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Datasheet for the decision of 22 May 2007

Case Number: T 0632/03 - 3.3.07

Application Number: 98306542.6

Publication Number: 0900592

IPC: B01J 23/881

Language of the proceedings: EN

Title of invention:
Mo-Bi-Fe-Ca based catalyst for the manufacture of acrylonitrile and hydrogen cyanide

Applicant:
Innovene USA LLC

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 56

Keyword:
"Inventive step (no) - problem and solution"

Decisions cited:
T 1067/97

Catchword:
-

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Case Number: T 0632/03 - 3.3.07

D E C I S I O N
of the Technical Board of Appeal 3.3.07
of 22 May 2007

Appellant: Innovene USA LLC
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Decision under appeal: **Decision of the Examining Division of the European Patent Office posted 7 February 2003 refusing European application No. 98306542.6 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: S. Perryman
Members: B. Struif
F. Rousseau

Summary of Facts and Submissions

I. The appeal lies from the decision of the examining division refusing European patent application 98 306 542.6 (published as EP-A-0 900 592), having a filing date of 17 August 1998 and claiming a priority date of 2 September 1997 (US 92 38 78). The application as filed comprised ten claims. Independent Claim 1 read as follows.

"A catalyst composition comprising a complex of catalytic oxides of iron, bismuth, molybdenum and calcium and characterised by the following empirical formula:

$$A_aB_bC_cD_dFe_eBi_fMo_{12}O_x$$

where

A = one or more of Li, Na, K, Rb and Cs or mixtures thereof

B = one or more of Mg, Mn, Ni, Co, Ag, Pb, Re, Cd and Zn or mixtures thereof

C = one or more of Ce, Cr, Al, Sb, P, Ge, La, Sn, V and W or mixtures thereof

D = one or more of Ca, Sr, Ba or mixtures thereof

and

a = 0.01 to 1.0; b and e = 1.0 - 10; c, d and f = 0.1 to 5.0 and x is a number determined by the valence of requirements of the other elements present."

II. The decision of the examining division was based on claims 1 to 10 filed with letter of 4 March 2002 as the

sole request in view of *inter alia* the following documents:

D1: EP-A-0 685 260
D2: EP-A-0 239 071
D3: US-A-4 873 217

Amended claim 1 reads as follows:

"A catalyst composition comprising a complex of catalytic oxides of iron, bismuth, molybdenum and calcium and characterised by the following empirical formula:

$$A_aB_bC_cD_dFe_eBi_fMo_{12}O_x$$

where

A = one or more of Li, Na, K, Rb and Cs or mixtures thereof
B = one or more of Mg, Mn, Ni, Co, Ag, Pb, Re, Cd and Zn or mixtures thereof
C = one or more of Ce, Cr, Al, Sb, P, Ge, La, Sn, V and W or mixtures thereof
D = **Ca, optionally** one or more of Sr, Ba or mixtures thereof

and

a = 0.01 to 1.0; b = 1.0 - 10; c, d and f = 0.1 to 5.0**; e = 1.7 to 10** and x is a number determined by the valence of requirements of the other elements present." (Emphasis added by the Board to indicate the differences vis-à-vis claim 1 as filed).

III. The examining division held that:

(a) Claim 1 was not novel over Example B-6 of D2 when taken into account the teaching of the whole document. Although the composition of example B-6 did not disclose the amount of Fe to be 1.7 to 10, the general amount of Fe could be up to 3 so that the claimed subject-matter was met. These arguments also applied to D3, in particular example XVI-1, in which the amount of Fe could be up to 10. Thus, the claims were not allowable under Article 54(1) EPC.

(b) As regards inventive step, the claimed subject-matter was considered to be too broad compared to what was demonstrated in the examples of the application as filed. Inventive step could not be accepted for catalytic compositions where B and C were selected from Mn, Co, Ag, Pb, Re, Cd, Zn, and Cr, Al, Sb, P, Ge, La, Sn, V, respectively. The argument that those elements were conventional promoters as shown by documents could not be accepted, since in view of D2 as the closest state of the art, it could not be expected that those elements might have the same effect.

(c) In additional remarks the following was pointed out: The amendment made to claim 1 violated Art. 123(2) EPC, since there was no basis in the application as filed for the amendment e = 1.7 introduced in claim 1, since that value was based on a generalization of a specific example. Since the alleged effect was based on a selection, the criteria of selection invention should be met.

IV. On 10 March 2003, the applicant (appellant) filed a notice of appeal against the above decision, the prescribed fee being paid on the same day. With the statement setting out the grounds of appeal filed on 6 May 2003, the appellant maintained the claims underlying the decision under appeal. Auxiliarily, they requested that in claim 1 of the main request the feature "e = 1.7 to 10" be replaced by the feature "e = 2 to 9".

