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**Datasheet for the decision
of 20 September 2021**

Case Number: T 2787/18 - 3.3.06

Application Number: 11806374.2

Publication Number: 2593222

IPC: B01J29/56, B01J29/80,
B01J37/02, B01D53/94

Language of the proceedings: EN

Title of invention:

COPPER CONTAINING ZSM-34, OFF AND/OR ERI ZEOLITIC MATERIAL FOR
SELECTIVE REDUCTION OF NOX

Patent Proprietor:

BASF SE

Opponents:

Umicore AG & Co. KG

Headword:

BASF/ZSM-34

Relevant legal provisions:

EPC Art. 56

RPBA Art. 12(4)

Keyword:

Inventive step - (no)

Late-filed evidence - request could have been filed in first instance proceedings (yes)

Decisions cited:

T 1467/13

Catchword:



Beschwerdekammern

Boards of Appeal

Chambres de recours

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Case Number: T 2787/18 - 3.3.06

D E C I S I O N
of Technical Board of Appeal 3.3.06
of 20 September 2021

Appellant:

(Patent Proprietor)

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Decision under appeal:

**Decision of the Opposition Division of the
European Patent Office posted on 11 October 2018
revoking European patent No. 2593222 pursuant to
Article 101(3) (b) EPC.**

Composition of the Board:

Chairman J.-M. Schwaller
Members: S. Arrojo
 R. Cramer

Summary of Facts and Submissions

- I. The appeal filed by the proprietor (the appellant) is directed against the decision of the opposition division to revoke European patent No. 2 593 222 for non-compliance with the requirements of Article 56 EPC.
- II. With its statement of grounds of appeal dated 20 February 2019, the appellant contested the decision and submitted ten sets of amended claims as main and first to ninth auxiliary requests.
- III. In its reply, opponent (also respondent) requested to dismiss the appeal and raised objections under novelty and inventive step.
- IV. In response to the board's preliminary opinion that auxiliary request 3 appeared to comply with the requirements of the EPC, the appellant with a letter dated 22 July 2021 requested to maintain the patent on the basis of auxiliary request 3 as its new main request, or alternatively, on the basis of one of auxiliary requests 5 to 9, renumbered as auxiliary requests 1 to 5.

Claim 1 of the new **main request** reads:

"A copper containing ZSM-34 zeolitic material having a silica to alumina mole ratio ranging from 10 to 15 and a copper content, reported as CuO, ranging from 1 to 10 wt.-%, based on the total weight of the calcined zeolitic material, and having an alkali metal content, reported as metal oxide, ranging from 1.5 to 12 wt.-%."

Claim 1 of new **auxiliary request 1** (formerly fifth auxiliary request) corresponds to that of the main request, with the alkali metal content being further restricted to the range "1.5 to **5 wt.-%**".

- V. At the oral proceedings held on 20 September 2021 the discussion focused on inventiveness of the main and first auxiliary requests and on the admissibility of auxiliary requests 2 to 5. The enabling disclosure of documents D1 and D14, disputed originally by the patent proprietor, was no longer contested. At the closure of the debate, the final requests of the parties were the following:

The appellant requested to set aside the decision under appeal and to maintain the patent on the basis of the main request filed as 3rd auxiliary request on 20 February 2019, or as an auxiliary measure, on the basis of one of auxiliary requests 1 to 5, respectively filed as fifth to ninth auxiliary requests on 20 February 2019.

The respondent requests to dismiss the appeal.

- VI. To facilitate the readability of this decision, the board will use the following abbreviations:

ZSM-34: Zeolite with framework of ZSM-34;

Cu/ZSM-34: Zeolite with framework of ZSM-34 containing copper and potassium;

ERI: Zeolite with framework of erionite;

OFF: Zeolite with framework of offretite;

Cu/KOFF: Zeolite with framework of offretite containing copper and potassium;

SCR: Selective catalytic reduction.

