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**Datasheet for the decision
of 26 February 2010**

Case Number: T 1623/09 - 3.3.07

Application Number: 03748820.2

Publication Number: 1558383

IPC: B01J 31/00

Language of the proceedings: EN

Title of invention:

Vanadium catalysts and a process for the direct conversion of methane into acetic acid

Applicants:

Instituto Superior Técnico

Opponent:

-

Headword:

-

Relevant legal provisions:

EPC Art. 123(2)

Relevant legal provisions (EPC 1973):

-

Keyword:

"Amendments - added subject-matter (yes)"

Decisions cited:

-

Catchword:

-



Case Number: T 1623/09 - 3.3.07

D E C I S I O N
of the Technical Board of Appeal 3.3.07
of 26 February 2010

Appellants: Instituto Superior Técnico
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Representative: Pereira da Cruz, Joao
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Decision under appeal: Decision of the Examining Division of the
European Patent Office posted 10 February 2009
refusing European patent application
No. 03748820.2 pursuant to Article 97(1) EPC.

Composition of the Board:

Chairman: S. Perryman
Members: D. Semino
F. Rousseau

Summary of Facts and Submissions

I. The appeal lies from the decision of the Examining Division refusing European patent application No. 03 748 820.2 originating from international application PCT/PT2003/000015 having an international filing date of 15 October 2003 and published as WO 2004/037416. The application as filed comprised 4 claims, which read as follows:

"1. Catalyst system and process for the direct one-pot conversion of methane into acetic acid, under relatively mild conditions, characterized by containing a vanadium complex, a peroxodisulfate salt and trifluoroacetic acid."

"2. Catalyst system and process according to claim 1, characterized by the use of a vanadium complex with di- or poly-dentate ligands coordinated by nitrogen and oxygen (N,O) atoms or by oxygen and oxygen (O,O) atoms."

"3. Catalyst system and process according to claims 1 and 2, characterized by the use of a complex of vanadium in the +4 or +5 oxidation state, with ligands derived from aminoalcohols, (hydroxyimino)dicarboxylic acids, hydroxypyrones, trifluoroacetic acid, triflic acid or an inorganic acid."

"4. Catalyst system and process according to claims 1, 2 and 3, characterized by the use on [sic] carbon monoxide."

- II. In its decision posted on 10 February 2009 the Examining Division refused the application on the grounds that the application did not comply with the requirements of Articles 84 and 123(2) EPC. The decision was based on a single set of amended claims 1-3 filed with letter dated 20 May 2008 wherein both product claim 1 and process claims 2 and 3 included inter alia an amendment defining in parenthesis the relatively mild conditions as follows: "(temperatures below 100°C and pressures up to 30 atm)".
- III. According to the decision no basis was present in the application as filed for the attempted definition of "relatively mild conditions" in claims 1-3, namely for temperatures below 100°C and pressures up to 30 atm and the use of parentheses in said definition led to lack of clarity.
- IV. On 1 April 2009, the applicants (appellants) filed a notice of appeal against the above decision, the prescribed appeal fee being paid on the same day. With the statement setting out the grounds of appeal filed on 9 June 2009, the appellants submitted four sets of claims 1-3 as main, first, second and third auxiliary requests. Claims 1-3 according to the main request read respectively as follows:

"1. Catalyst for the direct one-pot conversion of methane into acetic acid, at pressures in the range of 2 - 32 atm, the catalyst comprising a vanadium complex with the metal in the +4 or +5 oxidation state and with di- or poly-dentate ligands coordinated by nitrogen and oxygen (N,O) atoms or by oxygen and oxygen (O,O) atoms derived from aminoalcohols, (hidroxyimino)dicarboxylic

acids, hydroxypyranones, trifluoroacetic acid or triflic acid."

"2. Process for the direct one-pot conversion of methane into acetic acid, at pressures in the range of 2 - 32 atm, characterised by the association of a vanadium complex, according to claim 1, with a peroxodisulfate salt, in trifluoroacetic acid, without requiring the use of carbon monoxide."

"3. Process for the direct one-pot conversion of methane into acetic acid, at pressures in the range of 2 - 32 atm, characterised by the association of a vanadium complex, according to claim 1, with a peroxodisulfate salt, in trifluoroacetic acid, in the presence of carbon monoxide as one of the carbonylating agents."

Claims 1-3 according to the first, second and third auxiliary requests corresponded to claims 1-3 according to the main request with the addition of the temperature process conditions as "at temperatures below 100 °C" in the first auxiliary request, "at temperatures in the range of 80 - 100 °C" in the second auxiliary request and "at the typical temperature of 80 °C" in the third auxiliary request.

V. After receiving a communication accompanying the invitation to oral proceedings, in which *inter alia* objections according to Article 123(2) EPC were raised regarding the ranges of temperatures and pressures in the claims according to all requests and the limitation of claim 1 according to all request to a catalyst comprising only a vanadium complex, the appellants

informed the Board that they did not intend to attend the oral proceedings.

VI. Oral proceedings were held on 26 February 2010 in the absence of the appellants.

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

(a) the upper value of the pressure of 32 atm corresponds to the value of 27 atm of the examples, when the change in temperature between 25 and 80°C has been taken into account;

(b) the experiments with ¹³C-enriched methane mentioned in the application were conducted at low pressure, using commonly supplied bottles of ¹³C-enriched methane at 2 atm, which justifies the lower limit of 2 atm;

(c) the upper value of temperature of 100°C comes from the indication of the interval 100-500°C for the prior art and the intent to operate at lower values;

(d) the lower limit of 80°C for the temperature comes from the examples.

