Datasheet for the decision
of 21 May 2007

Case Number: T 0332/03 - 3.3.05
Application Number: 97903169.7
Publication Number: 0881989
IPC: C02F 1/46
Language of the proceedings: EN

Title of invention:
Electrochemical treatment of water contaminated with nitrogenous compounds

Applicant:
Enpar Technologies Inc.

Opponent:
-

Headword:
Groundwater electrochemical decontamination / ENPAR

Relevant legal provisions:
EPC Art. 56

Keyword:
"Inventive step (yes)"

Decisions cited:
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Catchword:
-
Case Number: T 0332/03 - 3.3.05

DECISION
of the Technical Board of Appeal 3.3.05
of 21 May 2007

Appellant: Enpar Technologies Inc.
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Decision under appeal: Decision of the Examining Division of the European Patent Office posted 13 November 2002 refusing European application No. 97903169.7 pursuant to Article 97(1) EPC.

Composition of the Board:
Chairman: M. Eberhard
Members: J.-M. Schwaller
H. Preglau
Summary of Facts and Submissions

I. The appeal was lodged against the decision of the examining division to reject European patent application No. 97903169.7. The decision was based on claims 1-15 submitted with letter of 8 December 1999.

II. The following prior art documents were inter alia relied upon during the examination proceedings:

D1 = WO 92/13117
D5 = WO 95/29129
D6 = CA-A-2098511

III. In the decision, the examining division held that the subject-matter of claim 1 then on file did not involve an inventive step. The arguments can be summarised as follows:

The subject-matter of claim 1 differs from the processes known from D1 or D2 in that it is directed towards in-situ groundwater treatment, with the contaminated water remaining in its native environment during treatment by providing the electrodes in the ground.

The problem to be solved by the present invention may be regarded as the modification of the electrolytic process for nitrogenous contaminant removal known from either D1 or D2 in order to allow for in-situ groundwater treatment, with the contaminated water remaining in its native environment during treatment.
It would be obvious for the skilled person to include the features relating to in-situ groundwater treatment known from either D5 or D6, especially in view of the known advantages associated with these features, in the known process of either D1 or D2 and thus to arrive at the solution proposed in claim 1.

IV. With the grounds of appeal dated 10 March 2003, the appellant maintained as unique request the set of claims 1-15 on which the decision was based.

V. In a communication, the board objected to claim 1 of said request under inter alia the following aspects:

The requirements of support by the description were not fulfilled because owing to the arguments provided in the grounds of appeal, it appeared that the present invention could not be carried out successfully for any type of nitrogenous contaminant, in particular with water-insoluble organic ones, but only for a limited number of contaminants, namely the nitrate or ammonium species.

In view of the argumentation in the grounds of appeal, certain technical features appeared to be essential for solving the problem addressed in the present application; the features in question were:

(a) the oxygen-nitrogen-hydrogen (O-N-H) pH-Eh diagram (the appellant described this feature as a reliable guide for determining the limits Eh-voltage that will favour gaseous nitrogen),

(b) the direct transformation of ammonium or nitrate to nitrogen gas, and
(c) the contaminant had to be in solution in the water.

Claim 1 lacking these features, the arguments for inventiveness based thereon could not be taken into consideration and as a consequence, claim 1 was considered as lacking an inventive step on the one hand over D1 in combination with D5 (or D6) and, on the other hand, over D2 in combination with either D5 or D6.

VI. In a letter dated 12 July 2006, the appellant filed observations in response to the board's communication along with a set of amended claims 1-14 replacing those on which the decision was based.

VII. Following another communication of the board dated 26 April 2007, wherein an inconsistency of wording within claim 1 was objected to, the appellant submitted with a letter dated 4 May 2007 a replacement page containing an amended claim 1 as well as unamended claim 2.

