DECISION
of 6 July 2004

Case Number: T 0094/03 - 3.2.7
Application Number: 93111528.1
Publication Number: 0582121
IPC: C23G 1/08
Language of the proceedings: EN

Title of invention:
Process for stainless steel pickling and passivation without using nitric acid

Patentee:
Henkel Kommanditgesellschaft auf Aktien

Opponents:
Eka Chemicals AB
CONDOROIL IMPIANTI s.r.l.
USINOR
Solvay (Société Anonyme)

Headword:
-

Relevant legal provisions:
EPC Art. 100(b), 114(1), 114(2)

Keyword:
"Late submitted material - documents partly admitted"
"Amended requests filed about one month before date of oral proceedings - admitted"
"Disclosure - enabling (no)"

Decisions cited:
T 0651/90

Catchword:
-

EPA Form 3030 06.03
Case Number: T 0094/03 - 3.2.7

DECISION
of the Technical Board of Appeal 3.2.7
of 6 July 2004

Appellant I: Henkel Kommanditgesellschaft auf Aktien
(Proprietor of the patent) D-40191 Düsseldorf (DE)

Representative: Gervasi, Gemma, Dr.
NOTARBARTOLO & GERVASI Srl,
Corso di Porta Vittoria, 9
I-20122 Milano (IT)

Appellant II: USINOR
(Opponent III)
Immeuble "La Pacific"
11 Cours Valmy
TSA 10001 - La Défense 7
F-92070 La Défense Cedex (FR)

Representative: Bernasconi, Jean Raymond
c/o Cabinet Lavoix,
2, Place d'Estienne d'Orves
F-75441 Paris Cedex 09 (FR)

Decision under appeal: Interlocutory decision of the Opposition
Division of the European Patent Office posted
22 November 2002 concerning maintenance of
European patent No. 0582121 in amended form.

Composition of the Board:
Chairman: A. Burkhart
Members: H. E. Hahn
E. Lachacinski
Summary of Facts and Submissions

I. The patentee and opponent III lodged appeals against the decision of the Opposition Division to maintain the European patent No. 0 582 121 in amended form on the basis of claims 1 to 5 according to the second auxiliary request.

II. Oppositions by four opponents had been filed against the patent as a whole and were based on Article 100(a) EPC (lack of novelty and lack of inventive step) and Article 100(b) EPC (that the patent did not disclose the invention in a manner sufficiently clear and complete for it to be carried out by a person skilled in the art) and Article 100(c) EPC (matter extending beyond the content of the application as filed). Opponent I withdrew its opposition with a letter dated 21 December 2001.

The Opposition Division held that that all oppositions were admissible and that the invention was sufficiently disclosed. The Opposition Division further concluded that the subject-matter of the independent claims 1 of the main and first auxiliary request comprised inadmissible generalisations of an example and thus contravened Article 123(2) EPC. Claim 1 of the second auxiliary request was considered to meet the requirements of Articles 84 and 123(2) and (3) EPC. Furthermore, the priority was considered to be validly claimed and the novelty and inventive step of the subject-matter of claim 1 of the second auxiliary request were acknowledged.
III. With letter of 31 March 2003 opponent II withdrew its appeal and its opposition.

IV. With fax of 18 June 2004 opponent IV withdrew its opposition.

V. Oral proceedings were held on 6 July 2004.

(a) Appellant I (patentee) requested that the decision under appeal be set aside and that the patent be maintained on the basis of the sets of claims according to either the main request, or alternatively according to the first to ninth auxiliary request as filed on 4 June 2004 with letter of 3 June 2004.

(b) Appellant II (opponent III) requested that the decision under appeal be set aside and the patent be revoked in its entirety.