Reasons for the Decision

1. Main request - Inventive step

The board has concluded that the requirements of Article 56 EPC are not met for the following reasons:

1.1 Document **D1** (W. Arous et al., "*Selective catalytic reduction of nitric oxide with ammonia on copper (II) ion-exchanged offretite*", Catalysis Communications 6, 2005) discloses (see table 1) Cu/KOFF catalysts for SCR including an amount of potassium falling within the claimed range.

Document **D10** (WO 2008/132452 A2) discloses (example 2) a Cu/ZSM-34 catalyst for SCR.

1.2 Closest prior art

1.2.1 The respondent considered that any one of documents D1 and D10 could be regarded as the closest prior art, because the patent interpreted the meaning of ZSM-34 (see par. [0033]) in such broad terms, that the OFF materials disclosed in D1 would also fall within the scope of the ZSM-34 according to the invention.

1.2.2 The board disagrees with the respondent in this respect, because there is no reason to rely on the content of the description to interpret a clear feature such as "ZSM-34", which is a known zeolite structure. Thus, while OFF and ZSM-34 are considered to be closely related, they are not the same zeolites.

Therefore, document D10 and more particularly its example 2, is considered to represent the most promising springboard, because it is structurally closer (it uses the same zeolite) to the invention than document D1.

Claim 1 differs from this closest prior art in that the alkali metal content ranges from 1.5 to 12 wt.-% and in that the silica to alumina mole ratio ranges from 10 to 15. It should however be noted that these features are not necessarily different in D10, but simply not specified in its example 2.

1.3 Problem solved by the invention

- 1.3.1 According to the patent in suit (par. [0027]), the invention intends to solve the problem of providing *"cost-effective hydrothermally stable catalysts for SCR applications"*, which *"show a high activity over a wide temperature range"*.

The examples in tables 2-6 of the contested patent compare the NO conversion under different temperatures and for different ageing conditions between catalysts falling within the scope of protection (examples A-D) and embodiments containing alkali metal concentrations lower than the claimed range (examples E-F).

- 1.3.2 The appellant argued that examples A-F of the patent demonstrated that the above problem was surprisingly solved by the differentiating features of the invention.

The respondent counter-argued that the examples differed from one another in several aspects, so it was not possible to associate the observed effects with the

alkali metal content in particular, as these could be related to any of the other differences. Furthermore, the comparative examples E and F were not representative of the zeolites disclosed in the prior art, because the concentration of alkali metal was very low and appeared to be the result of multiple steps of ammonium exchange and calcination. In any case, the observed results showed that the catalysts containing more alkali metal had a worse performance after being aged, and there was little added value in improving the NO conversion only for fresh catalysts. Consequently, the problem solved by the invention was simply that of providing an alternative zeolitic material.

- 1.3.3 While the board tends to agree with the respondent in that the comparative examples represent an unrealistic reference point, it will be assumed for the sake of the argument (in the appellant's favour) that the tests in the patent provide a valid comparison with respect to the closest prior art.

A first comparison between the examples according to the invention (A-D) and the comparative tests (E-F) indicates that fresh catalysts containing higher amounts of alkali metals clearly lead to higher NO conversion when similar or even lower amounts of copper are used. However, the results for aged catalysts are less consistent, showing improved conversions at 200°C after ageing at 750°C, but slightly worse results at 450°C after the same ageing process. Concerning the catalysts aged at 800°C, while the performance of example B is similar despite the lower copper concentration, it is clearly worse for the other examples including higher alkali metal concentrations (examples A, C and D) at all temperatures.

The board considers that, in view of the inconsistencies in the results obtained under different ageing and temperature conditions, there is no basis to conclude that the invention provides the effects of increasing the hydrothermal stability and/or of showing high activity over a wide temperature range throughout the entire scope of protection. For instance, in examples A, C and D the NO conversions are clearly worse for all temperatures after the catalyst is aged at 800°C.

