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Datasheet for the decision of 29 August 2013

Case Number: T 1526/10 - 3.3.05

Application Number: 07014196.5

Publication Number: 1880758

IPC: B01J 8/06, C07C 51/25

Language of the proceedings: EN

Title of invention:
A method for gas-phase catalytic oxidation using a fixed bed reactor

Applicant:
Nippon Shokubai Co., Ltd.

Headword:
Treating agent/NIPPON SHOKUBAI

Relevant legal provisions:
EPC Art. 54(1)(2), 56, 84, 123(2)

Keyword:
"Clarity (yes): measurement method sufficiently characterised in the claims"
"Inventive step (yes): improvement not derivable from the state of the art"

Decisions cited:
-

Catchword:
-

Case Number: T 1526/10 - 3.3.05

DECISION
of the Technical Board of Appeal 3.3.05
of 29 August 2013

Appellant: (Applicant)	Nippon Shokubai Co., Ltd. 1-1, Koraibashi 4-chome, Chuo-ku, Osaka-shi, Osaka 541-0043 (JP)
Representative:	Albrecht, Thomas Kraus & Weisert Patent- und Rechtsanwälte Thomas-Wimmer-Ring 15 D-80539 München (DE)
Decision under appeal:	**Decision of the Examining Division of the European Patent Office posted 12 February 2010 refusing European patent application No. 07014196.5 pursuant to Article 97(2) EPC.**

Composition of the Board:

Chairman: G. Raths
Members: J.-M. Schwaller
C. Vallet

Summary of Facts and Submissions

I. This appeal lies from the decision of the examining division refusing European patent application No. 07 014 195.5.

Claim 1 of the main request (dated 8 September 2008) read as follows:

"*1. A method for gas-phase catalytic oxidation which is characterized by, in conducting gas-phase catalytic oxidation, using a fixed bed reactor wherein a treating agent for removing organic substance and/or carbides is disposed on the upstream side of the gas-phase oxidation catalyst layer in respect of the direction of the gas flow, in which method the adsorption capacity of the treating agent is at least 0.05% by mass, as measured by crotonaldehyde as an indicator of organic substance.*"

Claim 1 of auxiliary request 1 (dated 9 December 2009) read as follows:

"*1. A method for gas-phase catalytic oxidation which is characterized by, in conducting gas-phase catalytic oxidation, using a fixed bed reactor wherein a treating agent for removing organic substance and/or carbides is disposed on the upstream side of the gas-phase oxidation catalyst layer in respect of the direction of the gas flow, in which method the adsorption capacity of the treating agent is at least 0.05% by mass, as measured by crotonaldehyde as an indicator of organic substance wherein said treating agent is selected from the group consisting of silica alumina, silica-titania,* *silica-zinc oxide, silica-zirconia, alumina-titania, alumina-zinc oxide, alumina-zirconia, titania-zirconia, zinc oxide-zirconia, zeolite, magnesium carbonate and calcium carbonate.*"

Claim 1 of auxiliary request 2 (dated 26 January 2010) read as follows:

"*1. A method for gas-phase catalytic oxidation which is characterized by, in conducting gas-phase catalytic oxidation, using a fixed bed reactor wherein a treating agent for removing organic substance and/or carbides is disposed on the upstream side of the gas-phase oxidation catalyst layer in respect of the direction of the gas flow, in which method the adsorption capacity of the treating agent is at least 0.05% by mass, as measured by crotonaldehyde as an indicator of organic substance wherein said treating agent is selected from the group consisting of silica alumina, silica-titania, silica-zinc oxide, silica-zirconia, alumina-titania, alumina-zinc oxide, alumina-zirconia, titania-zirconia, zinc oxide-zirconia, zeolite, magnesium carbonate and calcium carbonate, the adsorption capacity the adsorption capacity being measured as follows:*
(i) 50 grams of the treating agent are filled in a fixed bed flowing apparatus and maintained at 350°C,
(ii) nitrogen gas after bubbling in crotonaldehyde maintained at 10°C is introduced thereinto from the upstream side of the treating agent at a rate of 170 ml/min for an hour,
(iii) after the adsorption treatment, the whole amount of the treating agent is heat-treated in air up to 500°C,

(iv) the mass change before and after the heat treatment is measured and
(v) the adsorption capacity of organic substance is determined by the following equation:
adsorption capacity of organic substance (mass%)
= [weight reduction (g)/treating agent (g)] x 100,
wherein the average diameter of the treating agent is within a range of 1 mm-15 mm."

