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D E C I S I O N
of 29 March 2000

Case Number: T 0776/98 - 3.4.2

Application Number: 90106051.7

Publication Number: 0390158

IPC: C25B 3/04, C25B 11/06

Language of the proceedings: EN

Title of invention:
Electrolysis cell and method of use

Applicant:
UNITED TECHNOLOGIES CORPORATION

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 56, 83

Keyword:
"Inventive step (yes - after amendments)"
"Disclosure - sufficiency (yes)"

Decisions cited:
-

Catchword:
-

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Case Number: T 0776/98 - 3.4.2

D E C I S I O N
of the Technical Board of Appeal 3.4.2
of 29 March 2000

Appellant: UNITED TECHNOLOGIES CORPORATION
United Technologies Building
1 Financial Plaza
Hartford, CT 06101 (US)

Representative: Klunker . Schmitt-Nilson . Hirsch
Winzererstrasse 106
D-80797 München (DE)

Decision under appeal: **Decision of the Examining Division of the European Patent Office posted 9 March 1998 refusing European patent application No. 90 106 051.7 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: E. Turrini
Members: R. Zottmann
V. Di Cerbo

Summary of Facts and Submissions

I. The appellant (applicant) lodged an appeal against the decision of the Examining Division to refuse European patent application No. 90 106 051.7 with the publication No. 0 390 158.

The reason given for the refusal was that the subject-matters of the independent claims did not involve an inventive step.

II. The following documents are cited in this decision:

D1: Catalysis Letters, vol. 1, 1988, J. C. Baltzer AG, Basel, Switzerland, pages 73 to 79;

D2: US-A-4 595 465;

D3: J. Electrochem. Soc., vol. 131, No. 7, 1984, pages 1511 to 1514;

D4: J. Am. Chem. Soc. 1984, 106, pages 5033 to 5034;

D5: US-A-4 668 349;

D7: J. Electrochem. Soc., June 1988, pages 1470 to 1471;

D8: J. Am. Chem. Soc. 99, 1 1977, pages 286 to 288;

D9: Römpps Chemie-Lexikon, 8th edition, Stuttgart 1983, pages 1608 to 1610; and

D10: Römpps Chemie-Lexikon, 8th edition, Stuttgart 1985,

page 3200, catchword "Phthalocyanin-Farbstoffe".

III. In communications pursuant to Article 110(2) EPC, in a conversation by telephone and during oral proceedings the Board of Appeal expressed its preliminary opinion that and why the application did not meet the provisions of the EPC.

To meet these objections, the Appellant reformulated the claims.

IV. At the end of the oral proceedings, which took place on 29 March 2000, the appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of the following application documents:

Claims: 1 to 3 filed with letter of 27 March 2000;

Description: to be adapted; and

Drawings: as originally filed.

V. The independent claim reads as follows:

"1. An electrolysis cell (2) being operable to reduce carbon dioxide to a product consisting essentially of methanol and/or formic acid, comprising an anode (4), a cathode (8), and, at the cathode side of said electrolysis cell (2), a material having catalytic effect containing at least one metal phthalocyanine, characterized in
that a solid polymer electrolyte (12) capable of

transporting positive ions is provided;
and that said material having catalytic effect constitutes simultaneously the cathode (8), said cathode being formed of

(a) at least one metal phthalocyanine, or

(b) a mixture of at least one metal phthalocyanine and at least one other catalytic or non-catalytic material."

The remaining claims 2 and 3 are dependent on claim 1.

VI. The arguments supporting the appellant's request are summarized as follows:

Though D8 discloses that metal phthalocyanines (hereinafter called MePcs) show low electrical conductivity, they have, however, ionic conductivity in electrolytic environment which property has not been examined in D8. Moreover, Pcs are semiconductors and thus have charge transport ability, as can be seen from D9. A cell with a cathode in the form of a layer consisting of a mixture of 20% Teflon with MePc powder did work in a satisfactory manner. Since Teflon cannot contribute to the electrical conductivity of MePc, the latter must be sufficiently conductive. The electrolysis cell described in the laboratory report dated 15 June 1987, as filed during the examining proceedings, shows the inventor's originally test cell and actually contains "stainless steel screens plated with indium to form the cathode active area". However, said cell was built almost two years earlier than the priority date of the application-in-suit and is not a

cell following the teaching of present claim 1.

