curve, how to perform such an analysis for mixtures containing crystalline components having separate easily assignable peaks, such as quartz with fluorite or quartz with either Al_2O_3 , MgO , CaCO_3 or bentonite.

However, D12 does not give any information as to how a quantitative analysis of mixtures of components having interfering peaks can be carried out, let alone how such an analysis can be done when all the peaks of one moiety are included in the XRD diagram of the other, which is the case for ZSM-5/ZSM-11 mixtures.

- 3.5 The question thus arises whether such information, in particular that contained in Professor Lin's declarations, may be considered as common general knowledge.

In his first declaration, Professor Lin submitted on page 7 that i) "the polycrystalline XRD pattern of the zeolite is formed by linear addition according the weight fractions of the two phases" and ii) "when CuK_α is used as a radiation source, ZSM-5 has a medium strength diffraction peak near $d = 3.65 \text{ \AA}$ in the range $2\theta < 40^\circ$ while ZSM-11 has not". On page 6 of the same

declaration, it is further stated that "the quantitative analysis of the ZSM-5 and ZSM-11 phases is a common technique for phase quantitative analysis" and reference is made to the Klug and Alexander book.

No further information, however, is given as to how the investigator can arrive at the determination of the ZSM-5/ZSM-11 weight ratio.

- 3.6 In paragraph 4 of his second declaration, the same expert identifies several diffraction peaks in the ZSM-5 pattern that have no corresponding peak in the ZSM-11 pattern, explaining that on that basis the identification of ZSM-5 or ZSM-11 or their combination is rendered possible.

The board notes in this respect that the presence of differentiating peaks indeed makes it possible to diagnose the presence of the ZSM-5 moiety, but since all the XRD peaks in the ZSM-11 diagram are also part of the ZSM-5 XRD diagram, this does not make it possible to conclude the presence of ZSM-11.

In paragraph 6 of his declaration, Professor Lin explains that one basis for a quantitative determination of the relative zeolite amounts would be to take the peak at $2\theta = 13.96^\circ$ (which corresponds to $d = 6.34 \text{ \AA}$) as a distinguishing peak for the ZSM-5 entity, since ZSM-11 has its nearest peaks at $2\theta = 13.30^\circ$ and 14.61° , respectively.

In paragraph 7, starting from the teaching of D12, the expert indicates the experimental protocol which, in his opinion, would render possible the quantitative

determination of ZSM-5 by an XRD method using the known internal-standard analysis. According to point 7 of the declaration, this method includes preparing five samples containing different known weight ratios of pure ZSM-11 and pure ZSM-5 zeolites by mechanical mixing.

After adding α -Al₂O₃ as an internal standard to all these samples, an XRD pattern for each sample is taken. Then the intensities of the 012 diffraction peak of ZSM-5 and the 116 diffraction peak of α -Al₂O₃ are calculated from the XRD patterns and the intensities $I_{\text{ZSM-5};012}/I_{\alpha\text{-Al}_2\text{O}_3;116}$ are plotted as a function of the weight fraction of the ZSM-5 in the mixture, thereby obtaining a straight line as the calibration curve.

An X-ray diffraction pattern is then generated with a sample of the unknown ZSM-5/ZSM-11 mixture, to which the same internal standard is added.

Based on the intensity ratio $I_{\text{ZSM-5};012}/I_{\alpha\text{-Al}_2\text{O}_3;116}$ thus measured, the content of ZSM-5 in the unknown sample is determined using the calibration curve and, by subtracting the amount of ZSM-5, the amount of ZSM-11 is obtained.

The respondent, however, contested that this quantitative method including steps a) to f) was well-known before the priority date for zeolite mixtures having similar XRD patterns, such as ZSM-5 and ZSM-11.

The appellant provided no evidence to show that this quantitative method was usually applied to this kind of

mixtures, although the burden of proof rests with him. In any case it cannot be inferred from D12 that the internal-standard analysis disclosed therein would be suitable for mixtures including zeolites with similar XRD patterns. In these circumstances, the board is not convinced that the method described in the declaration formed part of common general knowledge before the priority date of the disputed patent.