II. The following two documents quoted during the examination proceedings are relevant for the present decision:

D1: EP 1 270 065 A1

D2: GB 2 063 861 A.

III. In the contested decision, the examining division rejected the main, first and second auxiliary requests *inter alia* under Article 84 EPC, because the method for measuring the adsorption capacity defined in the respective claims 1 was insufficiently described. The division explained that the method lacked information, on the one hand, as regards the geometry and the length of the apparatus used for determining the adsorption capacity, and on the other hand, with respect to the flow of air used in the post-adsorption heat treatment and the duration of this treatment.

IV. With the grounds of appeal dated 15 June 2010, the appellant maintained the sets of claims underlying the decision and contested in particular the clarity objection raised by the first instance.

V. In a communication dated 22 December 2012, the board expressed the opinion that the absence of details in the claims of the main and first auxiliary request regarding the method for measuring the adsorption capacity of the treating agent gave rise to a lack of clarity.

However, claim 1 of auxiliary request 2 which contained a clear definition of the measurement method appeared however to meet the requirements of Article 84 EPC.

In its communication, the board also cited a new document:

D3: US 5 051 244,

which in the board's opinion destroyed the novelty of the claims 1 of the main request and of auxiliary request 1. In particular, in the passage at column 5, lines 3 to 53, D3 disclosed a catalytic process for treating an automotive exhaust gas which fell under the wording of the claims 1 of these requests, because the feature "gas phase catalytic oxidation" in said claims was so broad that the oxidation of hydrocarbons and carbon monoxide occurring in this exhaust gas treating process was encompassed by the broad wording "gas phase catalytic oxidation" defined in the claims 1 under dispute. Further, the zeolite used as a hydrocarbon adsorbent in said process fell under the wording "treating agent" defined in said claims 1.

VI. With a letter dated 1 March 2013, the appellant cancelled the requests underlying the decision and filed an amended set of claims 1 to 4, claim 1 of which reads as follows:

"1. A method for gas-phase catalytic oxidation for producing (meth)acrylic acid through two-stage reaction of propylene, isobutylene, t-butyl alcohol or methyl-t-butyl alcohol which is characterized by, in conducting gas-phase catalytic oxidation, using a fixed bed reactor wherein a treating agent for removing organic substance and/or carbides is disposed on the upstream side of the gas-phase oxidation catalyst layer in respect of the direction of the gas flow, in which method the adsorption capacity of the treating agent is at least 0.05% by mass, as measured by crotonaldehyde as an indicator of organic substance wherein said treating agent is selected from the group consisting of silica-alumina, silica-titania, silica-zinc oxide, silica-zirconia, alumina-titania, alumina-zinc oxide, alumina-zirconia, titania-zirconia, zinc oxide-zirconia, zeolite, magnesium carbonate and calcium carbonate, the adsorption capacity being measured as follows:
(i) 50 grams of the treating agent are filled in a fixed bed flowing apparatus and maintained at 350°C,
(ii) nitrogen gas after bubbling in crotonaldehyde maintained at 10°C is introduced thereinto from the upstream side of the treating agent at a rate of 170 ml/min for an hour,
(iii) after the adsorption treatment, the whole amount of the treating agent is heat-treated in air up to 500°C,
(iv) the mass change before and after the heat treatment is measured and
(v) the adsorption capacity of organic substance is determined by the following equation:

adsorption capacity of organic substance (mass%) = [weight reduction (g)/treating agent(g)] x 100, wherein the average diameter of the treating agent is within a range of 1 mm — 15 mm and which uses a fixed bed reactor wherein the treating agent is disposed on the downstream side of the catalyst for the first stage reaction and on the upstream side of the catalyst for the second stage reaction, in respect of the direction of the gas flow."