On the other hand, while it is clear that operating within the defined ranges of CuO and K₂O concentrations does not guarantee optimal performance over a broad range of ageing and temperature conditions, the results show that the CuO and K₂O concentrations can be optimised to ensure a good performance of the catalyst under specific temperature and ageing conditions (e.g. example B for 800°C aged catalysts at 450°C). This conclusion is in-line with par. [0028] of the patent (emphasis added by the board), which reads:

"Surprisingly, it was found that Cu/ZSM-34 catalysts containing high alkali metal contents can exhibit high performance even after severe hydrothermal aging when the Cu content is carefully controlled". Thus, by controlling the copper and alkali metal contents, the performance of the catalyst can be maximised for specific ageing/temperature conditions without necessarily increasing or even decreasing the copper consumption. Since it is well-known that metals are an expensive part of catalysts in general, the optimisation of the alkali metal/CuO concentrations is considered as a way of obtaining cost-effective catalysts for SCR applications.

It is thus concluded that the invention solves the problem of providing cost-effective catalysts for SCR applications.

1.4 Obviousness

- 1.4.1 Document D1 is a scientific article which relates to selective catalytic reduction of NO with Cu/KOFF catalysts treated with a predetermined number of copper exchange steps. According to this document (see abstract; page 284, left column, last lines; §4 "Conclusion" and figure 3), the NO conversion of the catalyst increases with the amount of added copper only up to a certain level. Further exchange steps do not lead to significantly higher conversions because once copper ions have been retained in the main channels formed by gmelinite cages, potassium ions will block the access to the cancrinite cages.

Document **D14** (same authors, *"Catalytic activity of Cu-offretite catalysts prepared by solid state ion exchange in the reduction of NO with NH₃"*, Studies in Surface Science and Catalysis, Vol. 15, 2005) discloses the same concept (see abstract and §3 "Results and discussion").

- 1.4.2 The appellant argued that the subject-matter of claim 1 at issue was not obvious in view of the combination of example 2 of D10 and the teachings in D1 or D14, because Example 2 of D10 taught away from the solution in claim 1, since it included ammonium exchange steps for removing potassium from the zeolite, which, as indicated in table 1 and par. [0015] of the patent, would lead to alkali metal concentrations lower than the claimed range.

Furthermore there was no reason to consider the teachings of documents D1 and D14 for solving the underlying technical problem, because these documents related to OFF, which was both structurally and functionally different from the ZSM-34 in example 2 of D10. The appellant referred in this respect to figures 10 and 11 of **D21** (Lillerud and Raeder, "*On the synthesis of erionite-offretite intergrowth zeolites*", Zeolites, Vol. 3, 1986), and observed that the diffractogram of ZSM-34 did not show some of the peaks formed by ERI, which implied that ZSM-34 could not be considered as a simple addition of OFF and ERI. Furthermore, col. 1, line 67 to col. 2, line 10 of **D18** (US 4 086 186 A1) explicitly indicated that while ZSM-34, ERI and OFF had similarities, ZSM-34 was distinguished from the others by certain chemical properties.

Finally, even if the contents of D1 or D14 were taken into account, the teachings relating to the role of potassium in blocking the cancrinite cage were not supported by evidence. The only experimental comparison (shown in figure 3 of D1) was not carried out using a Cu/KOFF with low amounts of alkali metal as comparator, but a different catalyst structure (Cu(76)NaY). The conclusions in these documents should therefore be regarded as mere speculative allegations and not as actual teachings.

- 1.4.3 The board disagrees with the above arguments and considers that a skilled person starting from example 2 of D10 as closest prior art, would consider the teachings of D1 or D14 for solving the underlying technical problem for the following reasons:

