Europäisches Patentamt

European Patent

Beschwerdekammern

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Chambres de recours

T171

Case Number: T 29 183

Office

Boards of Appeal



DECISION of the Technical Board of Appeal 3.4.1 of 23 July 1984

Appellant: Diamond Shamrock Technologies S.A. c/o Peterhens Treuhand AG Artherstrasse 12 CH-6301 Zug Switzerland

Crcnin, Brian Harold John, et al. Representative: c/O Eltech Systems Corporation 17 - 19, Chemin du Champ-d'Anier CH-1209 Le Petit Saconnex Geneva Suisse

Decision under appeal:

Decision of Examining Division 019 Office dated 9 August 1982 application No 78 101 621.7 EPC

of the European Patent refusing European patent pursuant to Article 97(1)

Composition of the Board:

Chairman: R. Kaiser Member: 0. Huber

Member:

P. Ford

SUMMARY OF FACTS AND SUBMISSIONS

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I. European patent application No. 78 101 621.7 entitled "Dimensionally stable bipolar electrode, electrolysis cell comprising an electrode and electrolysis method" (original title), filed on 8 December 1978 and published on 27 June 1979 (publication No. 0 002 511) and claiming priority of 9 December 1977 from a previous application in USA, was refused by decision of Examining Division 019 of the European Patent Office, dated 9 August 1982. The decision was based on Claims 1 - 9 received on 4 December 1981, Claim 1 was worded as follows:

"An electrode for electrolytic processes comprising glassy carbon, characterized in that the electrode is a dimensionally stable porous reticulated structure, formed of strands of the glassy carbon."

The grounds for refusal were, that a skilled man, if faced with the problem as outlined in the description of the application (great resistance to oxidation, improved mechanical stability) would be naturally led to apply the method disclosed in FR-A-2 086 156 to the manufacturing of porous electrodes, thus coming to the subjectmatter of Claim 1. According to the decision, Claim 1 and the dependent Claims 2 - 7, independent Claim 8 concerning an electrolysis cell and independent Claim 9 relating to a method for the electrolysis of an aqueous electrolyte were not allowable due to lack of inventive step (Article 52 (1) in conjunction with Article 56 EPC).

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- II. On 11 October 1982 the appellant lodged an appeal against the decision and paid the appeal fee. The appellant submitted a Statement of Grounds on 20 December 1982.

In reply to several communications issued by the rapporteur pursuant to Article 110 (2) EPC, the appellant filed a new set of 8 Claims and an amended description on 7 April 1984.

The present independent Claims 1, 7 and 8 read as follows:

- 1. An electrode for electrolytic processes which has a dimensionally stable porous or reticulated crack-free glassy carbon structure obtainable by infusing polyurethane resin strands or reticulated polyurethane foams with a curable furan resin or resin precursor, removing excess furan resin from the surface of the strands or foams, curing the furan resin or precursor to form a thermoset and hardened structure and rapidly heating said structure in a non-oxidizing atmosphere to a temperature of at least 1000°C.
- 7. An electrolysis cell comprising at least one set of an anode and cathode and means for impressing an electrolysis current on the cell, characterized in that a least one of the anode and/or cathode is formed by the porous or reticulated electrode structure according to any preceding claim.

8. Method for the electrolysis of an aqueous electrolyte between an anode and cathode, characterized by using as the anode and/or cathode the porous or reticulated electrode structure according to any one of Claims 1 to 6.

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The appellant has requested the cancellation of the decision refusing the European patent application, the grant of a patent on the basis of the documents filed on 7 April 1984, the reimbursement of the appeal fee.

REASONS FOR THE DECISION

- The appeal complies with Articles 106 to 108 and Rule 64 EPC. It is therefore admissible.
- There is no formal objection to the current claims. They are adequately supported by the original documents.
- 3. FR-A- 2 086 156 discloses a method of manufacturing an electrode for electrolytic processes by heating between 1000°C and 3000°C a mixture of 1 to 80 parts of weight of carbon strands and 99 to 20 parts of weight of a bonding agent, both of a low tendency to graphitization. The bonding agent which may be, among others, a furan resin serves to connect the carbon fibres so that upon heating a continuous plate of glassy carbon is produced. The known method uses an already prepared carbon strand and the resin as a bonding agent, whereas according to the present application the carbon material is prepared from a mixture of polyurethane resin strands and a furan

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resin to give a porous or reticulated glassy carbon structure. A comparison of the densities of these known electrode materials with the claimed material makes the difference clear, namely FR-A- 2 086 156, see Table on p. 8 and 9: 0.8 - 1.6 g/cm^3 depending upon the starting material; in the case of the claimed electrode, see US-A- 3 927 186, col.6, 1. 66 and the Examples: 0.035 - 0.08 and the porosity values (70 % and 45 %) in the Examples of the application g/cm^3 . Therefore, the claimed electrode material is different from that as manufactured by the method described in FR-A-2 086 156.

According to DE-A- 2 036 394 only the pores of a graphite anode for electrolytic processes are filled with glassy carbon, which has been produced by baking resins.

DE-A-1 299 287 discloses only the covering of the surface of a graphite electrode with a coating of an electrocatalytic agent.