- 3.7 The board further notes that Professor Lin proposes the use of "pure ZSM-5" and "pure ZSM-11" for the calibration. As pointed out by the respondent and in Professor Mortier's declaration (see page 3), when applying this method to zeolites it must be ensured that the samples used for calibration purposes are prepared from reference zeolites having the same or a very similar chemical composition as the zeolites mixture under investigation, in terms of cation and template contents and in terms of hydration and cation exchanged states. As further explained on page 3 of Professor Mortier's declaration, this is because the intensities of the diffraction peaks are determined by the unit cell content and are therefore greatly influenced by the presence or absence of templates or cations (in the present case the diamine as the template and the rare earth element as the cation) in said unit cell and by the hydration state of the zeolite (i.e. whether the zeolite is in the calcined form or not).

The effect on peak intensity of the hydration state and the presence of a template in the zeolite unit cell can be shown e.g. by comparing the calculated XRD diagram of a tetrapropylammonium ZSM-5 (e.g. on page 10 of

Professor Mortier's declaration) with that of a calcined ZSM-5 (e.g. as annexed to Professor Lin's declaration), from which comparison it can be seen that the most intense peak for the calcined material is at $d = 11.13 \text{ \AA}$, whereas in the case of the hydrated material containing tetrapropylammonium as the template, the most intense peak is at $d = 3.84 \text{ \AA}$.

Since the intensities of the peaks vary greatly with the unit cell content, it is necessary to take into account all these parameters when preparing the reference samples used for the calibration. If the cations, the templates, the hydration state and/or the cation exchange state of the reference zeolite differ from those of the zeolites in the mixture to be tested, the results would not be representative of the actual weight ratio of the compounds in the mixture and would be meaningless, as pointed out by the respondent in its letter of 29 July 2003 (pages 5-6). This was not contested by the appellant.

- 3.8 Furthermore, the patent in suit only describes the preparation of a rare earth-containing zeolite having a cocrystalline structure of ZSM-5 and ZSM-11 by direct synthesis, i.e. from a gel obtained from a mixture containing all the zeolite precursors including the rare earth compounds. It is, however, silent about the chemical composition, in particular as regards the kind and content of the rare earth element and template and as regards the hydration and ion-exchange state of the individual reference zeolites suitable for establishing the calibration curve. Therefore the skilled person is left without any guidance in this respect, since not only information is missing as to the characteristics

of the reference zeolites to be used for the calibration curve but also with respect to their method of preparation.

Although from paragraph [0004] of the patent in suit it appears that the direct synthesis (i.e. without use of an ion exchange operation) of a ZSM-5 containing rare earth element is known from the patent literature cited therein, the situation is different for the rare earth-containing ZSM-11, since no reference to published documents concerning the preparation of such a zeolite is made in the patent in suit.

In the board's judgment, in the absence of any guidance in the patent in suit as regards both the characteristics and method of preparation of the rare earth-containing reference zeolites suitable for the calibration curve and for a meaningful determination of the weight ratio of the zeolite moieties in the claimed product or in the examples in the disputed patent, the determination would require further experimentation and efforts involving more than routine experimentation and would thus amount to an undue burden for the skilled person.

3.9 Taking into account the fact that not only important information concerning the disputed X-ray diffraction based method is missing in the patent in suit, in particular:

- how a quantitative analysis can be performed for zeolitic entities having very similar X-ray powder diffraction patterns,

- which reference zeolites are supposed to be used for the calibration curve,
- how the said reference zeolites are to be prepared,

but also that the missing information cannot be considered as being comprised within the common general knowledge available to the skilled person at the reference date of the patent in suit, and that the further experimentation required for determining the missing data would represent an undue burden for the skilled person, the board concludes that the disclosure in the patent in suit is not sufficiently clear and complete for the skilled person to determine the weight ratio of the zeolite entities defined in the claims and in the examples and thus to carry out the invention.

The patent in suit therefore does not meet the requirements of sufficiency of disclosure set out in Article 100(b) EPC.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:

The Chairman:

A. Wallrodt

M. Eberhard