- While the board agrees with the appellant in that the ammonium exchange in example 2 of D10 will tend to reduce the amount of alkali metal in the zeolite, this does not imply that D10 teaches away from zeolites having an alkali metal in the range defined in claim 1 at issue, because the invention is not based on maximising the amount of alkali metal in the zeolite but on limiting the ion exchange steps to ensure that there is enough alkali metal in the zeolite to occupy the cancrinite cage and block the access to the copper ions (see par. [0030] of the patent). In any case, the ammonium exchange in example 2 of D10 does also not imply that the alkali metal content is decreased to a very low level, but simply that the alkali metal should be at least partially exchanged by copper (i.e. first ammonium and then copper), an idea which is not in contradiction with the invention. In fact, as indicated in par. [0019] of the patent, while the invention does not require an ammonium exchange step, it includes an alkali metal/copper ion-exchange process. Furthermore, as argued by the respondent, zeolites can have concentrations of potassium which are significantly higher than those shown in table 1 of the patent (for example, the potassium concentration of the KOFF in table 1 of D1 is 10.2 wt.-% when expressed as K_2O , which is much higher than the 2.29 wt.-% in table 1 of the patent), so there is no basis to conclude that the potassium concentration after an ammonium exchange step would necessarily fall outside the claimed range.

- As furthermore indicated in point 1.2.2 above, ZSM-34 and OFF are not the same zeolites, but they are closely related. According to tables 4 and 7 of D21, ZSM-34 is formed by major portions of OFF intergrowths. Similarly, D18 indicates (emphasis added by the board) that "*ZSM-34 is ... an intergrowth of very small*

erionite domains throughout an offretite structure" (col 2, lines 60-62). The diffractograms cited by the appellant (i.e. figures 10 and 11 of D21) also appear to indicate that ZSM-34 and OFF are structurally close, because the seven peaks corresponding to OFF overlap almost exactly with those of ZSM-34, with the latter including just one additional peak of smaller size (probably attributable to the minor portions of ERI intergrowth). In any case, the relevant question is not whether OFF and ZSM-34 are the same but rather whether they are similar or comparable in those aspects which might be of relevance when reading the teachings of D1 and D14, namely the indication that the cancrinite cage of the OFF structure retains alkali metals and blocks the access to copper ions to these sites. Since it is not contested that cancrinite cages are an important part of both ZSM-34 and OFF, the skilled man would expect the above teachings of D1 and D14 to be applicable to the cancrinite cages of ZSM-34 in D10, so he would take the teachings of these documents into account for solving the underlying technical problem.

- Concerning the argument that the teachings in D1 and D14 would be invalid, the board notes that the appellant appears to equate an (alleged) lack of evidence for a certain teaching with a non-enabling disclosure. The two are however different, because a direct and unambiguous disclosure can normally be implemented without the need of supporting experiments, unless it involves allegations which go directly against well established knowledge (which is not the case in D1 or D14). In any case, the board also notes that at least document D1 includes sufficient evidence to support its conclusions (see next point 1.4.4).

1.4.4 The tests in document D1 compare several Cu/KOFF catalysts which have been exposed to a different number of copper exchange steps and (thereby) include different concentrations of potassium and copper (see table 1 and figure 3, wherein the sub-index "n" in "Cu-KOFF_n" represents the number of copper exchange steps). As explicitly indicated on page 284, left col., last lines: *"Fig. 3 shows that the increase of copper exchange number is not necessary since the deNO_x efficiency is not improved after the second run"*. These observations lead to the conclusion that (see page 284, right col.) *"After the first exchange, almost all the remaining K cations are in cancrinite cages... and they cannot be easily exchanged by copper ions"*, and that (see page 285, left col.) *"Therefore, we could conclude that (i) copper ions are retained in the main channels formed by gmelinite cages and (ii) K ions "blocks" the cancrinite access to copper ions and prevent any copper migration"*.

Thus, contrary to the appellant's allegations, document D1 supports its conclusions by comparing the NO conversion obtained with Cu/KOFFs having different concentrations of copper and potassium, and indicates that an optimum catalytic performance can be obtained using only a few copper exchange steps. When reading these passages, the skilled person would recognise that by limiting the copper exchange steps to a point in which the remaining potassium occupies the cancrinite cage would represent an optimum solution for obtaining a cost-effective catalyst, because this would reduce the copper consumption while maintaining the catalytic performance.