VIII. The appellants implicitly requested in writing that the decision under appeal be set aside and a patent be granted on the basis of one of the sets of claims according to the main request or to the first, second and third auxiliary requests, all filed with the statement setting out the grounds of appeal.

Reasons for the Decision

1. The appeal is admissible.
2. *Amendments*
 - 2.1 Claims 1-3 according to all requests contains *inter alia* an amendment relating to the process pressure conditions, which specifies that the process takes place at "pressures in the range of 2 - 32 atm".
 - 2.1.1 In order for the requirements of Article 123(2) EPC to be met a basis is necessary both for the range as such and for the specific values of the end points, wherein specific values only disclosed in an example can provide the required basis for such end points only if the skilled person could recognise without any doubt that they are not closely related to the other characteristics of the working examples and apply directly and unambiguously to the more general context.
 - 2.1.2 The application as originally filed does not disclose in the general part any range of values for the pressure of the process for the conversion of methane, while specific individual values are disclosed in the examples, which are run at values of the methane and carbon monoxide partial pressures as given in the second and third column of the table on pages 7 and 8 (3, 5, 8 and 12 atm for the methane partial pressure and 5, 15 and 20 atm for the partial pressure of carbon monoxide when present). Some of these values are mentioned also in the part of the description

discussing the results in the table (page 3, second paragraph to page 4, third full paragraph), where in addition a partial pressure of carbon monoxide of 8 atm is mentioned (page 4, second paragraph).

- 2.1.3 The cited passages and the table do not therefore provide any explicit basis for a range of total pressure nor for the specific end point values 2 and 32 atm.
- 2.1.4 The appellants argued that the experiments with ^{13}C -enriched methane mentioned in the application were conducted at low pressure, using commonly supplied bottles of ^{13}C -enriched methane at 2 atm, which justifies the lower limit of 2 atm and that the upper value of the pressure of 32 atm corresponds to the value of 27 atm of the examples, when the change in temperature between 25 and 80°C has been taken into account.
- 2.1.5 The application as filed indeed mentions experiments with ^{13}C -enriched methane in the paragraph bridging pages 3 and 4, but neither the value of the pressure at which they are conducted is given, nor an indication of the source of ^{13}C -enriched methane is mentioned. Therefore, these passages cannot be taken as a direct and unambiguous disclosure of the specific value of 2 atm. Moreover, experimental conditions which may have been used by the applicants while testing the invention, but which are not mentioned in the application cannot provide any basis for amendments.

2.1.6 It is also correct that in a couple of examples the partial pressures of methane and carbon monoxide sum up to 27 atm (eighth line concerning $[\text{VO}\{\text{N}(\text{CH}_2\text{CH}_2\text{O})_3\}]$ and third line concerning $\text{Ca}[\text{V}(\text{HIDA})_2]$ in the table on pages 7 and 8 respectively) and that the pressures are measured at 25°C (page 9, note *b*), while the experiments are conducted at 80°C (page 9, note *a*) after heating the system (page 4, last paragraph). However, a number of hypotheses need to be made in order to derive a value of 32 atm for the total pressure from these data (including at least that the mixture is an ideal mixture of gases, that the ideal gas law is valid, that the volume of the reaction system is constant and that no reaction occurs before reaching the temperature of 80°C), which cannot be taken as something that the person skilled in the art would assume in the absence of any information in the application and of any evidence on the side of the appellants.

2.1.7 For these reasons, it is concluded that there is no direct and unambiguous disclosure in the application as filed of the feature "at pressures in the range of 2 - 32 atm" present in all claims according to all requests, so that they contain subject-matter which extends beyond the content of the application as filed.

2.2 Claim 1 according to all requests concerns a catalyst comprising a vanadium complex with the metal in the +4 or +5 oxidation state, and with di- or poly-dentate ligands coordinated by nitrogen and oxygen (N,O) atoms or by oxygen and oxygen (O,O) atoms derived from aminoalcohols, (hidroxyimino)dicarboxylic acids, hydroxypyranones, trifluoroacetic acid or triflic acid.

The presence of a peroxodisulfate salt in the catalyst is not claimed and, while trifluoroacetic acid may be present as one of the possible ligands, it is not necessarily the case.

- 2.2.1 Original claim 1 related to a catalyst system for the direct one-pot conversion of methane into acetic acid containing together with a vanadium complex, a peroxodisulfate salt and trifluoroacetic acid. A corresponding disclosure is present in the description (see in particular page 2, last paragraph), where all described vanadium complexes are always disclosed and used in combination with a peroxodisulfate salt and trifluoroacetic acid for the desired conversion of methane into acetic acid.
- 2.2.2 Since no direct and unambiguous disclosure of a catalyst comprising the vanadium complex without any requirement for the presence of a peroxodisulfate salt and trifluoroacetic acid can be inferred from the original application, claim 1 according to all request extends beyond the content of the application as filed.
- 2.3 For these reasons, the claims according to all requests do not meet the requirements of Article 123(2) EPC.
3. Since all requests of the appellants fall on Article 123(2) EPC and the Board has only the power to decide on filed requests, it is not necessary that the Board takes a position on any other issue.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar

The Chairman

S. Fabiani

S. Perryman