V. The arguments of the appellant can be summarized as follows:

(a) The amended claims met the requirements of Article 123(2) EPC. Although the ratio iron to molybdenum could be calculated to be 1.73285, the iron content and the parameter "e" were quoted to one decimal place. Furthermore, the description already demonstrated that the parameter "e" was essential and that a hierarchy of intermediate ranges was given from 2 to 9, preferably 2 to 8. The value of 1.7 was consistent with the preferred range and could be derived from the examples. As regards the auxiliary request, the basis for the amendment of feature "e" was given.

(b) The claimed subject-matter was novel over D2, since a combination of example B-6 and the general teaching in D2 could not be considered. In all the examples the atomic ratio of Mo to Fe was at most 12:1 and only the compositions in examples B1, B6 and C6 contained calcium. Thus, the skilled person would not consider a value in the overlapping

range of 1.7 and 3 as critical. The same kind of arguments applied *mutatis mutandis* with respect to the auxiliary request.

Having regard to example XVI-1 of D3 the same arguments applied as set out with respect to D2. Furthermore, there was a generic disclosure not only with respect to the iron content (d = 0.1 to 10) but also with respect to the molybdenum content (a being 2 to 12) and the tungsten content (b = 0 to 10; and a + b = 12). Since in example XVI-1 a + b = 14 the claimed requirement was not met, the teaching of that example could not simply be combined with the general disclosure. Thus, the claimed subject-matter of the main and the auxiliary request was novel over D3 as well.

(c) As regards inventive step, D2 was considered to be the closest state of the art. It was not obvious to increase the level of iron in the catalyst of D2 so that the parameter e = 1.7 to 10 was met. There was no motivation in the further cited prior art in order to do so. In particular, D2 and D3 were not directed to the problem posed namely to increase the hydrogen cyanide level in an ammoxidation process. In D3 the specific surface area of the catalyst particles should be controlled, which was far away from the inventive concept of the catalysts in suit.

VI. In a communication dated 19 March 2007, the Board objected to the amended term "e = 1.7 to 10" in the main request (Article 123(2) EPC) and questioned whether or not the composition of example 10 as filed

could form the basis for the amendment, since this example was not an embodiment of what was claimed, as it contained no metal C as defined. The proposed amendment of "e = 2 to 9" in claim 1 of the auxiliary request was considered to be allowable under Article 123(2) EPC.

As regards novelty of the auxiliary request, the disclosure of documents D2 or D3 should be considered. As regards inventive step, D2 was considered to be a possible candidate for the closest state of the art. Having regard to the test results in table 1 of the application in suit, the Board had difficulties to see any improved effect, in particular vis-à-vis the Ca containing catalyst of example 6B of D2. Thus, the problem should be reformulated in a less ambitious way, for example to provide an alternative catalyst composition.

VII. In reply to the invitation for oral proceedings communication the Board was informed by letter of 30 March 2007 that the appellant would not be attending oral proceedings.

VIII. Oral proceedings were held on 22 May 2007 in the absence of the appellant as announced. The oral proceedings were continued in the absence of the appellant in accordance with Rule 71(2) EPC.

IX. The appellant had requested in writing that the decision under appeal be set aside and that a patent be granted on the basis of claims 1 to 10 underlying the decision under the appeal, or auxiliarily on the basis

of an amended claim 1 as set out in the grounds of appeal filed 6 May 2003 and current claims 2 to 10.

Reasons for the Decision

1. The appeal is admissible.

Amendments

Main request

2. Claim 1 has been amended in that the content of Fe was defined to be "e = 1.7 to 10". That feature was objected to in the decision under appeal as violating Article 123(2) EPC. The appellant referred to example 10 of the application as filed as a basis for that amendment.

2.1 Example 10 discloses a catalyst composition which is prepared by using silica gel as support material (page 7, line 2, claim 2) and having the following composition (table I):

$Cs_{0.1}K_{0.1}Ni_{6.2}Mg_{2.5}Fe_2Bi_{0.5}Ca_{1.5}Mo_{13.85}O_x$.

According to claim 1 as filed the elements Ca, Fe, Bi and Mo and one of the components from each group A, B and C are obligatory. Component of group C is one or more of Ce, Cr, Al, Sb, P, Ge, La, Sn, V and W or mixtures thereof. However, the composition of example 10 does not contain any element from group C. Thus, example 10 does not illustrate the claimed subject-matter and for that reason alone cannot provide a basis for the amendment effected.