Dependent claims 2 to 4 represent specific embodiments of the subject-matter of claim 1.

As a main and sole request, the appellant requested that the contested decision be set aside and that the case be remitted to the examining division with the order to grant a patent on the basis of this new set of claims.

Reasons for the Decision

1. *Amendments*

In the board's view, the subject-matter of claim 1 of the sole request on file is based on claims 1, 2, 6 and 7 as well as on the passages at page 6, lines 10 to 19; page 7, lines 28 to 32 and page 16, line 26 to page 17, line 2 of the application as filed.

Dependent claims 2, 3 and 4 have a basis in claims 3, 4 and 5 of the application as filed, respectively.

It follows that amended claims 1 to 4 of the sole request on file meet the requirements of Article 123(2) EPC.

2. *Clarity*

The details of the method for measuring the adsorption capacity of the treating agent were originally described at page 16, line 26 to page 17, line 2 of the application as filed. The introduction of these details into the subject-matter of claim 1 at issue now allows, in the board's view, that the measurement method be sufficiently characterised and that the boundaries of claim 1 be clearly defined.

The examining division's arguments as to the alleged absence of information regarding the reactor and the post-adsorption heat treatment are – in the board's view - unfounded, because as explained by the appellant, the determination of the adsorption capacity necessarily involved adsorption of the test substance to <u>saturation level</u> followed by <u>complete</u> removal of the substance. The point of achievement of the saturation was easily determinable by following the weight increase of the treating agent until it no longer increases due to the reaching of the saturation equilibrium state; similarly, the complete removal of the substance upon heat treatment was also easily determinable by following the weight decrease of the treating agent until it no longer decreased, because the desorption equilibrium state was achieved. The factors that the examining division considered to have an influence on the results of the capacity measurement and for which it raised an alleged lack of information can be put aside for the following reasons:

- It is well known to the skilled person that the adsorption capacity in the equilibrium state depends only on the concentration and temperature of the adsorbate, and so the shape and length of the apparatus has no particular influence on the measurement results.

- The heat-treatment is conducted for the purpose of eliminating the crotonaldehyde adsorbed by the treating agent; it follows that the heat treatment has to be carried out so that the amount of air passed is sufficient for the elimination of adsorbed crotonaldehyde, and as a matter of common sense, the treatment is to be carried out until the mass of treating agent reaches the equilibrium conditions (i.e. does no longer change). It follows that the particular details regarding the flow of air and the duration of the heat-treatment also will have no influence on the measurement results.

Thus, based on common general knowledge and on the guidance given in the description, the skilled person can easily conduct the adsorption capacity measurement to obtain results which are quite independent of the factors that the examining division considered to have a decisive influence.

It follows that claim 1 meets the requirements of Article 84 EPC.

3. *Novelty*

The board no longer maintains its novelty objection based on D3, since the claimed subject-matter has been substantially restricted by the specification of the oxidation reaction (*production of (meth)acrylic acid through a two-stage reaction of propylene, isobutylene, t-butyl alcohol or methyl-t-butyl alcohol*), and the further specification of the average diameter (*within a range of from 1 to 15 mm*) and of the location of the treating agent (*downstream the catalyst for the first reaction stage and upstream the catalyst for the second reaction stage*) and, in the meantime, has been rendered novel by introduction of these features into claim 1 at issue.

Further, neither D1 nor D2 discloses at least the type of treating agent, its adsorption capacity and its specific location in a process for producing (meth)acrylic acid through a two-stage oxidation reaction of propylene, isobutylene, t-butyl alcohol or methyl-t-butyl alcohol.

4. *Inventive step*

By applying the problem-solution approach, the board came to the following conclusions.

4.1 The invention concerns a method for gas-phase catalytic oxidation using a fixed bed reactor, in particular a method for producing acrylic or methacrylic acid (collectively referred to as "meth(acrylic acid)) wherein in a first stage the hydrocarbons are converted into unsaturated aldehydes by gas phase catalytic oxidation, and then in a second stage the unsaturated aldehydes are converted into meth(acrylic) acid (page 1, lines 4 to 16 of the application in suit).

4.2 As to the starting point for assessing inventive step, the board considers that document D2 represents the closest state of the art.