VI. The decision is based on the following documents:

D3: GB-A-2 000196


D24: EP-A-0 541 448

D37: Elektrochemie, H. Ebert, Vogel-Verlag, 2nd Edition 1979, pages 88 and 89, tables 19-20 (= enclosure 9)

Annex 2: Declaration of Prof. Achille DeBattisti


Enclosure 6: Potential vs. Temperature diagram, Dr W. Ingold KG-Frankfurt data (derived from enclosure 5)

Enclosure 7: "Skoog-Leary-Graph", graphical display of Table 20-1 from page 490 of enclosure 4

Enclosure 8: Handbook of Chemistry and Physics, 77th Edition, 1996-1997, pages 8-81, 8-84 and 8-86

Enclosure 10: "Skoog-Küster-Graph", with respective tables (graphically displaying the combined values of the Skoog-Leary-Graph and the Küster-Thiel-Graph

VII. The independent claims 1 and 4 of the main request as filed on 4 June 2004 read as follows (differences to claim 1 as granted are in bold):

"1. Process for the sole pickling of stainless steel providing only for descaling and dechromized surface
layer removal, consisting in placing the material to be treated in a bath kept at a temperature ranging from 30°C to 70°C, having the following initial composition:

a) H$_2$SO$_4$ at least 150 g/l
b) Fe$^{3+}$ at least 15 g/l
c) HF at least 40 g/l
d) H$_2$O$_2$ 35% by w. added with known stabilizers, 1-20 g/l
e) additives of the non-ionic surfactant class (emulsifiers, wetting agents, polishing agents) as well as of the acid attack inhibitor class: approx. in a whole amount of 1 g/l, in the bath being continuously fed: an air flow equal to at least 3 m$^3$/h per m$^3$ bath, through a diffuser distributing the flow in the liquid mass, and if required, quantities of ingredients a) and c) securing a concentration of at least 80 g/l of H$_2$SO$_4$ and of HF in an amount to give a concentration of free fluoride in the bath of at least 25 g/l and a bath pH below 1.5, and of additives e) in order to secure the optimal concentration of 1 g/l (as whole amount), and characterized in that a stabilized H$_2$O$_2$ (35% by w.) is fed continuously in the bath in quantity adjusted to keep the redox potential of the bath at a value of at least 250 mV and less than 350 mV."

"4. Process for the sole pickling of stainless steel providing only for descaling and dechromized surface layer removal, consisting in placing the material to be treated in a bath kept at a temperature ranging from 30°C to 70°C, having the following initial composition:

a) H$_2$SO$_4$ at least 150 g/l
b) Fe$^{3+}$ at least 15 g/l
c) HF at least 40 g/l
d) H$_2$O$_2$ 35% by w. added with known stabilizers, 1-20 g/l
e) additives of the non-ionic surfactant class (emulsifiers, wetting agents, polishing agents) as well as of the acid attack inhibitor class: approx. in a whole amount of 1 g/l, in the bath being continuously fed: an air flow equal to at least 3 m³/h per m³ bath, through a diffuser distributing the flow in the liquid mass, and if required, quantities of ingredients a) and c) securing a concentration of H₂SO₄ between 100 and 150 g/l, free fluoride (added as HF) between 20 and 30 g/l, and a bath pH below 1.5, and of additives e) in order to secure the optimal concentration of 1 g/l (as whole amount), and characterized in that a stabilized H₂O₂ (35% by w.) is fed continuously in the bath in quantity adjusted to keep the redox potential of the bath at a value of at least 250 mV and less than 350 mV."

VIII. Appellant I argued essentially as follows:

The late filed documents illustrate the general knowledge of the skilled person and are taken from standard text books. These documents are a reaction to the communication of the Board annexed to the summons for the oral proceedings and should therefore be allowed into the proceedings. Similarly, the ten requests represent a reaction to the Board's opinion presented in said communication and should therefore be allowed.

It is doubtful whether the normal hydrogen electrode (NHE) represents the standard to which redox potentials are quoted since in the practical environment of a real plant (i.e. non-laboratory environment) it causes problems. Although many documents do not quote the reference electrode at all, most of those which specify
the same refer to an Ag/AgCl electrode (cf. e.g. D24, column 2, lines 36 to 45; D3 or D11). The skilled person would expect that said redox potential of 700 mV of the initial pickling bath according to Table 1 of the application as filed has been measured at the working temperature of the pickling experiments, i.e. 50°C. Furthermore, the skilled person would immediately realize that the NHE would not fit with the disclosed 700 mV potential of the disclosed pickling bath according to said Table 1 of the application as filed. Thus, there would be only the choice between the most common Ag/AgCl or calomel reference electrodes, the latter one having a potential which is 44 mV higher than the other. For the reasons given by Prof. Battisti the skilled person would select the Ag/AgCl reference electrode. It is admitted that there exists a contradiction concerning the upper working temperature limit of the calomel electrode (cf. enclosures 4 and 5). Although an inner electrolyte of NaCl can be used for measuring of a redox potential the skilled person would not use the same because it was unreliable (cf. the reasons given by Prof. Battisti). The potential value of "approx. 700 mV" according to Table 3 of the application as filed for the same pickling bath can be derived by using the slopes for the said reference electrode (see e.g. enclosures 6, 7 or 10), and leads, since the quoted temperature of 30-35°C is lower, to a lower redox potential of about 675-680 mV. Thus, there exists no inconsistency between these two potential values. The burden of proof for the insufficient disclosure lies with the opponent who failed to submit the corresponding evidence. The skilled person can check via electrochemical experiments whether a work piece has been only pickled or additionally has been
passivated. Therefore the patent enables the skilled person to carry out the claimed process.

