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**Datasheet for the decision
of 29 June 2010**

Case Number: T 0209/07 - 3.3.07

Application Number: 98114580.8

Publication Number: 0895809

IPC: B01J 23/20

Language of the proceedings: EN

Title of invention:

Niobium-containing aqueous solution for use in producing niobium-containing oxide catalyst

Applicants:

Asahi Kasei Kabushiki Kaisha

Headword:

-

Relevant legal provisions:

EPC Art. 123(2)

Relevant legal provisions (EPC 1973):

EPC Art. 54, 111(1)

Keyword:

"Amendments: added subject-matter (no)"
"Novelty (yes) - choice from two lists"

Decisions cited:

-

Catchword:

-

Case Number: T 0209/07 - 3.3.07

**DECISION
of the Technical Board of Appeal 3.3.07
of 29 June 2010**

Appellants: Asahi Kasei Kabushiki Kaisha
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Osaka 530-8205 (JP)

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Decision under appeal: Decision of the Examining Division of the
European Patent Office posted 28 July 2006
refusing European patent application
No. 98114580.8 pursuant to Article 97(1)
EPC.

Composition of the Board:

Chairman: S. Perryman
Members: D. Semino
B. ter Laan

Summary of Facts and Submissions

- I. The appeal lies from the decision of the Examining Division refusing European patent application No. 98 114 580.8 (published under No. 0895809).

- II. The application as filed contained 18 claims, wherein claims 1 to 8 concerned a niobium-containing aqueous solution for use in producing a niobium-containing oxide catalyst, claims 9 to 11 a niobium-containing oxide catalyst for use in a catalytic oxidation or ammoxidation of propane or isobutane in the gaseous phase, claims 12 to 14 a process for producing said catalyst, claim 15 a process for producing acrylic acid or methacrylic acid using said catalyst and claims 16 to 18 a process for producing acrylonitrile or methacrylonitrile using said catalyst.

In particular process claims 12 to 14 had the following wording:

"12. A process for producing a niobium-containing oxide catalyst for use in a catalytic oxidation or ammoxidation of propane or isobutane in the gaseous phase, wherein said niobium-containing oxide catalyst comprises an oxide of a plurality of active component elements including niobium, said process comprising mixing a niobium-containing aqueous solution with an aqueous mixture or aqueous mixtures containing compounds of active component elements of said oxide catalyst other than niobium, to thereby provide an aqueous compound mixture, and drying said aqueous compound mixture, followed by calcination, said niobium-containing aqueous solution comprising water having dissolved therein a dicarboxylic acid, a niobium compound and optionally ammonia, wherein the molar ratio (•) of said dicarboxylic acid to the niobium contained in said niobium compound satisfies the following relationship: $1 \cdot (\cdot) \cdot 4$, and the molar ratio (•) of said ammonia to the niobium contained in said niobium compound satisfies the following relationship: $0 \cdot (\cdot) \cdot 2$."

"13. The process according to claim 12, wherein said active component elements are niobium, molybdenum, vanadium, and at least one element selected from the group consisting of tellurium and antimony."

"14. The process according to claim 12 or 13, wherein said aqueous compound mixture is provided so as to further contain a silica sol in an amount such that said oxide catalyst further comprises a silica carrier in an amount of from 20 to 70 % by weight, based on the total amount of said oxide and said silica carrier, said silica carrier having supported thereon said oxide of a plurality of active component elements."