D3 is the closest prior art with respect to claim 1 since it is the only document disclosing the use of Pc and the production of substantial amounts of HCOOH and CH_3OH. Though Pcs are used there as catalysts, it is stressed that deposition of these catalysts on carbon (hereinafter called C) electrodes with a very smooth surface is important for their catalytic abilities. It does not contain any suggestion that Pcs or a **mixture** of said catalyst with a further component including C deposited on a solid polymer electrolyte (hereinafter called SPE) membrane will produce HCOOH or CH_3OH. The remaining documents also do not guide the skilled person in that direction.

Reasons for the Decision

1. *Amendments*

In the Board's opinion, there are no objections under Article 123(2) EPC against the claims since they do not contain subject-matter which extends beyond the content of the application as originally filed. In particular, as to features (a) and (b) reference is made to page 6 last paragraph.

2. *Sufficiency (requirements of Article 83 EPC)*

According to alternative (a) of claim 1, the cathode is formed exclusively of at least one MePc. In its communications, the Board called in question the

suitability of such cathodes for electolysis cells, since, according to D8, their (specific) electrical conductivity seems to be very low. Document D8 provides conductivity parameters of MePcs and states that MePcs partly oxidized with iodine show anisotropic metallic conductivity. According to Table I, NiPc, CoPc and FePc have (specific) electric conductivity of 1 x 10^{-11}, 2 x 10^{-10} and 2 x 10^{-10} $Ù^{-1}(cm^{-1})$ and it is mentioned there that these compounds are organic semiconductors.

However, the conductivities of the MePcs are measured at room temperature. Since an electrolysis cell with a solid electrolyte can easily - and usually is operated - at higher temperatures and since conductivity is rising in substance exponentially, since the cross-section of the cathode is relative high and since its thickness is very low (see the drawing of the application-in-suit), conductivity of the cathode layer seems to be or can easily be made high enough in electrolytic environment for the purpose. In addition, D10 (mentioned during the oral proceedings) stresses that MePcs are used for fuel cells due to their semiconductor properties.

According to D4, formation of $Co(Pc)^-$ is important for the reduction of CO_2 on carbon electrodes modified with adsorption of CoPc, suggesting that, in addition to semiconductor conductivity, ionic conductivity of MePcs in an electrolytic environment could occur.

Taking moreover into account the appellant's submission that a cell with a cathode in the form of a layer consisting of a mixture of 20% Teflon with MePc powder worked in a satisfactory manner, whereby it seems that

Teflon does not contribute to conductivity, the Board sees no reason to doubt that the skilled person, on the basis of the teachings of the application as originally filed, is able to construct a cell working in a satisfying manner.

3. *Novelty*

3.1 D1 describes an electrolysis cell and method for reduction of CO_2 to hydrocarbon products including CH_3OH at a Cu cathode in contact with a SPE consisting of Nafion. The CO_2 is fed to the cathode in the gas phase while the counter electrode reactant is a solution of H_2SO_4. It is mentioned that Cu alone is completely inactive for hydrogenation whereas Cu alloy catalysts have shown activity for the hydrogenation. D1 is silent regarding MePcs.

3.2 D2 discloses a rather complicated device for the reduction of CO_2 to oxalates. It comprises two photosystems and three chambers separated by two membranes consisting of Nafion with photosensitizers deposited thereon. Among a lot of other catalysts MePcs may be used as such photosensitizers. The electrodes are separated from said membranes and are immersed in fluidic electrolytes. Not any material for said electrodes is mentioned in D2.

3.3 According to D3, MePcs deposited on C electrodes are found to catalyze the electroreduction of CO_2 to HCOOH in aqueous acid solutions saturated with CO_2 by electrolysis. At pH above 5 HCOOH is formed; CH_3OH is also produced at lower pH values. A glassy C rod is polished and cleaned prior to depositing the catalyst,

namely MePcs. A thin layer of ca. 10 µg of MePc is deposited on the C surface. Only CoPc and NiPc are used. It is emphasized that graphite and glassy C seem to be specific in their ability to utilize Pcs as catalysts for CO_2 reduction.

