BESCHWERDEKAMMERN DES EUROPÄISCHEN PATENTAMTS

BOARDS OF APPEAL OF THE EUROPEAN PATENT OFFICE

CHAMBRES DE RECOURS DE L'OFFICE EUROPEEN DES BREVETS

Publication in the Official Journal [redacted] / No

File Number: T 871/90 - 3.4.2

Application No.: 84 307 667.0

Publication No.: 0 145 262

Title of invention: Method for removing dissolved oxygen from acqueous media

Classification: B01D 19/00, C02F 1/20, C23F 11/18, B01J 31/08

DECISION

of 25 June 1992

Proprietor of the patent: WESTINGHOUSE ELECTRIC CORPORATION

Opponent:
01) Bayer AG, Leverkusen Konzernverwaltung RP Patente Konzern
02) Hoechst AG

Headword:

EPC Article 56 EPC

Keyword: "Inventive step (no)"

Case Number : T 871/90 - 3.4.2

DECISION
of the Technical Board of Appeal 3.4.2
of 25 June 1992

Appellant : (Proprietor of the patent)	WESTINGHOUSE ELECTRIC CORPORATION Westinghouse Building Gateway Center Pittsburgh Pennsylvania 15235 (US)
Representative :	van Berlyn, Ronald Gilbert 23, Centre Heights London, NW3 6JG (GB)
Respondent : (Opponent 01)	Bayer AG, Leverkusen Konzernverwaltung RP Patente Konzern Bayerwerk W - 5090 Leverkusen (DE)
Respondent : (Opponent 02)	HOECHST AKTIENGESELLSCHAFT, Frankfurt (Main) Werk Ruhrchemie Postfach 13 01 60 W - 4200 Oberhausen 11 (DE)
Decision under appeal :	Decision of Opposition Division of the European Patent Office dated 29 August 1990 revoking European patent No. 0 145 262 pursuant to Article 102(1) EPC.

Composition of the Board :

Chairman : E. Turrini
Members : C. Black
L.C. Mancini

Summary of Facts and Submissions

I. European patent application No. 84 307 667.0, (publication No. 0 145 262), was granted on 9 March 1988.

II. The patent was opposed by Bayer AG (Opponent I and Respondent I) and Hoechst AG (Opponent II and Respondent II). Both Opponents requested revocation of the patent on the ground of Article 100(a) EPC. Opponent I also submitted 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 (Article 100(b) EPC).

In respect of the objection under Article 100(1) EPC the oppositions were supported by the following documents:

(D1) DE-C-1 201 262;
(D2) DE-C-1 112 047;
(D3) DE-A-1 642 494;
(D4) VGB Speisewassertagung, 1967, pp. 53 to 48, H. Kurapkat, "Die katalytische Reduktion von Sauerstoff in wäßriger Lösung";
(D5) 41^{st} annual meeting of the International Water Conference, Pittsburgh, Pennsylvania, October 20, 21 and 22, 1980, pp. 3 to 8, F.B. Martinola et al., "Saving energy by catalytic reduction of oxygen in feedwater";
(D6) VGB Kraftwerkstechnik 61, Vol. 1, January 1981, pp. 59 to 61, P. Thomas, "Betriebserfahrungen mit der katalytischen Reduktion von Sauerstoff in Wasser";
(D7) EP-A-0 065 687;
(D8) VGB Konferenz "Chemie im Kraftwerk", October 26 and 27, 1983, pp. 11 to 13, F. Martinola et al., "Ein

starkbasischer Anionenaustauscher mit zwei Funktionen";

(D9) Symposium on determination of dissolved oxygen in water, 60th ASTM annual meeting, Atlantic City, N.J., June 20, 1957, pp. 3 and 12 of "A study of the accuracy of methods of testing for dissolved oxygen in high-purity water" by K.G. Stoffer;

(D10) Manual on industrial water and industrial waste water, 2nd Edition, 1965, ASTM special technical publication No. 148-H, pp. 309 to 320, "Tentative methods of test for dissolved oxygen in industrial water";

(D11) M.L. Hitchman, Measurement of dissolved oxygen, John Wiley & Sons, 1978, pp. 172 and 173, "Summary of chemical methods"; and

(D12) Standard methods for the examination of water and waste water, 11th Edition, American Public Health Association, Inc., 1962, pp. 16 to 19, "Expression of results" and "Precision and accuracy".