As indicated in the description of the application, see the last paragraph of p. 1 and the first paragraph of p. 2, electrodes are known which are prepared by impregnating polyurethane resin strands with volatile and/or decomposable compounds at the curing temperature of \leq 500°C or by impregnating porous graphite with volatile ammonium salts or with compounds capable of decomposing at the curing temperature of \leq 500°C. The missing step of heating to a temperature of at least 1000°C in comparison with an electrode according to claim 1 already yields a different electrode material. The process of manufacture as defined in Claim 1 of the application is completely described in US-A- 3 927 186. However, in this specification it is only said that glassy carbon manufactured in this way is useful for high temperature applications in the presence of neutral or reducing gas conditions or in a vacuum, see the Abstract.

Thus, the electrode, as set out in Claim 1, the electrolysis cell according to Claim 7 and the method for the electrolysis of an aqueous electrolyte according to Claim 8 are new.

- The question now to be examined is whether the subjectmatter of any of Claims 1, 7 and 8 involve an inventive step.
- 4.1. According to the description of the application, see p. 2, third paragraph, it is an object of the invention to provide novel dimensionally stable, porous electrodes made of glassy carbon which offer a great resistance to oxidation and a much improved mechanical stability. It is a constant preoccupation of manufacturers of electrodes to improve the mechanical stability and to reduce the deterioration in use of their products, see FR-A-2 086 156, p. 1 and p. 5, 11. 22 - 34.
- 4.2. According to the main Claim and the Examples of the application the structure of the claimed electrode is porous or reticulated. It consists of a mat of glassy carbon full of voids so that its density is very low. In contrast thereto, the electrode fabricated according to the method described in FR-A- 2 086 156 is a continuous plate with a relatively high density of 1,4 2,0 g/cm³ after

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heating to a temperature of at least 1000°C so a transformation to glassy carbon may take place. Given these fundamental structural differences, it was impossible for a person skilled in the art to foresee that a <u>porous</u> electrode prepared by impregnating polyurethane resin strands with volatile and/or decomposible compounds at the curing temperature ($\frac{4}{500}$ °C) would be improved by application of some method steps known from FR-A- 2 086 156, namely use of furan resin as impregnating material (see claim 3 in FR-A- 2 086 156) and additional heating to a temperature of at least 1000°C after reaching the curing temperature of the impregnating material (furan resin).

- 4.3. As the state of the art FR-A- 2 086 156, DE-A- 2 036 394) shows, improvements in electrodes were primarily achieved by reducing their porosity, so that the teachings of the prior art point away from the solution according to the subject-matter of the present application. Therefore, a man skilled in the art knowing the method for producing the porous or reticulated glassy carbon material described in US-A- 3 927 186 would never have the idea of using the known glassy carbon structure as electrode material for electrolytic processes even taking into account the fact that glassy carbon, in the form of a continuous plate, is used for the same purpose (FR-A- 2 086 156) or that the pores of graphite electrodes are filled with glassy carbon (DE-A- 2 036 394).
- 4.4. Finally DE-A- 1 299 287 is irrelevant, since it deals only with coated compact graphite electrodes.
- 4.5. Thus, the electrode in Claim 1 involves an inventive step (Article 56 EPC).

- 5. Claim 1 is allowable in accordance with Article 52 (1) EPC.
- Dependent Claims 2 6 relate to special embodiments of the invention in Claim 1 and are thus allowable.

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- 7. The allowability of Claim 1 involves the allowability of Claims 7 and 8 relating to an electrolysis cell and a method for electrolysis respectively using electrodes according to Claim 1.
- The effective description meets the requirements of Rule 27 EPC.
- An application was made for the reimbursement of the appeal fee in accordance with Rule 67 EPC.

The Examining Division pointed out in the Communication dated 19 May 1981 that the independent Claim 7 filed on 3 December 1980 was not allowable on the issue of lack of inventive step, reference being made to the observations in the first Communication of 29 July 1980. In answer to the Communication dated 19 May 1981, the appellant filed a new set of Claims on 4 December 1981, Claim 1 of which was similar to Claim 7 filed on 3 December 1980. Apart from amendments to the wording only, the facultative feature "and may be provided ... with a coating of an electrocatalytic agent" was omitted. Therefore, it was hardly surprising that the Examining Division likewise considered Claim 1 filed on 4 December 1981 as non-allowable. The non--allowability of a claim excludes the grant of a patent and the consequence is that the application had to be refused. The refusal was also based in the main on the same arguments as indicated in the two preceding Communications.

Prior to the issuance of the decision Oral Proceedings were not requested. Under these circumstances, the appellant was not denied its right under Article 113 (1) EPC to present comments.

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The Refund of the appeal fee can only be entertained when there has been a substantial procedural violation (Rule 67 EPC). In the opinion of the Board, no such violation occured in the present case.

ORDER

For these reasons it is decided that:

- The decision of Examining Division 019 of 9 August 1982 is set aside.
- 2. The case is remitted to the first instance with the order to grant a European patent on the basis of the following documents:

Description and 8 Claims received on 7 April 1984 provided that on page 3, 1. 2, the word "obtained" is replaced by "obtainable" (cf. Claim 1) and on page 5, line 4 and 9, and on page 7, 1. 24, the word "percursor" is amended to read "precursor".

 The request for reimbursement of the appeal fee pursuant to Rule 67 EPC is refused.

The Registrar:

The Chairman:

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