D2 (claim 1) discloses a two-stage gas phase catalytic oxidation process for producing acrylic acid comprising subjecting a mixture of propylene, molecular oxygen, and steam to a first-stage catalytic oxidation for converting propylene into acrolein and subjecting the gases thus formed to a second-stage catalytic oxidation to convert the acrolein into acrylic acid. The process according to D2, claim 1, is *inter alia* characterised in that the first-stage catalytic oxidation reaction is carried out in a reaction zone comprising a plurality of elongated unit reaction zones extending in the direction of flow of the gases and connected in parallel arrangement, each of the unit reaction zones comprising a reaction region comprising a bed of an oxidation catalyst and a cooling region comprising a bed of a solid inactive material disposed contiguously to the reaction region on the downstream side thereof, and the temperatures of the reaction region and of the cooling region being controlled independently.

The solid material used in the downstream cooling region is supposed to be inactive with respect to propylene, acrolein, and acrylic acid at the vicinity of the catalytic oxidation reaction temperature. Specific examples of such materials are α-alumina, alundum, mullite, carborundum, stainless steel, copper, aluminum, and ceramics (D2, page 4, lines 31, 32 and 37 to 42). A bed of a similar solid and inactive material can be provided on the upstream side of the catalyst bed thereby forming a gas preheating region (D2, page 4, lines 17 to 19).

4.3 As to the technical problem to be solved by the invention, this is defined in the application as filed (page 2, lines 20 to 25) to be the provision of a process for producing (meth)acrylic acid by gas phase catalytic oxidation which enables stable continuous operation over a prolonged period while maintaining high yield level and suppressing increase in the pressure loss, in other words the provision of an improved process.

4.4 As a solution to the above problem, the invention proposes the process according to claim 1 at issue, which is in particular characterised in that a treating agent for removing organic substance and/or carbides is disposed on the downstream side of the catalyst for the first stage reaction and on the upstream side of the catalyst for the second stage reaction, the treating agent having an adsorption capacity of at least 0.05% by mass as measured by crotonaldehyde as an indicator of organic substance, and said treating agent being selected from the group consisting of silica-alumina, silica-titania, silica-zinc oxide, silica-zirconia, alumina-titania, alumina-zinc oxide, alumina-zirconia, titania-zirconia, zinc oxide-zirconia, zeolite, magnesium carbonate and calcium carbonate, and the average diameter of the treating agent being within a range of 1 to 15 mm.

4.5 As to the question whether the above problem has been solved by the solution proposed in claim 1 at issue, the results in Table 1 of the application show that, in comparison to a treating agent with an adsorption capacity of 0.01% (such as the ceramic balls in examples 11 and 12), those agents having an adsorption capacity above the threshold value defined in claim 1 (see examples 9, 10 and 13 to 19) provide for a higher yield in acrylic acid over a prolonged period and a lower pressure loss at the second stage of the process. In view of this evidence for an improvement the board concludes that the above problem is successfully solved.

4.6 On the question whether the solution proposed in claim 1 at issue was derivable from the state of the art, the following comments apply:

- Documents D1 and D2 both disclose a two-step gas-phase catalytic oxidation method for producing (meth)acrylic acid in a shell-and-tube heat exchanger type reactor making use of solid inert particles making use of a cooling zone comprising solid inactive material upstream and downstream of the reaction zone.

- D3 relates to a process for treating an engine exhaust gas stream.

None of the documents now on file discloses the characterising features defined in point 4.4 above, let alone with the aim of solving the problem identified under point 4.3 above.

It follows that the skilled person faced with the problem underlying the application would not derive the solution proposed in claim 1 from the state of the art, let alone would arrive in an obvious manner at the subject-matter defined in claim 1.

Therefore, having regard to the state of the art, the subject-matter of claim 1 at issue, and by the same token that of dependent claims 2 to 4, which includes all the features of claim 1, is not obvious to a person skilled in the art and thus meets the requirements of Article 56 EPC.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the first instance with the order to grant a patent on the basis of the claims filed with letter dated 1 March 2013.

The Registrar:	The Chairman:
C. Vodz	G. Raths