III. By decision of 29 August 1990 the Opposition Division revoked the European patent pursuant to Article 102(1) EPC.

The Opposition Division held that the contested patent disclosed the invention in a manner sufficiently clear and complete for it to be carried out by a person skilled in the art but that Claim 1 did not contain inventive subject-matter.

IV. The Proprietor (Appellant) lodged an appeal against this decision on 5 November 1990. The Statement of Grounds of Appeal and an amended Claim 1 were filed on 8 January 1991.

V. In accordance with requests submitted by the Appellant and the Respondent I the Board with communication of 9 March 1992 issued a summons to oral proceedings to be held on 6 May 1992.

In its communication the Board expressed the provisional opinion that the subject-matter of Claim 1 on file was novel but did not involve an inventive step, having regard to document D5 or D6 and the technical knowledge of the skilled person.

VI. With fax of 5 May 1992 the Appellant informed the EPO and the Respondents that he was unable to be present at the oral proceedings to be held on the following day because of a strike in the public sector, in particular the air traffic, in Germany. He requested that the oral proceedings be postponed.

VII. The oral proceedings to be held on 6 May 1992 were cancelled, although the Representatives of the Respondents I and II appeared on 6 May 1992 before the Board. With communication of 11 May 1992 the Board summoned the parties to new oral proceedings to be held on 25 June 1992.

VIII. With letter of 12 May 1992 the Respondent I requested that the costs unnecessarily incurred by him because of the late cancellation of the oral proceedings to be held on 6 May 1992 be refunded by the Appellant.

The same request was filed by the Respondent II with letter of 9 June 1992.

IX. At the oral proceedings on 25 June 1992, the Appellant requested that the decision under appeal be set aside and that the patent be maintained on the basis of the following documents:

- Claim 1 as filed with letter of 8 January 1991,
- Claims 2 to 6 as granted,
- description pages 1 to 6 of the granted patent,
- unique figure of the granted patent.

The Respondents I and II requested that the appeal be dismissed.

Both of them, moreover, abandoned the request concerning the apportionment of costs.

X. Independent Claim 1 according to the Appellant's request reads as follows:

"A method for removing dissolved oxygen from a supply of aqueous medium comprising discharging a stream of aqueous medium from said supply thereof; injecting hydrogen gas into said discharged stream of aqueous medium and intimately mixing said hydrogen with said aqueous medium; characterized by pressurizing the intimately mixed stream to a pressure of between 80-150 psig (551 to 1034 kPa), and contacting said pressurized, intimately mixed stream, at a temperature of 15-40 degrees C, with an effective amount of palladium or platinum, dispersed on a stable carrier material, for a contact time between 0.5 and 2 minutes whereby to react the hydrogen and dissolved oxygen to produce a deoxygenated aqueous medium stream having a dissolved oxygen content of below about 10 parts per billion by weight.".

Claims 2 to 6 are dependent on Claim 1.

XI. In support of his requests the Appellant essentially argued as follows:

While it is true that the cited prior art documents deal with the removal of dissolved oxygen from an aqueous medium, the Respondents have failed to take into account the fact that in the present invention there are two clearly defined steps, namely intimately mixing hydrogen gas and the aqueous medium and then pressurising the intimately mixed stream. This means that the mixing intentionally takes place at a pressure which is lower than the pressure which exists during contact with the catalyst. A high degree of hydrogen dissolution is thereby obtained, this being a requirement for the very high degree of oxygen removal enabled by the invention. The prior art documents fail to show or suggest a distinction between the pressure which prevails at the time the hydrogen is added to the aqueous medium and the pressure which prevails after mixing the hydrogen and aqueous medium.