The new claim 1 reads as follows:

"Procedure for in-situ treatment of groundwater contaminated by dissolved nitrate or ammonium species, wherein:
[2] the water is groundwater in its native aquifer in the ground, and the treatment procedure is carried out in-situ, the contaminated water remaining in the aquifer during the procedure;
[3] the procedure includes the following steps:
[4] measuring the pH of the nitrogen-contaminated groundwater;"
[5] determining, by use of the Nernst equation, for that pH as measured, the voltages V-Ngas-upper and V-Ngas-lower, being the theoretical limiting Eh voltages between which the predominant nitrogen phase is nitrogen gas;
[6] providing a pair of electrodes in the nitrogen-contaminated-water, in the ground;
[7] so arranging the electrodes in the ground, that the electrodes are in electrolytic communication with each other through the nitrogen-contaminated-water, in the ground;
[8] thereby creating an electrochemical cell in the water, in the ground, being either an electrolytic or a galvanic cell, the nitrogen-contaminated-water comprising the electrolyte of the cell;
[9] measuring the Eh voltage V-Eh of the nitrogen-contaminated-groundwater;
[10] controlling the voltage V-cell, being the voltage as measured between the in-ground electrodes of the in-ground cell, to such a value as to maintain an Eh voltage in the water in the vicinity of one of the electrodes at a voltage V-Ngas, being an Eh voltage between V-Ngas-upper and V-Ngas-lower;
[11] whereby the dissolved nitrate or ammonium species in the groundwater is directly transformed, in-situ, in the ground, into nitrogen gas;
[12] the Eh voltage of a solution being the redox voltage generated in the solution by comparison with a standard hydrogen electrode."
VIII. The appellant presented *inter alia* the following arguments:

Feature [5] is a key feature that aids in distinguishing claim 1 from the old prior art procedures. Claim 1 does however not recite that the actual pH-Eh diagram for N-O-H is the thing that must be used in making the determination defined in feature [5], because there are other ways of presenting the thermodynamic constructs than the pH-Eh diagram itself and which can be used as a look-up reference tool. The pH-Eh diagram itself is just one way among others of representing the Nernst theoretical construct.

The limits V-Ngas-upper and V-Ngas-lower, as recited in claim 1, are theoretically calculated values, derived from the Nernst equation. Usually the Nernst-calculated values for Eh-voltage and pH are not an accurate prediction of the real levels of Eh-voltage and pH, as actually measured. But in the case of in-situ groundwater contaminated with nitrate or ammonium, the Nernst calculations for N-O-H phases do, surprisingly, give an accurate-enough prediction as to what the Eh-voltage should be, that a commercially-practicable treatment system can be based on the prediction. The inventive step lies in the purposeful coordination of the combination of procedural steps recited in claim 1 which actually result in the ideal that only N₂ gas - and none of the toxic phases of N-O-H - is the final form of nitrate or ammonium species, installation after installation, year after year.
IX. The appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of claims 1-2 submitted with letter dated 4 May 2007 and claims 3-14 submitted on 12 July 2006.

Reasons for the Decision

1. Allowability under Article 123(2) EPC

Claims 1-14 of the present request have a basis as follows in the international application published as WO 97/30941:
- claim 1: claims 1, 2, 14, 20; page 1, lines 4-7; page 3, lines 1-9; page 16, lines 10-12 of the PCT application
- claims 2-5: respectively claims 3, 5, 6, 7 of the PCT application
- claim 6: claim 8; page 4, line 15 of the PCT application
- claim 7: claim 9 of the PCT application
- claim 8: page 15, lines 4-6 of the PCT application
- claim 9: claim 15 of the PCT application
- claim 10: claims 18 and 19 of the PCT application
- claim 11-14: respectively claims 21, 23, 24 and 4 of the PCT application.

Accordingly, the requirements of Article 123(2) EPC are complied with.

2. Essential feature

In its communication, the board submitted that the oxygen-nitrogen-hydrogen (O-N-H) Eh-pH diagram would
appear to be a feature essential to the performance of the invention. As argued by the appellant, the Eh-pH phase diagram as shown in Figure 1 is derived by calculation, by use of the Nernst equation, and is therefore to be regarded as simply a manner of graphing, or presenting in visual form, the Nernst equation (see also page 3, line 26 to page 4, lines 9 of the application). As further pointed out by the appellant, there are other ways of representing the Nernst theoretical construct, such as look-up tables which are equivalent to the diagram. Sets of the Nernst relationships appropriate to the nitrogenous compounds which are available in table form are mentioned in the paragraph bridging pages 3 and 4 of the description. Under this aspect, the recitation in claim 1 of the Nernst equation as an essential feature for determining the theoretical limiting Eh voltages between which the predominant nitrogen phase is nitrogen gas - instead of the O-N-H phase diagram - is acceptable.