IX. Appellant II argued essentially as follows:

The late filed facts and evidence submitted by the patentee with letter of 3 June 2004 should not be introduced into the procedure because they were late filed and not relevant. Due to the late filing of said evidence the appellant was unable to produce a counter-technical opinion with respect to the technical opinion of Prof. Battisti (i.e. Annex 2 of said letter dated 3 June 2004) which should only be considered as one party's opinion. Appellant II had insufficient time to verify the experiments of Annex 1. The ten new sets of claims filed on 4 June 2004 should also not be allowed because they were, together with said evidence, obtained on 16 June 2004 since appellant I had submitted them only by post and not by fax. This action was considered to be unfair and represented an abuse of procedure.

The patent as granted does not enable the skilled person to carry out the claimed pickling process of claim 1. Claim 1 comprises as an essential feature that a redox potential of the pickling bath should be maintained in a range of from 250 to less than 350 mV but the patent fails to specify which reference electrode has to be used. Also the application as filed does not specify which reference electrode has to be used. The standard with respect to which redox potentials are quoted unless otherwise stated is the normal hydrogen electrode (NHE), which potential at standard conditions by definition is set at 0 mV.
Therefore the skilled person would interpret the said redox potential as being referred to NHE. Thereby the skilled person would carry out a process which is outside a redox potential range intended to be used by the patent in suit. Consequently, the skilled person cannot put the invention into practice. Even taking account of the example according to Table 1 of the application as filed the skilled person cannot conclusively derive that an Ag/AgCl reference electrode charged with an inner electrolyte of 3 M KCl should have been used in combination with a platinum measuring electrode. The application as filed, although mentioning a redox potential of 700 mV of the initial pickling bath of said example, neither unambiguously specifies the temperature at which said potential was measured – it is not conclusive that the temperature was the same at which the pickling experiments were carried out, it could be any temperature – nor does it specify all ingredients of the pickling bath, i.e. the counter ions of the ferric ions are not specified as well as the type of the "additives" comprised in the said bath. These components, however, influence the redox potential of the bath. The skilled person could have chosen a calomel reference electrode as suggested by document D3. The redox potential difference of such a calomel electrode with an inner electrolyte of saturated KCl compared with an Ag/AgCl having the same inner electrolyte is only 44 mV. Furthermore, as admitted by the patentee (cf. letter of 3 June 2004, page 9, last paragraph) the claimed process may sometimes result at least for some types of stainless steel in passivated work pieces below 350 mV. The arguments of Prof. Battisti are not particularly relevant since the knowledge of an expert is not the
same as that of the skilled person and even he needs a reasoning of nine pages to arrive at the said Ag/AgCl reference electrode. It has not been proven that everybody uses a 3 M or 3.5 M KCl electrolyte solution as the inner electrolyte. As can be derived from document D29 an expert did not succeed in properly pickling stainless steel according to the process claimed in the patent in suit (cf. D29: Declaration of Mr Zavattoni, page 1, point 3; page 5, third paragraph; page 6, first paragraph). The skilled person is thus unable to carry out the process of the patent in suit which therefore does not meet the requirements of Article 100(b) EPC.

Reasons for the Decision

1. Admissibility of late filed documents Annex 1, Annex 2 and enclosures 1 to 10

1.1 Appellant I submitted for the first time on 4 June 2004, i.e. three days later than the one month time limit set prior to the date of the oral proceedings before the Board, the Annexes 1 and 2 together with the enclosures 1 to 10 with its letter dated 3 June 2004. Hence these documents are to be treated as late filed.