Proper weight should, moreover, be given to the clear discrepancy between the values of residual dissolved oxygen stated in D6 and D5. Since it is an object of the present invention to achieve low residual oxygen values, an accurate evaluation of these references is important. In particular, the lower residual oxygen value of 0.0025 mg/l = 2.5 ppb reported in D6 is a clear misprint and should be 0.025 mg/l = 25 ppb, as disclosed in D5.

The claimed method is a "once-through" process. None of the cited documents discloses such a process consisting of the combination of the steps of intimately mixing the hydrogen with the aqueous medium, pressurising the mixture to between 551 and 1034 kPa and "once-through" contacting the mixture with palladium or platinum at a temperature range of 15 to 40 °C and for a contact time between 0.5 and 2 minutes. The surprising result of this combination is that in a single passage through the catalyst, the contact time of merely 0.5 to 2 minutes proves to be quite sufficient to achieve the desired reduction in oxygen content.

XII. The Respondents contested the Appellant's view and submitted essentially the following counter-arguments:

Catalytic reduction of oxygen dissolved in an aqueous medium by means of hydrogen introduced into the aqueous medium is a well-known method in the prior art (see D1 and D3 to D8). The mixing of gaseous hydrogen with water is described by Henry's law, according to which a given pressure is necessary. Moreover, since the catalytic reaction takes place under volume reduction and exothermal conditions, it will be promoted by an increase of the pressure and a reduction of the temperature. It is thus obvious that the intimately mixed stream must be pressurised and have a suitable temperature, whereby the provision of a separate pressurising step does not bring any advantage. The claimed ranges for pressure, temperature and contact time are normal for the skilled person, having regard, for example, to D6 which may be considered as the closest prior art document. The result achieved, i.e. a residual oxygen amount below 10 ppb by weight, is, moreover, not surprising, because it was already achieved in the prior art (see, for instance, D1 and D3).

During the oral proceedings, the Representative of the Respondent I admitted that, concerning the discrepancy between D5 and D6, the value of 25 ppb mentioned in D5, which is a publication of the Respondent I, is, indeed, correct, the discrepancy being due to a typing error in

D6. However, the method of D5 would lead to lower residual oxygen amounts, if the oxygen measurements were carried out by means of the modern device "Orbisphere 2713" used in the contested patent (see page 3, lines 49 to 52) instead of the methods used in D5 (see page 4, end of left-hand column). In his opinion, the method of Claim 1 of the contested patent essentially corresponds to that of D5 (or D6) with the difference that the residual oxygen amount is more precisely determined and a separate pressurising step is envisaged, which, however, is not advantageous.

Reasons for the Decision

1. The appeal is admissible.

2. Allowability of the amendments

2.1 Independent Claim 1 is based on the originally filed Claim 1 with the amendments that the original pressure interval of 60 to 150 psig has been replaced by the narrower interval of 80 to 150 psig and the contact time interval of 0.5 to 2 minutes has been added.

The new pressure interval is disclosed on page 4, lines 8 to 22 of the original description, when read in conjunction with page 7, lines 5 to 8. The limitation of the claim is not based on an arbitrary restriction of the originally claimed pressure interval, but merely represents a reduction of said interval by using a value already envisaged within the original document. Moreover, it appears that the skilled person can readily recognise the value of 80 psig described in the specific Example I as not so closely associated with the other features of the example as to determine the effect of that embodiment of the invention as a whole.

The contact time interval is disclosed on page 5, lines 12 to 15 of the original description.

Dependent Claims 2 to 6 correspond to the originally filed Claims 2 to 6.

Furthermore, the originally filed documents have been essentially amended so as to meet the requirements of Rule 35(12) EPC.

2.2 Therefore, the amended version of the patent, on the basis of which the Appellant requests that the patent be maintained, does not contravene the requirements of Article 123(2) and (3) EPC, since it neither includes subject-matter extending beyond the content of the application as originally filed, nor do the amendments to the claims have the effect of extending the scope of protection conferred.