- III. The decision of the Examining Division was based on a main request and an auxiliary request both filed at the oral proceedings before the Examining Division on 13 June 2006. Claim 1 according to the main request corresponded to claim 1 as originally filed in which the niobium-containing aqueous solution had been limited by a disclaimer. Claim 1 according to the auxiliary request instead was reworded as a use claim directed to the use of a niobium-containing aqueous solution in producing a niobium-containing oxide catalyst.
- IV. The following documents were *inter alia* cited:
- D1: JP-A-6 321543 (abstract and English translation)
D2: EP-A-0 318 295
D3: T.F. Limar et al., "Oxalato-compounds of niobium", Russian Journal of Inorganic Chemistry, Volume 9(10), 1964, pages 1288-1291
D4: M.M.A. Sekar et al., "Low-temperature synthesis and properties of microwave resonator materials", Materials Science & Engineering B38 (1996), pages 273-279
- V. The decision under appeal can be summarized as follows:
- (a) The introduction of a disclaimer into claim 1 of the main request, which intended to exclude the disclosure of D3, did not meet the requirements of Article 123(2) EPC, since D3 contained a highly relevant technical teaching and could not be considered as an accidental anticipation.
- (b) The disclosure of an aqueous solution of $\text{Nb}_2(\text{C}_2\text{O}_4)_5$ for the production of a niobium-containing oxide catalyst in D2 implied a stoichiometric ratio of 2.5 between oxalate and niobium, so that D2 anticipated the use of claim 1 according to the auxiliary request.
- (c) The arguments of the applicants that the compound $\text{Nb}_2(\text{C}_2\text{O}_4)_5$ did not exist or that, if it existed, it would not be soluble in water, could not be accepted on the basis of D2 itself and D4.
- VI. On 4 October 2006, the applicants (appellants) filed a notice of appeal against the above decision, the prescribed appeal fee being paid on the same day. Several requests were filed by the appellants with the statement setting out the grounds of appeal and with the letters of 21 May 2010 and 25 June 2010 which were later withdrawn. With letter of 21 May 2010 the appellants also filed *inter alia* an Expert Opinion by Prof. M. Kakihana (D8).
- VII. Oral proceedings were held on 29 June 2010. At the oral proceedings the appellants withdrew all pending requests and filed an amended set of two claims as main request, which

read as follows (amendments with respect to the claims as originally filed in bold, deletions in strike-through):

~~"12.~~ **1.** A process for producing a niobium-containing oxide catalyst for use in a catalytic oxidation or ammoxidation of propane or isobutane in the gaseous phase, wherein said niobium-containing oxide catalyst comprises an oxide of ~~a plurality of active component elements including niobium~~ **niobium, molybdenum, vanadium, and at least one element selected from the group consisting of tellurium and antimony, as active components,** said process comprising mixing a niobium-containing aqueous solution with an aqueous mixture ~~or aqueous mixtures~~ containing compounds of ~~active component elements of said oxide catalyst other than niobium~~ **molybdenum, vanadium, and at least one element selected from the group consisting of tellurium and antimony,** to thereby provide an aqueous compound mixture, and drying said aqueous compound mixture, followed by calcination, said niobium-containing aqueous solution comprising water having dissolved therein a dicarboxylic acid, a niobium compound and optionally ammonia, wherein the molar ratio (•) of said dicarboxylic acid to the niobium contained in said niobium compound satisfies the following relationship: $1 \cdot (\cdot) \cdot 4$, and the molar ratio (•) of said ammonia to the niobium contained in said niobium compound satisfies the following relationship: $0 \cdot (\cdot) \cdot 2$."

~~"14.~~ **2.** The process according to claim 12 or 13 (*sic*), wherein said aqueous compound mixture is provided so as to further contain a silica sol in an amount such that said oxide catalyst further comprises a silica carrier in an amount of from 20 to 70 % by weight, based on the total amount of said oxide and said silica carrier, said silica carrier having supported thereon said oxide of a plurality of active component elements."

VIII. The arguments of the appellants can be summarised as follows:

- (a) Claim 1 of the main request resulted from the combination of claims 12 and 13 as originally filed with the deletion of one out of two options. In particular, the mixing of the niobium-containing aqueous solution with an aqueous mixture containing compounds of the other active elements corresponded to the specific description of the process in paragraphs [99]-[101] of the original application and to all examples.
- (b) D1 and D3 did not disclose processes for the production of a niobium-containing oxide catalyst for use in a catalytic oxidation or ammoxidation of propane or isobutane and were therefore not relevant for novelty.

- (c) D2 did not provide sufficient disclosure of the compound $\text{Nb}_2(\text{C}_2\text{O}_4)_5$, which was mentioned therein. Such a compound did not exist, as motivated in the Expert Opinion D8 and was nothing more than an imaginary compound invented while taking only charge balance into consideration. Therefore, in this respect, D2 did not contain an enabling disclosure, so that it could not be used against novelty. Moreover, no disclosure was present in D2 of mixing a niobium-containing aqueous solution with an aqueous mixture containing compounds of molybdenum, vanadium and at least one element selected from the group consisting of tellurium and antimony. On the contrary, according to D2 the addition of the molybdenum component as the last ingredient to a solution already containing vanadium, tellurium and niobium components, was preferred.