2.2 Furthermore, since the content of the elements are based on Mo_{12}, feature "e" of that example 10 must be recalculated. The correct Fe-value can be calculated to be 1.73285 which is different from the amended value of 1.7. In the application as filed, there is no basis for any such generalisation.

2.3 Finally, in example 10 as filed, the amount of Fe is closely associated with specific amounts of Bi, Mo and Ca as obligatory component and with specific amounts of Ce, Ni and Mg so as to determine the conversion and the acrylonitrile production in a significant degree (see table I). Since the lower limit of feature "e" is restricted to a preferred embodiment (example 10), it is not admissible under Article 123(2) EPC, having regard to the functional and structural relationship among such features, to extract an isolated feature (here e = 1.7) from a set of features (here a specific exemplified catalyst composition), which had been originally disclosed in combination for that embodiment (Case Law of the Boards of Appeal of the European Patent Office, 5th edition, 2006, III.A.1.1, in particular T 1067/97). Consequently, the specific parameter (Fe content) of said exemplified composition cannot be singled out and generalized so as to provide the lower limit of a general range.

There is no other basis in the application as filed for the amended feature "e = 1.7 ...".

2.4 From the above it follows that the amendment effected cannot directly and unambiguously be derived from the application as filed. Hence, the main request does not

meet the requirements of Article 123(2) EPC and is not allowable.

Auxiliary request

3. There is a general disclosure for the features b and e being in the range of 2 to 9 in the application as filed (page 4, lines 27 and 28). Thus, there is an original basis to limit feature "e" to 2 to 9. Consequently, claim 1 of the auxiliary request is allowable under Article 123(2) EPC.

Novelty

4. The examining division was of the opinion that the claimed subject-matter lacked novelty over D2 and D3.

4.1 D2 discloses a process for producing a composite oxide catalyst, wherein a Mo-Bi composite oxide catalyst represented by the following formula is produced by a process comprising incorporating the compounds as respective element sources into a composite in an aqueous system and subjecting the composite to heat treatment, characterized in that use is made as a Bi source of a compound of bismuth carbonate complex of (a) Bi and Na, or (b) Bi, Na and X or (c) Bi and X which comprises at least a part of each of the required Na and/or X

$Mo_aBi_bCo_cNi_dFe_eNa_fX_gY_hZ_iSi_jO_k$

wherein: X represents Mg, Ca, Zn, Ce and/or Sm; Y represents K, Rb, Cs and/or Tl; Z represents B, P, As and/or W; a-k represent atomic ratios, respectively, and when a equals 12, b = 0.5 to 7, c = 0 to 10, d = 0 to 10, c+d = 1 to 10, e = 0.05 to 3, f = 0.01 to 1, g =

0 to 1, h = 0.04 to 0.4, i = 0 to 3, j = 0 to 48 and k is a numeral which satisfies the oxidation state of the other elements (claim 1).

4.1.1 In example 6-B of D2, a catalyst having the following composition is produced:

$Mo_{12}W_{0.5}Bi_1Co_4Fe_1Na_{0.05}Ca_{0.1}K_{0.06}Si_2Al_5$

That composition is prepared by dissolving ammonium paramolybdate in water and adding ammonium paratungstate under agitating. On the other hand, ferric nitrate and cobalt nitrate are dissolved in water under heating. These two solutions are mixed. To the mixed solution is added a solution of potassium nitrate. Then, bismuth subcarbonate in which Na and Ca have been complexed and colloidal silica and particulate alpha-Al_2O_3 are added, and the mixture is mixed with agitation. The mixture is then dried, heated and calcinated. Thus, the presence of Si and Al in the above composition results from the use of colloidal silica and Al_2O_3 support material.

4.1.2 The examples as filed are prepared by mixing an aqueous solution of ammonium heptamolydate with a silica sol to which a slurry containing the compounds, preferably nitrates of the other elements, is added (page 7, first paragraph). Whilst in the composition of example B-6 the presence of Al and Si is indicated, in table I of the application as filed the compositions are indicated without mentioning silicon, although it has been added. Indeed, according to claim 2 as filed silica and alumina can be used as inert support either alone or in the form of a mixture and can thus be present in the

metal oxide composition. Consequently, the presence of Si and Al in the metal oxide composition of example B-6 does not provide any distinction over the claimed subject-matter. Furthermore, the appellant had never argued in that respect.

4.1.3 The composition of example B-6 as such is not novelty destroying for the claimed subject-matter, since the content of obligatory Fe is 1 and thus outside the claimed range of 2 to 9. However, the examining division was of the opinion that example B-6 in combination with the general disclosed range for Fe anticipated the claimed range.