3. Novelty

3.1 Claim 1 refers to a method for removing dissolved oxygen from a supply of aqueous medium, the method comprising the steps of discharging a stream of aqueous medium from said supply thereof, injecting hydrogen gas into said discharged stream of aqueous medium, intimately mixing said hydrogen with said aqueous medium and contacting the intimately mixed stream with an amount of palladium or platinum dispersed on a stable carrier material, so as to react the hydrogen and dissolved oxygen to produce a deoxygenated aqueous medium stream.

Claim 1 does not explicitly mention whether the method is repetitive or of a "once-through" kind, and does not give any information concerning the carrier material of the catalyst.

3.2 The result of obtaining a residual dissolved oxygen content of below about 10 ppb by weight is achieved according to Claim 1 by:

(a) pressurising the intimately mixed stream to a pressure of 80 to 150 psig, before the contact with the catalyst takes place;
(b) keeping the temperature of the pressurised, intimately mixed stream at 15 to 40 °C;
(c) contacting the pressurised, intimately mixed stream with the catalyst for a time between 0.5 to 2 minutes.

3.3 The method of catalytic reducing oxygen dissolved in an aqueous medium by means of hydrogen injected into the aqueous medium is well known in the prior art. For example, documents D1 and D3 to D8 disclose a method such as mentioned in section 3.1 above.

None of these documents, however, discloses the step (a) of pressurising the mixture of aqueous medium and hydrogen before the contact with the catalyst takes places. Moreover, none of them, when taken alone, discloses values of all parameters mentioned in Claim 1, i.e. pressure and temperature of the mixture as well as contact time thereof with the catalyst, within the claimed intervals (see section 3.2 above), the result being thereby achieved, that the dissolved oxygen content is below about 10 ppb by weight.

3.4 Therefore, the subject-matter of Claim 1 is novel in the sense of Article 54 EPC.

4. Inventive step

4.1 Feedwater used in the primary and secondary coolant systems of nuclear power reactors, in heat generating stations or in industrial processes normally contains dissolved oxygen causing the destructive phenomenon of corrosion. Skilled persons have, therefore, recognised the need to remove said oxygen from the water, before it could oxidise the construction materials. Physical and chemical methods have been envisaged in the prior art; among the chemical methods, a particularly simple and energy saving process consists in the catalytic reduction of oxygen by means of hydrogen introduced into the water. Indeed, this method was already known at the priority date of the patent in suit, for example from D5 or D6, which are equivalent as referring to an evaluation of the same experiments and can been considered as the closest prior art.

Therefore, starting from the method according to D5 or D6, the objective problem underlying the patent in suit can be seen in the determination of optimal operating conditions so as to achieve a higher degree of oxygen removal, i.e. a residual oxygen content below 10 ppb by weight, whereby it is assumed that the wrong value of 2.5 ppb cited on page 60 of D6 is replaced by 25 ppb, as known from D5, in conformity with the admission of the Respondent I at the oral proceedings (see section XII above).

Considering that the majority of dissolved oxygen will normally have to be removed, to what extent depending on the particular materials and operating conditions, it appears that no inventive step can be perceived in the fact alone of stating this problem.

4.2 Said problem is solved by the steps above mentioned in section 3.2.

4.2.1 According to the method of the patent in suit, hydrogen gas has first to be injected in an aqueous medium. The phenomenon of dissolving a gas in a liquid is described by Henry's law, according to which the solubility of the gas is proportional to its pressure at constant temperature. Depending on the amount of hydrogen to be dissolved, which normally corresponds to the stoichiometric amount to react with the dissolved oxygen or a slight excess over that amount, a suitable value of the hydrogen pressure will be chosen. Once a dynamic equilibrium between the gaseous and the liquid phase is established, the saturation amount of the dissolved hydrogen will depend on the temperature and pressure conditions; indeed, in order to avoid that the dissolved hydrogen gas leaves the liquid, thereby not reacting with the oxygen, the pressure should, according to Henry's law, be kept as high, the temperature as low, as possible.

The above in section 3.2 mentioned step (a) of Claim 1 of pressurising the intimately mixed stream, before the contact with the catalyst takes place, is, therefore, regarded as an obvious measure for avoiding a decrease of the amount of dissolved hydrogen, which is needed in the subsequent catalytic process.