3. The objection under Article 84 EPC (lack of consistency within claim 1) raised in the board's communication of 26 April 2007 has been overcome by the substitution of the feature "nitrate or ammonium species" for the expression "nitrogenous contaminant" in item [11].

4. Novelty was not contested in the impugned decision. As the board also considers that the subject-matter claimed is novel over the known prior art, no further comments on this matter are needed.
5. **Inventive step**

5.1 **Closest prior art**

5.1.1 In both the contested decision and the first board's communication, the closest prior art to claim 1 then on file was considered as being represented by either D1 or D2.

5.1.2 Claim 1 of the present request now includes the features that the groundwater is contaminated by dissolved nitrate or ammonium species and that these species are electrochemically and directly transformed into nitrogen gas. Owing to the recitation of these features, D1 no longer represents the closest prior art for the following reasons.

5.1.3 D1 (page 1, line 10 to page 2, line 11) discloses a process wherein a composition containing energetic compounds such as nitratoesters, nitramines and/or other nitro-group-bearing compounds, combustible fuels, oxidants, and combinations thereof, is desensitized in a controlled manner by electrolysis, said composition being placed in contact with two electrodes and an electric current being passed from one to the other through the composition. One or more of the active compounds undergoes an electrolytic conversion in an oxidation or reduction reaction (or both) at the appropriate electrode, thereby lowering the susceptibility of the composition to initiation. The process is applicable to solid energetic compounds and most conveniently performed by macerating the solid material and combining it with a liquid to form a slurry prior to placing it in contact with the
electrodes. The preferred liquid for the slurry is water (see page 4, lines 10-21). As indicated on page 3, lines 16-37 of D1, the process is applicable to a wide range of compositions including various formulations of propellants and explosives. Examples of specific components, including oxidizers, are among others, ammonium nitrate, ammonium perchlorate, ammonium picrate, diazodinitrophenol, diethylnitramine dinitrate, lead azide, mannitol hexanitrate, nitrocellulose, nitroglycerine or pentaerythritol tetranitrate.

5.1.4 D1 (page 1, lines 29-33; claims 2, 3, 13 and 16) discloses that nitrogen gas can be generated when nitro-group-bearing compounds are desensitized, in which case at least a portion of the nitro groups in the nitro-group-bearing compounds is converted to either amino groups or to alcohol groups and nitrogen gas. However, the direct production of nitrogen gas from either nitrate or ammonium species is not disclosed in D1.

Under these circumstances, since the purpose of D1 is not the deliberate production of nitrogen gas from a water contaminated by dissolved nitrate or ammonium species, this document no longer represents the closest prior art to the subject-matter of present claim 1. Therefore D2 is considered to represent the closest prior art.

5.2 It remains thus to establish whether the subject-matter claimed meets the requirements of Article 56 EPC, starting from D2, the latter disclosing - as can be seen below - a process for treating water in which a
nitrate contaminant is electrochemically reduced into nitrogen gas.

5.2.1 D2 discloses (claim 1; page 2, lines 15-20) a process for electrochemically reducing nitrates to nitrogen gas in aqueous solutions. This process comprises providing a nitrate-containing aqueous solution into the cathodic compartment of an electrolytic cell in which a cation-exchange membrane separates the cathodic and anodic compartments (the anodic compartment containing an aqueous solution of sulfuric acid), the cell containing a pair of electrodes, and applying a voltage to the cell, so that the electrodes are in electrolytic communication with each other through, inter alia, the nitrate-contaminated water and thereby creating an electrolytic cell wherein the nitrate-contaminated water comprises the electrolyte of the cell.

In the example at page 2, lines 46-55 of D2, 5% sulfuric acid is fed to the anodic compartment and the pH is maintained constant in the cathodic compartment by adding HCl. As recognized by the appellant, for doing this, the pH must necessarily be measured. At a voltage of from 1000 to 1400 mV, about 90% of the nitrate can be converted into nitrogen gas.