4.1.4 D2 discloses at least ten exemplified compositions which include obligatory Fe in combination with other elements (see example A-1, comp. example A-2, examples A-2, A-3 and B-1, comp. example B-1, example B-6, comp. example B-2, examples C-1 and C-6). There are further eight exemplified compositions in which the X-component (see point 4.1 above) and its content have been modified (see tables B-1 and C-1). From these eighteen exemplified compositions only examples B1, B6, C2 and C6 contain in addition to Fe also calcium as a component and only example B-6 discloses also the other obligatory elements as claimed. Since Ca is not an obligatory element of D2, as index g for element X can be 0, there is a longer list of exemplified catalyst compositions which may be modified with respect to the Fe content being 0.05 to 3 (claim 1).

4.2 From the above it follows that even if the examples and the disclosure in the general description were considered, it was necessary to make a multiple

selection in the following respect: select from about eighteen exemplified compositions those, in which the non-obligatory metal Ca is present; select from the remaining four examples only example B-6; select as modifying element only Fe and select an appropriate content "e" to be for example 3.

4.3 There is no pointer in D2 to the particular combination of features in the catalyst composition now being claimed. For destroying novelty it is not sufficient, to associate, in the knowledge of the invention, the specific type of catalyst composition (containing Ca), the specific metallic element (Fe) and the specific content thereof, selected from many possibilities offered by the prior art document so as to create a catalyst composition of the application in suit. Quite to the contrary, it is necessary that the claimed combination should directly and unambiguously be derived from that document. Hence, D2 is not novelty destroying for the subject-matter of claim 1.

4.4 D3 discloses a catalyst useful for the oxidation of propylene, said catalyst having a composition represented by the following formula:

$Mo_aW_bBi_cFe_dA_eB_fC_gD_hO_x$

wherein Mo denotes molybdenum, W denotes tungsten, Bi denotes bismuth, Fe denotes iron, A denotes at least one element selected from the group consisting of nickel and cobalt, B denotes at least one element selected from the group consisting of alkali metal, alkaline earth metal and thallium, C denotes at least one element selected from the group consisting of phosphorus, tellurium, antimony, tin, cerium, lead, niobium, boron, arsenic, manganese and zinc, D denotes

at least one element selected from the group consisting of silicon, aluminum, titanium and zirconium, and O denotes oxygen;
and further a, b, c, d, e, f, g, h and x denote atomic ratios respectively, and when a=2 to 12, b=0 to 10 and a+b=12, then c=0.1 to 10, d=0.1 to 10.0, e=2 to 20, f=0.005 to 3.0, g=0 to 4.0, h=0.5 to 15 and x is a numerical value determined depending upon the atomic values of the other elements than oxygen and wherein said unfired material powder particles formed in said centrifugal flow coating device further comprise at least one of nickel and cobalt, at least one of alkali metal, alkaline earth metal and thallium, at least one of silicon, aluminum, titanium and zirconium, and optionally, at least one of phosphorus, tellurium, antimony, tin, cerium, lead, niobium, boron, arsenic, manganese, zinc and tungsten (claim 7). In example XVI-1 a catalyst composition consists of $Mo_{12}W_2Ni_8Bi_1Fe_1Si_{1.35}Cs_{0.2}Mg_{1.0}Ca_{1.0}$.

4.4.1 The exemplified composition as such is not novelty destroying, since the content of Fe is 1 and thus outside the claimed range of 2 to 9. On the other hand example XVI-1 is the only example of a longer list of exemplified compositions, which contains Ca, which is not an obligatory component according to the teaching of D3.

4.4.2 The selection criteria are similar to those indicated with respect of D2 above so that the same considerations as outlined under points 4.2 and 4.3 above apply *mutatis mutandis* with respect to novelty over D3. Consequently, there is no direct and

unambiguous disclosure in D3 to make the claimed selection within the disclosure of D3.

4.4.3 Consequently, claim 1 of the auxiliary request is novel over D2 and D3.

Inventive step

Problem and solution

5. The application in suit concerns a Mo-Bi-Fe-Ca based catalyst composition for the manufacture of acrylonitrile and hydrogen cyanide. Such a catalyst composition is known from D2, which the appellant and the examining division regarded as the closest prior art document. The Board has no reason to deviate from that approach as can be gathered from the following:

5.1 The application in suit relates to an improved catalyst for use in the ammoxidation of unsaturated hydrocarbons, such as propylene, to the corresponding unsaturated nitrile, such as acrylonitrile, which unexpectedly provides increased yields in coproduct HCN without significant decrease in the yield of the unsaturated nitrile (page 1, first paragraph). According to D2, it is well known that "Mo-Bi composite oxide catalysts are useful for selective reactions such as ... a vapor phase catalytic ammoxidation reaction for producing acrylonitrile from propylene..." (page 2, lines 6 to 8). In such prior art reactions acrylonitrile has been attained as a major product while hydrogen cyanide has been maintained as the major coproduct (application in suit, page 2, first paragraph). According to the application as filed, the alleged increase in HCN is

due to the presence of Ca in the composition (see also page 6, second paragraph and table I). The exemplified catalyst of D2 discloses a composition having the same components as those of the application as filed (see above points 4.1 to 4.3), and is used for the same purpose.