IX. The appellants requested that the decision under appeal be set aside and that the matter be remitted for further prosecution on the basis of the claims of the main request submitted at the oral proceedings on 29 June 2010.

Reasons for the Decision

1. The appeal is admissible.
2. *Amendments*
 - 2.1 Claim 1 of the main request corresponds to claim 12 as originally filed with the following amendments:
 - (a) the oxide catalyst now specifically comprises "an oxide of niobium, molybdenum, vanadium, and at least one element selected from the group consisting of tellurium and antimony as active components" instead of the generic "oxide of a plurality of active component elements including niobium";
 - (b) the process now specifically comprises mixing a niobium-containing aqueous solution with "an aqueous mixture containing compounds of molybdenum, vanadium, and at least one element selected from the group consisting of tellurium and antimony" instead of "an aqueous mixture or aqueous mixtures containing compounds of active component elements of said oxide catalyst other than niobium".
 - 2.2 Amendment (a) corresponds to original claim 13. Since original claim 13 was directly dependent on original claim 12, the introduction of that additional feature is not objectionable under Article 123(2) EPC.
 - 2.3 As far as amendment (b) is concerned, according to original claim 12, a niobium-containing aqueous solution is mixed

with an aqueous mixture **or with aqueous mixtures** (emphasis added by the board) containing compounds of active component elements of said oxide catalyst other than niobium. While the latter option (aqueous mixtures in plural) leaves it open in which order the solutions of the different components are mixed, the former (aqueous mixture in singular) implies that an aqueous mixture of the active components other than niobium is prepared first and is only then mixed with an aqueous solution containing niobium. The singular option corresponds in particular to the preferred embodiment in the description with reference to specific compounds of molybdenum, vanadium, tellurium and antimony (paragraphs [0099]-[0101]), and also to the procedure used in all the examples. Therefore, amendment (b) finds its basis in the application as originally filed. It is noted that, in agreement with amendment (a), it is further specified in the claim which active component elements other than niobium are present in the aqueous mixture, namely molybdenum, vanadium and at least one element selected from the group consisting of tellurium and antimony.

- 2.4 Claim 2 of the main request corresponds to claim 14 as originally filed. However, in this claim a clerical error appears in that the dependence on original claims 12 and 13 has not been amended. That error should still be corrected.
- 2.5 In view of the above, it can be concluded that the amendments find their basis in the application as originally filed so that the requirements of Article 123(2) are fulfilled.
3. *Novelty*
- 3.1 Document D1 describes a niobium oxide sol and a method for producing it (claims 1 and 2, English translation). Although a general indication is given that the niobium oxide sol can be used as a niobium source for a catalyst (paragraph [0001] in the English translation), no method of producing such a catalyst is disclosed. Therefore, D1 does not prejudice the novelty of present claim 1.
- 3.2 Document D3 concerns the production of oxalato-compounds of niobium by crystallisation from a solution of niobium hydroxide in oxalic acid or ammonium, potassium or sodium oxalates (Summary on page 1291). No niobium-containing catalyst, nor a method of production of such a catalyst is described, so that D3 does not anticipate the subject-matter of claim 1 according to the main request.
- 3.3 Document D2 discloses a process for producing a nitrile which comprises subjecting an alkane and ammonia in the gaseous state to catalytic oxidation in the presence of a solid oxide catalyst comprising molybdenum, vanadium, tellurium and niobium (claim 1). Such a process is referred