1.2 The Board concurs with the arguments of appellant I that the documents Annex 2 and enclosures 1 to 10 (wherein enclosure 1: Curriculum vitae of Prof. Battisti; enclosure 2: Publication list of Prof. Battisti; enclosure 3: "Sufficiency of Disclosure" statements made by Henkel KGaA) were submitted in order to support appellant I's position with respect to the
Board's negative opinion concerning an insufficiency of disclosure. Furthermore, these documents (namely enclosures 4 to 10) represent common general knowledge which is taken from standard text books. The Board takes into consideration the expert opinion of Prof. Battisti according to said Annex 2 as well as enclosure 3.

The Board in view of the above consideration exercises its discretion under Article 114(1) EPC and introduces the documents Annex 2 and the enclosures 1 to 10 into the proceedings.

1.3 The Board concurs with appellant II that it would be unfair to introduce the late filed document Annex 1, concerning experimental redox potential values of several pickling bath composition, into the proceedings because appellant II had not enough time to repeat and verify the experiments described therein.

As a consequence the Board exercises its discretion and disregards Annex 1 in accordance with Article 114(2) EPC.

2. **Admissibility of appellant I's requests**

2.1 With the letter dated 3 June 2004 appellant I filed a new main request and nine auxiliary requests on 4 June 2004.

2.2 The Board in exercising its discretion decides to admit the said ten requests filed on 4 June 2004 into the proceedings, which were filed as a reaction to the Board's negative opinion with respect to the
Article 123(2) issue as set out in the communication annexed to the summons for the oral proceedings.

3. Lack of enabling disclosure (Article 100(b) EPC)

3.1 Claim 1 of the main request comprises as an essential feature that the redox potential of the pickling bath should be maintained in a range of from 250 to less than 350 mV. Although this feature is most essential for the patent the specification fails to specify that a reference electrode and particularly which reference electrode has to be used. The same is valid for the application as filed which similarly is totally silent in this respect. The decision T 651/90 cited by appellant I is not considered to be relevant for the present case since the patent in suit does not contain any reference to any standard procedure, let alone one using a specific reference electrode, for determining the redox potential.

The patentee admitted that the feature of the redox potential represents an essential feature of the claimed stainless steel pickling process. During the opposition procedure the patentee had declared for the first time that the values specified in the patent in suit are with reference to the \([\text{Pt/Ag/AgCl/Cl}^-]\) electrode (cf. letter dated 31 August 2001, paragraphs 7.2 to 7.5) and only as a reaction to the Board's communication dated 18 March 2004 did the patentee specify with its letter of 3 June 2004 that the inner electrolyte of the said reference electrode is a solution of 3 M KCl.
3.2 The standard with respect to which redox potentials are quoted unless otherwise stated is the normal hydrogen electrode (NHE), which potential at standard conditions (25°C) by definition is set at 0 mV. It has also to be considered that redox potentials may be measured using any reference electrode and thereafter the measured potentials may be recalculated to refer to those of the NHE. Therefore the skilled person would normally interpret the said redox potentials of the patent – since no reference electrodes are quoted – as being referred to NHE. Thereby the skilled person would carry out a process which is outside any redox potential range defined in claim 1 intended to be used by the patentee.

3.2.1 Also the result to be achieved by the pickling process of claim 1, i.e. the desired effect of (sole) pickling of stainless steel without any passivation thereof (i.e. only descaling and dechromized surface layer removal), does not help the skilled person in finding the reference electrode. If the skilled person would use the NHE as reference he could carry out a pickling process using a bath composition falling under the definition of claim 1 and would obtain pickled stainless steel work pieces which would not be passivated since the redox potential of the used pickling bath would be well below the critical passivation potential of the stainless steel.

3.2.2 The Board concurs with appellant I that the skilled person can check via electrochemical experiments whether a work piece has been only pickled or additionally has been passivated. However, these experiments do not enable the skilled person to derive
the redox potential at which said work piece has been solely pickled (i.e. if it has not been passivated) and whether it has been pickled within the intended redox potential range (e.g. close to 250 mV), or not (e.g. at 230 mV).