5.2 The definition of the technical problem to be solved should normally start from the technical problem actually described in the application in suit in relation to the closest state of the art indicated there (Case Law, *supra*, I.D.4.3.2). According to the application in suit, an ammoxidation catalyst should be provided which increases the yields of hydrogen cyanide coproduct produced during the manufacture of acrylonitrile while maintaining the acrylonitrile production levels at substantially the same level (page 3, lines 6 to 11). That problem is directed to an improvement over the state of the art.

5.3 The question arises whether or not there is any evidence on file that such a problem has effectively solved. As stated in the communication of the Board, the test results in table I of the application in suit only show a minor increase in the yields of hydrogen cyanide in spite of a decrease in acrylonitrile production (comparative example 1 and examples 7 to 11). Furthermore, the Board had difficulties to see any improved effect from the data on file (communication, point 3.2).

5.4 Since the appellant did not address this objection, the Board can only speculate in which way the test results on file reflect a comparison with any specific prior

art catalysts and whether or not reaction conditions (not specified in the claims) may be responsible for the test results shown rather than the catalyst composition as such. In any case, the comparative compositions do not reflect catalyst compositions of the closest prior art document D2, since they do not contain Ca.

5.4.1 As stated in the communication mentioned above, there is no evidence on file for any improvement vis-à-vis the Ca containing catalyst of example 6B of D2 (point 3.2). The Ca containing Mo-Bi-Fe catalyst of example B-6 of D2 comes closer to the claimed subject-matter than any of the comparative examples of the application in suit, since it contains Ca within the claimed range. Since the catalyst as claimed differs from the composition of example 6-B of D2 only in that the content of Fe is higher, in the absence of any evidence it cannot be assumed that this difference has any beneficial effect on the catalytic performance of the catalyst. Since the appellant has not made any attempt to file further test results in that respect, the Board cannot accept from the data on file that an improved effect over the closest state of the art, for example in respect of the hydrogen cyanide production, can be achieved.

5.5 As regards the argument of the appellant that the desirability of increasing the hydrogen cyanide production capability has not been mentioned in D2, the following comments are given:

5.5.1 When assessing the objective problem, it is not important whether this problem is mentioned in the

closest prior art; what matters is what the skilled person objectively recognizes as the problem when comparing the closest state of the art with the invention (Case Law, *supra*, I.D.4.3.1). From the comparative tests on file, an objectively recognized improvement over the closest state of the art cannot be derived.

5.5.2 Furthermore, alleged advantages to which the patent proprietor merely refers without offering sufficient evidence supported by any comparison with the closest prior art, cannot be taken into consideration in determining the problem underlying the invention (Case Law, *supra*, I.D.4.2), or in assessing whether or not the problem has been solved. Since the problem cannot be formulated in terms of an improvement over D2, it has to be reformulated.

5.6 Hence, the technical effects shown in the application in suit only justify the formulation of a technical problem in relation to D2 which is less ambitious than described on page 3, first and second paragraph of the application as filed. Thus, the problem effectively solved over D2 may therefore only be seen in providing an alternative catalyst composition for ammoxidation.

Obviousness

6. It remains to be decided whether the claimed subject-matter is obvious having regard to the documents on file.

According to D2, the general content of Fe may be in the range of 0.05 to 3 (see claim 1). The composition

of the claimed subject-matter differs from that of example B-6 only in that the Fe content is higher. Since there is no evidence on file that the selected different parameter "e" provided a specific technical effect, the selection is arbitrary. Since the only problem that can be accepted as solved was to provide an alternative composition to that of D2, almost any modification of the known catalyst composition within the limits suggested by D2 itself can be regarded as a feasible alternative by the person skilled in the relevant art and is therefore obvious. This includes a modification of the content of Fe to 3 in the composition of example B-6 of D2 to something which falls within the subject-matter of claim 1. The claimed subject-matter is thus obvious.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar The Chairman

N. Maslin S. Perryman