In this connection, the Appellant argued that the step of pressurising the mixture of aqueous medium and hydrogen before the contact with the catalyst takes places is very important for the invention; however, he failed to give any evidence that such a measure produces any surprising effect or advantage with the regard to the case known from D5 or D6, in which the entire system is held at a given pressure.

A further reason for pressurising said intimately mixed stream derives from the fact, that the catalytic reduction of oxygen in feedwater takes place under volume reduction. According to the principle of Le Chatelier, the increase in pressure resulting from said pressurising step will then positively influence said reaction in the direction of the product with smaller volume. The skilled person, therefore, expects that a lower residual amount of oxygen can be achieved, if a suitable pressure is applied before the catalytic reaction takes place.

As regards the claimed pressure interval, it is clear that it should not be lower than the pressure needed to inject the desired amount of hydrogen into the water.

A hint to values of the same order of magnitude as those claimed is, furthermore, given by documents D5 (see Table 4 on page 5, Figure 6) and D6 (see page 61, left-hand column, end of first paragraph, Figure 6), referring to a catalytic reduction commercial unit operating at 5 bar (500 kPa).

4.2.2 As regards the operating temperature, considering that the reaction between oxygen and hydrogen is highly exothermal, the above mentioned principle of Le Chatelier already gives a hint to the fact that lower amounts of residual dissolved oxygen can be better obtained by choosing a low operating temperature, so as to positively influence the exothermal reaction.

A temperature value contained in the claimed temperature interval is, moreover, disclosed in said documents D5 and

D6 (see the passages above referred to), according to which the commercial unit of Figure 6 works at a pressure of 5 bar and a temperature of 25 °C.

4.2.3 With reference to the third parameter, i.e. the contact time, the claimed range appears to be normal for the process in suit, as it can be inferred from documents D5 (see Figures 5 and 7) and D6 (see Figures 5 and 7, the specific flow rate having to be converted in the residence time, as in D5).

It is clear from the cited graphs, that the residual oxygen amount tends to an asymptotic value depending on the used catalyst. Since analogous graphs are expected to be valid in the present case, the skilled person will have no difficulties in choosing a range for the contact time, of which the lower value corresponds to the beginning of the asymptotic part of the curve and the upper value depends on economic considerations.

4.2.4 At the priority date of the patent in suit, methods based on the catalytic reduction of oxygen in water and giving residual oxygen amounts below 10 ppb by weight were not unusual. Depending on the operating conditions, the catalyst and the apparatus, following results can, for instance, be mentioned:

- below 7.15 ppb, this value representing the lowest detectable limit at the priority date of D1 (see D1, column 3, lines 36 to 42 and the Appellant's letter of 31 March 1990, page 1 of observations);
- oxygen-free (see D3, page 6, lines 22, 23);
- below 10 ppb, mainly about 5 ppb (see D4, page 57, "Kondensataufbereitung" and page 58, first paragraph of left-hand column);
- below 10 ppb (see D7, Examples 1, 3 and 5).

It should be noted that, at the priority dates of D3, D4 and D7, methods already existed to detect such low amounts of dissolved oxygen in industrial water, as it is proved by D10 (see "Non-Referee Method D" on page 319).

It can so be concluded, that it is not at all surprising that with the method according to present Claim 1 a residual amount of oxygen below 10 ppb by weight can be achieved. This result should rather be regarded as a foreseeable consequence of the claimed operating conditions, which, however, do not go beyond the limits of the technical knowledge of the skilled person.

4.3 For the above reasons, the subject-matter of Claim 1 does not involve an inventive step in the sense of Article 56 EPC and Claim 1 is, therefore, not allowable under Article 52(1) EPC.

5. Claims 2 to 6 are subordinate to Claim 1 and, consequently, are also not allowable.

Thus, the grounds for opposition mentioned in Article 100(a) EPC prejudice the maintenance of the European patent.

Order

For these reasons, it is decided that:

The appeal is dismissed.

The Registrar: The Chairman:

P. Martorana E. Turrini