3.4 D4 discloses the electrocatalytic reduction of aqueous solutions of CO_2 to CO using CoPc as catalyst. The CoPc is deposited on pyrolytic graphite or C by adsorption in a monolayer coverage (see also D5 column 1 lines 30 to 38).

3.5 D5 discloses the electrocatalytic reduction of aqueous solutions of CO_2 to CO using transition metal complexes with square planar geometry, e. g. MePcs. Preferably CoPc is adsorbed on a glassy C electrode, polished with alumina and sonicated.

3.6 D7 describes electrochemical reduction of CO_2 to hydrocarbons with one or two C atoms at Cu electrodes supported on SPE membrane, preferably Nafion. It is said that Cu is electrocatalytically active for promoting high rate CO_2 reduction in CO_2 saturated aqueous solutions. Said document is silent with respect to Pcs.

3.7 Documents D8, D9 and D10 deal with the properties of MePcs and, respectively, semiconductors but use of said MePcs in electrolysis cells or a similar use is not mentioned there.

3.8 The other prior art documents on file are farer away from the electrolysis cell as defined by claim 1 than the above-described documents.

3.9 Thus the subject-matter of claim 1 is considered as being novel in the meaning of Article 54 EPC. Novelty of the independent claims confers novelty also on the dependent claims.

4. *Inventive Step*

4.1 In view of the fact that D3 is the only prior art document that discloses the use of MePc as catalyst at the cathode and the production of CH_3OH and HCOOH, the opinion of the appellant can be accepted that none of the cited prior art documents comes nearer to the subject-matter of claim 1 than D3.

The main difference between the cell according to claim 1 and that of D3 consists in that a SPE capable of transporting positive ions is provided and that the cathode is formed of (a) at least one metal phthalocyanine, or (b) a mixture of at least one metal phthalocyanine and at least one other catalytic or non-catalytic material.

With these measures the cathode can be manufactured in an easier manner and nevertheless the efficiency for the conversion of CO_2 to CH_3OH and HCOOH is very high (see e.g. EP-A-0 390 158 column 1 lines 38 to 40 and column 3 last paragraph).

The problem underlying the solution according to claim 1 is, therefore, to further develop the electrolysis cell according to D3 such that the above effects are obtained.

4.2 Though use of a SPE instead of a liquid electrolyte may

be suggested by the teaching of one of the documents D1 or D7, there is, however, no hint at the structure of the cathode according to features (a) or (b) in one of the cited documents. When MePcs are used as cathode material, they are deposited as a thin layer on a smooth C surface, see sections 3.3 to 3.5 above.

There is not any pointer that MePcs in the absence of any additional electrode material which has good electrical conductivity is suitable as cathode material.

By a mixture of materials the skilled person understands a homogeneous blend of components which, therefore, differs fundamentally from a material having been obtained by depositing a first material on a second smooth material.

Since none of the documents on file discloses or suggests a cathode made of a material as defined in features (a) or (b), and, beyond that, not all of the remaining features, the skilled person would not arrive at an electrolysis cell with all essential features of claim 1 without inventive skill if starting from another document than D3 as nearest prior art.

4.3 Therefore, the subject-matter of claim 1 involves also an inventive step as defined in Article 56 EPC with respect to the prior art documents on file.

The dependent claims concern particular embodiments of the subject-matter of claim 1 and are, therefore, likewise inventive.

5. In the result, the Board of Appeal takes the view that the claims comply with the requirements of the EPC. This applies also to the drawing. However, the description will have to be adapted to these claims and the relevant prior art will have to be disclosed in the introductory part of the description (Rule 27(1) 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

Claims: 1 to 3 filed with letter of 27 March 2000;

Description: to be adapted; and

Drawings: as originally filed.

The Registrar: The Chairman

P. Martorana E. Turrini