- to as ammoxidation; propane and isobutane are exemplified as alkanes (page 2, lines 9-11).
- 3.3.1 The solid oxide catalyst is produced by a process which comprises adding to an aqueous solution of ammonium metavanadate, V_2O_5 , V_2O_3 , $VOCl_3$ or VCl_4 , in any order:
(i) an aqueous solution of ammonium niobium oxalate, $NbCl_3$, $NbCl_5$ or $Nb_2(C_2O_4)_5$,
(ii) an aqueous solution of telluric acid or TeO_2 ; and
(iii) an aqueous solution of ammonium paramolybdate, MoO_3 , $MoCl_5$, phosphomolybdic acid, silicomolybdic acid or a heteropoly acid which contains mixed-coordinate molybdenum and vanadium, subsequently heating the mixture, concentrating the mixture to dryness and calcining the resulting dry solid (claim 10).
- 3.3.2 Although in the above preparation process the order of addition of the metal elements is not specified, it is stated in D2 that it is desirable to add the molybdenum component, for example in the form of an aqueous solution of ammonium paramolybdate, last of all as it facilitates obtaining a uniform aqueous solution (page 2, lines 53-56).
- 3.4 Therefore, D2 describes a process for producing a niobium-containing oxide catalyst for use in a catalytic ammoxidation of propane or isobutane in the gaseous phase, wherein said niobium-containing oxide catalyst comprises an oxide of niobium, molybdenum, vanadium and tellurium as active components, said process comprising mixing a niobium-containing aqueous solution with aqueous mixtures containing compounds of molybdenum, vanadium and tellurium, to thereby provide an aqueous compound mixture, and drying said aqueous compound mixture, followed by calcination. It remains to be determined whether the process of D2 includes the addition of the niobium-containing aqueous solution to the aqueous compound mixture as the last component, in combination with the use of a niobium-containing solution with the • and • values within the claimed ranges.
- 3.5 The process of claim 10 of D2 specifies that the addition of the niobium-containing solution, the tellurium-containing solution and the molybdenum-containing solution to the vanadium-containing solution may take place "in any order". This means that several options are left open, including those in which the niobium-containing solution is added as the last component. Even if some of the options are not the preferred ones (see points 3.3.1 and 3.3.2, *supra*), they are encompassed by the wording of claim 10.
- 3.6 As far as the composition of the niobium-containing solution is concerned, four options are listed in claim 10, namely an aqueous solution of ammonium niobium oxalate, $NbCl_3$, $NbCl_5$ or $Nb_2(C_2O_4)_5$.

- 3.6.1 There can be no doubt that an ammonium niobium oxalate solution contains oxalate and ammonia ions together with niobium-containing ions; however, in the absence of a specific chemical formula or further information in D2, the exact molar ratios of those components are not known.
- 3.6.2 The same does not hold for the aqueous solution of $Nb_2(C_2O_4)_5$. It can be accepted that $Nb_2(C_2O_4)_5$ does not exist as such in view of D8, in which it is stated that that compound had never been described in the art (pages 7 and 8 of D8, "2. Observation based on authoritative remarks about niobium complexes"). At the same time, according to D8, several ions with specific oxalate to niobium molar ratios are formed when niobium compounds and oxalate compounds are dissolved, such as $[NbO(C_2O_4)_3]^{3-}$ with a ratio of 3 or $[NbO(C_2O_4)_2(H_2O)_2]^{3-}$ with a ratio of 2 (end of page 2 to page 4, "1-1 Oxo-oxalato complexes of niobium"). Hence, the skilled person could only understand the disclosure of an aqueous solution of $Nb_2(C_2O_4)_5$ in D2 as referring to a solution containing oxalate and niobium in a ratio 5:2 (equal 2.5). Such a solution does not contain ammonia so that the molar ratio of ammonia to niobium is 0.
- 3.7 In view of the above, it can be concluded that D2 discloses, as one of a number of options, the use of a niobium-containing solution with a molar ratio of 2.5 and a molar ratio of 0 and also, as one of several possibilities, the addition of the niobium-containing solution as the last component to the aqueous mixture. However, it does not directly and unambiguously disclose those two features in combination. Such a combination would result from a choice out of two lists (the list of niobium compounds and the list of the possible order of addition), which is neither explicitly nor implicitly disclosed in D2.
- 3.8 For these reasons, novelty of the process of claim 1 of the main request with respect to the disclosure in D2 has to be acknowledged.
- 3.9 As claim 2 is a specific embodiment of claim 1, it also fulfils the requirements of Article 54 EPC.

Remittal

4. In view of the above and since the present claims are clear, it is concluded that the main request fulfils the requirements of Articles 123(2) and 54 EPC as well as Article 84 EPC, so that the reasons for the refusal of the application by the Examining Division no longer exist. As the substantive issue of inventive step has not been addressed in the appealed decision and in view of the request of the appellants and of the new formulation of the claims, which may require additional evidence in order to show the presence of technical advantages related to the technical difference between the claimed process and the one

of D2, the Board considers it appropriate to exercise the power conferred by Article 111(1), second sentence, EPC to remit the case to the Examining Division for further prosecution.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The matter is remitted to the first instance for further prosecution on the basis of the claims of the main request submitted at the oral proceedings on 29 June 2010.

The Registrar

The Chairman

S. Fabiani

S. Perryman