3.3 Appellant I argued that the skilled person would derive the reference electrode to be used taking account of the example according to Table 1 of the application as filed.

The Board is not convinced that the skilled person would conclusively only select an Ag/AgCl reference electrode charged with an inner electrolyte of 3 M KCl in combination with a platinum measuring electrode.

3.3.1 The application as filed mentions a redox potential of 700 mV for an initial pickling bath which was prepared to contain 150 g/l H$_2$SO$_4$, 50 g/l HF, 15 g/l Fe$^{3+}$, 5 g/l stabilized H$_2$O$_2$ and 1 g/l additives (cf. Table 1, example). Table 1 further reveals that said pickling bath was used to determine the weight loss at 50°C (cf. Table 1).

3.3.2 Thus, the specification fails to define all the ingredients of the said pickling bath since neither the counter ions of the ferric ions nor the specific type of the said "additives" comprised in the pickling bath are specified. These components, however, influence the redox potential of the bath.

3.3.3 Furthermore, in the Board's view the specification does not unambiguously specify the temperature at which said initial potential was measured. Although appellant I
argued that the temperature for measuring the redox potential of the initial pickling solution was the same temperature of 50°C at which the pickling experiments were carried out the Board is not convinced that it is conclusively the same temperature. Particularly, since an identical pickling bath (cf. Table 3) – which is used for pickling experiments at a lower temperature of 30-35°C – is stated to have an initial redox potential of "approx. 700 mV". Appellant I argued in this context that the term "approx. 700 mV" actually would mean a value of "about 675-680 mV" as can be derived, e.g. from the curve according to the "Potential vs. Temperature" diagram of enclosure 6. It appears to be more credible to the Board that in such a case the skilled person would specify the redox potential to be "approx. 680 mV" rather than "approx. 700 mV". Hence these arguments are not convincing.

As a consequence, the skilled person does not know at which temperature said initial redox potential of 700 mV of the pickling bath has been measured.

The identification of the reference electrode to be used is not possible when the exact temperature for measuring the initial redox potential is not known, let alone when the composition of the pickling bath thereof is not totally defined.

3.3.4 Assuming that the skilled person realizes that the NHE is not the intended reference electrode, he would most presumably chose one of the most common reference electrodes, namely either a calomel electrode or a Ag/AgCl electrode in combination with a measuring electrode (compare e.g. D3, page 2, lines 50 to 54; D37,
pages 87 to 88; D38, page 425). According to document D3 any electrode if only they are inert to the pickling solution, for example noble metals such as Pt, Au or Rh, may be used as the measuring electrode (cf. D3, page 2, lines 50 to 54).

3.3.5 The skilled person thus could have chosen a saturated calomel reference electrode in combination with a Pt measuring electrode as suggested by document D3 (cf. page 2, lines 57 to 61). The redox potential difference of such a calomel electrode with an inner electrolyte of saturated KCl compared with an Ag/AgCl having the same - saturated KCl - inner electrolyte is about 44 mV (depending upon the literature cited). In this context it should be borne in mind that the temperature range of from 30-70°C according to the pickling process of the patent in suit implies a possible change of the redox potential which can be in the same order of about 40 mV. This is on the one hand caused by the temperature dependency of the pickling bath and on the other hand by the temperature dependency of the reference electrode.

Taking account of the fact, that the temperature at which the redox potential of 700 mV of the initial pickling bath has been measured is not specified, and that the temperature according to the Nernst equation $E=E_0 + \frac{RT}{zF} \cdot \ln \frac{[ox]}{[red]}$ has a great impact on the value of the redox potential (compare in this context document D3, page 2, lines 47 to 49) it is credible that the skilled person could have chosen a calomel electrode.
3.3.6 In such a case the skilled person would not even have realized that he is using a wrong reference electrode because he could pickle the stainless steel within the meaning of the patent in suit, i.e. he could carry out a sole pickling process without any passivation. This is due to the fact that there exists a broad overlap of the relative redox potentials (compare also paragraph 3.2.2 above). Furthermore, as admitted by the patentee the claimed process may sometimes, at least for some types of stainless steel, result in passivated work pieces below a redox potential of 350 mV (cf. letter of 3 June 2004, page 9, last paragraph).