Therefore, when starting from example 2 of D10 and in view of the content of document D1, the skilled man

would solve the underlying technical problem of providing cost-effective catalysts for SCR applications by controlling the potassium concentration after each ion exchange (be it ammonium exchange or copper exchange), so as to ensure that the final alkali metal concentration of the Cu/ZSM-34 is such that potassium only occupies the sites on the cancrinite cages, because he is aware that further addition of copper would increase the manufacturing costs without improving the catalyst performance. In view of the results in figure 3 of D1, this optimum point might be reached after one, two or three copper exchanges, at alkali metal concentrations between 4,05 and 5,18 wt.-% (corresponding to a range of 4,89 to 6,25 wt.-% measured in K₂O basis). The combination of example 2 of D10 with the teachings of D1 therefore leads to a Cu/ZSM-34 catalyst with an alkali metal concentration falling within the claimed range.

- 1.4.5 The additional differentiating feature, namely the silica to alumina ratio ranging from 10 to 15, has not been linked to any specific technical effect, so its only contribution is that of providing an alternative.

This range is nonetheless common for ZSM-34 catalysts, as apparent from the 2nd table of col. 4 (preferred silica/alumina ratio of 10-55) and table 4 of D18 (silica/alumina of examples 2 and 3 equal to 13.4).

The board therefore concludes that working within the claimed silica to alumina ratio of 10 to 15 would represent a trivial (or at least obvious) alternative for a person skilled in the art.

- 1.4.6 In view of the above argumentation, the board concludes that the subject-matter of claim 1 is not inventive in

view of the combination of example 2 of D10 and the teachings of D1 and D18.

2. Auxiliary request 1 - Inventive step

2.1 The subject-matter of claim 1 of this request corresponds to that of the main request with the sole difference that the alkali metal content is restricted to a range of 1.5 to 5 wt.-%.

2.2 Since the optimal alkali metal values according to D1 include at least one example falling within this restricted range (Cu/KOFF₃ having an alkali metal concentration expressed as K₂O of 4,89 wt.-%), the same arguments and conclusions as presented for the main request apply to this request.

2.3 The subject-matter of claim 1 is therefore not inventive in view of the combination of example 2 of D10 and the teachings of D1 and D18.

3. Auxiliary requests 2-5 - Admittance

3.1 Since these requests were filed as auxiliary requests 6 to 9 with the statement of grounds of appeal and considering that none of them were filed during first instance proceedings, their admittance is governed by Article 12(4) RPBA 2007.

3.2 It is noted that claims 3 and 5-8 as granted and as originally filed were drafted using conditional features, i.e. defining different restrictions for different degrees of ageing or for different silica to alumina ratios. In the requests at issue, all the conditional features have been amended by deleting the conditions but not the associated restrictions.

3.3 The appellant argued that these requests should be admitted because they were a reaction to the conclusions presented by the opposition division in the decision under appeal.

3.4 The board notes that during opposition proceedings several auxiliary requests were filed including combinations of claim 1 as originally filed with some of these conditional features. The opposition division argued (see §5.4.1.3 of the appealed decision) that when the conditions were not fulfilled the restrictions associated therewith did not apply. Since the conditions were rather specific, it followed that these features did not restrict the subject-matter for most of the embodiments falling within the claims.

The board considers that the deletion of the conditional features cannot be seen as an attempt to overcome objections presented during the first instance, because the opposition division did not consider the conditional features to be unallowable, but simply to be insufficient (when combined with claim 1) for establishing patentability.

3.5 Consequently, these requests represent an attempt to pursue embodiments going beyond those which were presented and defended in front of the first instance (analogous to decision T 1467/13, reason 3.1). The board therefore concludes that auxiliary requests 2 to 5 at issue could and should have been filed during the first instance proceedings, and thus exercises its discretion under Article 12(4) RPBA-2007 not to admit them into the appeal proceedings.

3.6 As an addendum and for the sake of completeness, the board notes that since the deleted conditional features

were part of the dependent claims as filed and as granted, the amendments would have likely given rise to additional problems under Articles 123(2) and/or (3) EPC.

4. Since none of the requests presented by the appellant is considered to be admissible and allowable, the proprietor's appeal does not succeed.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:

The Chairman:



A. Pinna

J.-M. Schwaller

Decision electronically authenticated