5.2.2 Accordingly, the subject-matter of present claim 1 is distinguished from D2 by the following features:
[2] the water is groundwater in its native aquifer in the ground, and the treatment procedure is carried out in-situ, the contaminated water remaining in the aquifer during the procedure;
[5] determining, by use of the Nernst equation, for the pH as measured in step [4], the voltages V-Ngas-upper
and \( V_{\text{Ngas-lower}} \), being the theoretical limiting Eh voltages between which the predominant nitrogen phase is nitrogen gas;
[6] providing a pair of electrodes in the nitrogen-contaminated water, in the ground;
[9] measuring the Eh voltage \( V_{\text{Eh}} \) of the nitrogen-contaminated-groundwater;
[10] controlling the voltage \( V_{\text{cell}} \), being the voltage as measured between the in-ground electrodes of the in-ground cell, to such a value as to maintain an Eh voltage in the water in the vicinity of one of the electrodes at a voltage \( V_{\text{Ngas}} \), being an Eh voltage between \( V_{\text{Ngas-upper}} \) and \( V_{\text{Ngas-lower}} \).

5.2.3 Starting from D2, the technical problem to be solved by the subject-matter of claim 1 may be seen in the provision of a low cost process for treating groundwater contaminated with dissolved nitrate or ammonium species, avoiding the production of harmful gaseous products.

5.2.4 In view of the information in the description, it is credible in the absence of evidence to the contrary that the above problem has been effectively solved by the process as defined in claim 1.

5.2.5 The solution as proposed in claim 1 of the present request involves an inventive step for the following reasons.

5.2.6 As pointed out in item 5.1.4 supra, D1 does not disclose a process for the treatment of water contaminated with dissolved nitrate or ammonium species in which these species are directly transformed into
nitrogen gas. D1 describes that certain nitrate- or ammonium-containing compounds can be desensitized electrochemically, and that in the case of nitro-group-bearing compounds, a portion of the nitro groups may be converted to alcohol and nitrogen gas. Neither the ammonium nor the nitrate species however belong to this family of compounds described in D1 as being desensitized into alcohol and nitrogen gas.

In this context and since D1 does also not disclose the distinguishing features [2], [5], [6], [9], [10] identified under item 5.2.2 supra, any lack of inventive step argumentation based on D1 in order to arrive at the subject-matter of present claim 1 would be based on hindsight.

5.2.7 The skilled person would also not find the solution to the problem identified above in either of the documents D5 or D6, because although the provision of an electrochemical cell for the treatment of contaminated groundwater in its native aquifer is disclosed in both D5 (claims 1, 5 and 7) and D6 (claim 1), the contaminants to be treated in these documents are of a different type. D5 (page 1, lines 3-4; claim 1) relates to the treatment of water, especially groundwater, contaminated with organic contaminants, such as halogenated hydrocarbons. According to the passage bridging pages 12 and 13, the process of D5 may also be used to treat aliphatics, aromatics and polyaromatics with halogen and nitrogen group substituents. Examples include solvents such as carbon tetrachloride, tetrachloroethene, and hexachloroethane; hexachlorobenzene, nitrosamines, explosives such as trinitrotoluene, PCP's, nitro-PAH's, and certain
pesticides. D6 is concerned with inhibiting the formation of, and alleviating, acidity in water residing in an aquifer.

Since neither D5 nor D6 furthermore disclose either of the distinguishing features [5], [9], or [10] identified under item 5.2.2 supra, let alone these features in combination, they cannot suggest using these features in a process for electrochemically reducing nitrate or ammonium species dissolved in aqueous solutions or in groundwater to nitrogen.

5.2.8 The other documents cited in the European search report also do not describe or suggest the combination of features [5], [9] and [10] for an in situ treatment of groundwater contaminated by dissolved nitrate or ammonium species and their direct transformation into nitrogen gas.

5.2.9 There is also no hint that the distinguishing features [5], [9] and [10] would belong in combination to the common general knowledge of a skilled person faced with the problem of treating groundwater contaminated by dissolved nitrate or ammonium species, so that the subject-matter of present claim 1 can also not be considered as being obvious in view of D2 taken in combination with the common general knowledge.

5.2.10 Accordingly, for the reasons developed above, the subject-matter of claim 1 cannot be considered obvious to a person skilled in the art in view of the cited prior art. The claims 2-14 being dependent on claim 1, these claims therefore also meet the requirements of Article 56 EPC.
Order

For these reasons it is decided that:

The decision under appeal is set aside.

The case is remitted to the department of first instance with the order to grant a patent on claims 1 and 2 submitted with letter of 4 May 2007 and claims 3-14 filed on 12 July 2006, with a description and figures to be adapted.

The Registrar: The Chairman:

C. Vodz M. Eberhard