3.3.7 The Board concurs with appellant II that the arguments of Prof. Battisti with respect to the selection of a particular reference electrode and more particularly of a specific inner electrolyte are not particularly relevant since the knowledge of a scientific expert is not the same as that of a skilled person. Even Prof. Battisti requires a lengthy reasoning to arrive at the said Ag/AgCl reference electrode having an inner electrolyte of 3 M KCl.

It has also to be considered that the change of solubility of NaCl with the change of temperature is much smaller than that of KCl (cf. enclosure 8, pages 8 to 84 and 8 to 86) so that the temperature dependency of a reference electrode containing the saturated NaCl electrolyte is smaller than that containing the KCl electrolyte.

Therefore in the Board's view the skilled person would also consider to use an Ag/AgCl reference electrode with an inner electrolyte of NaCl which is also
commercially available. In the case of a saturated NaCl solution this would lead to a potential of the Ag/AgCl reference electrode which is about 10 mV higher than that of an identical electrode but containing a saturated KCl solution (cf. D38, page 425). Hence also this reference electrode would have allowed the skilled person to carry out the sole pickling process of the patent in suit. The conclusions of paragraph 3.4.6. above thus apply *mutatis mutandis* to the Ag/AgCl reference electrode containing a saturated NaCl electrolyte.

The Board is also not convinced that the person skilled in the art would select a less concentrated 3 M or 3.5 M KCl solution which would result in an estimated potential difference of only about ±5 mV over the temperature range of from 30 to 70°C for the Ag/AgCl electrode.

Saturated solutions have the advantage that the concentration is reproducible even if the temperature changes (provided that solid salt is present) and that they are immune to water evaporation. A nearly saturated solution, such as the 3 M or 3.5 M KCl solution, has the disadvantage that the concentration can change due to evaporation of water. Since the electrode is to be used at higher temperatures the said evaporation could be an issue so that saturated solutions could be preferred.

Pickling baths are made from demineralised water and the acids (i.e. technical grade H₂SO₄ and HF) and thus will not contain any substantial amounts of Cl⁻-ions so that the skilled person could equally select a less
concentrated 1 M KCl solution as the inner electrolyte. This would result in a redox potential which is only about 8 mV smaller than that of the calomel electrode containing a saturated KCl electrolyte (cf. enclosure 5, page 146, penultimate table).

Furthermore, as is evident from the several documents (cf. enclosures 4 and 5, documents D37 and D38) many different concentrations of the inner electrolyte are known and would actually be suitable for measuring the redox potential of the said pickling bath. However, the standard text books do not provide clear suggestions as to how to select the most appropriate one.

In the context of the Ag/AgCl electrode, enclosure 5 only discloses concentrations of 1 a and particularly of 1 M and 3 M (cf. page 146, tables), while document D37 (cf. page 89, Table 20) only mentions 1 a, 0.1 n (= 0.1 M) and 1 n (= 1 M) which result in standard potentials of 222.3 mV, 289.4 mV and 236.8 mV, respectively, whereas document D38 only mentions the saturated electrolytes thereof (cf. page 425).

According to enclosure 4 (cf. page 491, right-hand column, second paragraph) Ag/AgCl electrodes are normally prepared with a saturated or 3.5 M KCl solution which would result at 25°C in a potential of 199 mV for the saturated KCl electrolyte and of 205 mV for the 3.5 M KCl electrolyte, respectively (cf. enclosure 4, Table 20-1). These values have to be compared with a potential of 207 mV of the intended Ag/AgCl reference electrode containing the 3 M KCl electrolyte (cf. enclosure 10, Table).
Consequently, the conclusions of paragraph 3.3.6. above apply mutatis mutandis to the Ag/AgCl reference electrode containing a 1 M or a saturated KCl electrolyte.

3.4 Taking account of the paragraphs above, no "one-way street" situation exists which conclusively leads the skilled person to the intended Ag/AgCl reference electrode containing the 3 M KCl electrolyte.

3.5 The Board therefore considers that the skilled person is unable to carry out the process of the patent in suit. Consequently, the ground of opposition according to Article 100(b) EPC prejudices the maintenance of the patent in suit.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The patent is revoked.

The Registrar: 

The Chairman:

G. Nachtigall 
A. Burkhart