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Bezeichnung der Erfindung: An apparatus for controlling the electrical
Title of invention: parameters of an electrode reaction in an
Titre de l'invention : electrochemical cell

Klassifikation / Classification / Classement : G 01 N 27/46

ENTSCHEIDUNG / DECISION

vom / of / du 3 October 1986

Anmelder / Applicant / Demandeur :

Patentinhaber / Proprietor of the patent / Titulaire du brevet : International Business Machines

Hartmann & Braun AG

Einsprechender / Opponent / Opposant :

Stichwort / Headword / Référence :

EPÜ / EPC / CBE Articles 52(1), 54 and 56
"Novelty" "Inventive Step"

Leitsatz / Headnote / Sommaire

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Case Number : T 20/85

D E C I S I O N
of the Technical Board of Appeal 3.4.1
of 3 October 1986

Appellant : (Opponent)
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Representative :

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Decision under appeal : **Decision of Opposition Division of the European Patent Office dated** 23 November 1984 **rejecting the opposition filed against European patent No.** 20 898 **pursuant to Article 102(2) EPC.**

Composition of the Board :

Chairman : K. Lederer
Member : J. Roscoe
Member : F. Benussi

I

Summary of Facts and Submissions

I. European patent No. 20 898 was granted to the Respondent on 17 August 1983 with 4 claims in response to the European patent application No. 80 101 917.5 filed on 10 April 1980 claiming the priority of an earlier application made in the United States on 18 June 1979. Claim 1, the only independent claim, was worded as follows:

1. An apparatus for controlling the electrical parameters of an electrode reaction in an electrochemical cell having a counter electrode (16), a reference electrode (14) and a working electrode (12), the apparatus comprising circuit means adapted to supply current to the electrochemical cell (10) under the control of an externally applied control voltage (V_{in}) characterised in that the circuit means comprises a linear voltage to current converter means (20; 18, 20') the output impedance of which is much larger than the impedance of the electrochemical cell, said voltage to current converter means (20; 18, 20') having a non-inverting input (17) to which the said control voltage (V_{in}) is applied, an inverting input (19) which is connected to the reference electrode (14) and an output which is connected to the counter electrode, the working electrode (16) being connected to the ground potential of the said circuit means.

II. The Appellants filed notice of opposition against the European patent on 29 February 1984 requesting that it be completely revoked on the ground that the subject-matter of Claim 1 was lacking in novelty with respect to DE-A-2 155 935 and that of the remaining claims lacking in inventive step. Attention was also drawn by the opponents to the disclosure in the article by F. Tödt entitled "Die

Fachgruppe Elektrochemische Sonderverfahren", which appeared in Achema Jahrbuch 1962/1964 at pages 168-172. In what follows the above-mentioned patent document and article will be referred to as documents (A) and (B) respectively.

III. The Respondents requested that the opposition be dismissed. In support of their arguments they referred to the following textbooks:

"Integrated Circuit Pocket Book" by R.G. Hibberd published by Newnes-Butterworth, London 1972 (page 132) hereafter referred to as document (C), and "Operational Amplifiers" by G.B. Clayton, Second Edition, Butterworth Scientific LONDON 1979 (page 302) hereinafter referred to as document (D).

IV. The Opposition Division rejected the opposition in a decision of 23 November 1984. The reasons given for the decision were **inter alia** that document (A) nowhere stated that the amplifier 80 shown in Fig. 8 was a linear voltage-to-current converter having an output impedance much greater than the impedance of the electrochemical cell, and Fig. 6 of document (B) was essentially equivalent to that figure. Since this impedance relationship was an essential feature of Claim 1 the subject-matter of the claim was novel with respect to the circuits shown in documents (A) and (B).

Furthermore both of these documents confirmed the well-known practice in the field of the patent-in-suit, as acknowledged by reference to Fig. 1, of providing a voltage source when operating the cell in the potentiostatic mode. To use a voltage to current converter instead of a voltage source when operating in this mode the skilled man would have to depart from the path marked by the prior art, even though it was known to use a current source when operating in the galvanostatic mode.

Although the potentiostatic mode of operation had been known for a long time the prior art offered no evidence that any effort had been directed to solving the problems summarized in the patent associated with the known potentiostatic technique. Since these problems would be easily recognised and the advantages of the solution provided by the invention were indisputed it must be concluded that the invention was not obvious.

V. The Opponents filed notice of appeal on 15 January 1985 and paid the appeal fee at the same time. The Statement of Grounds was filed on 29 March 1985.

In this the Appellants (Opponents) argued that in the drawings of the patent-in-suit the electrochemical cell was shown in a simplified circuit. Thus, although it was stated that the output impedance of the converter (18,20,20') was much greater than the impedance of the cell, this impedance was not shown as a resistance in the line between the output of the converter and the counter electrode of the cell. If it were the circuit representation would correspond to that of Fig. 8 of document (A) in which a resistor 98 is shown in that line. The fact that a voltmeter 100 was connected across it meant that resistor 98 was necessarily high ohmic, and since the cell had a low resistance, the essential feature of Claim 1 that the output impedance of the converter was much greater than that of the cell was present.

VI. In their submissions in response the Respondents stated that the conclusion drawn by the Appellants concerning the value of resistance 98 was contrary to basic teaching, as was illustrated by pages 1059-1060 of the textbook Intermediate Physics by C.T. Smith (document E) and page 119 of Electricity and Magnetism by M. Nelkon (document F). They went on to say that the linear characteristic of the voltage

to current converter of Claim 1 arose from the stated relationship between the output impedance of the amplifier and the cell impedance.

VII. In a letter filed on 4 December 1985 the Appellants cited the article "Electrochemisches Messverfahren..." by Herrn Dr. Teske, which appears at pages 18-25 of VGB-Speisewassertagung 1971 (document G). This was said to describe with reference to Fig. 1 on page 19 the circuit shown in Fig. 6 of document B. In this description of the circuit (page 19, para. 3 left-hand col.) the transistor amplifier performing a potentiostatic function was stated to have output values of 5 volts and 2 milliamps. It thus had an output impedance of 2.5 K-ohms. According to the Appellants this was much larger than the impedance of the measuring cell which was normally far less than 500 ohms. These values could be substantiated by calculations based on the electrode dimensions and the properties of the cell liquid. The ratio of output impedance to cell impedance was thus 2000-200.

VIII. In an response filed 12 June 1986 the Respondents, while accepting the quoted value of 2500 ohms for the output impedance of the amplifer denied that the quoted value for the cell impedance had any basis in document (E).

IX. In a further letter filed on 26 July 1986 the Appellant accepted this lack of basis but contented that the skilled man knows that the measuring cell impedance could be, for example, about 100 ohms.

X. Though the Appellants do not explicitly say so it emerges clearly for the fact that their attack on all the claims is maintained that they are requesting that the decision of the Opposition Division be set aside and the patent revoked. The Respondents request that the appeal be dismissed.

II

Reasons for the Decision

1. The appeal complies with Articles 106 to 108 and Rule 64 EPC and is, therefore, admissible.

2. Fig. 8 of document (A) discloses an apparatus for controlling the electrical parameters of an electrode reaction in an electrochemical cell comprising a counter electrode 72, a working electrode 70, and a reference electrode 74. The apparatus includes circuit means 76 to 98 for supplying current to the cell under the control of an externally applied control voltage (taken from the tapping or resistor 92) and hence exhibits the combination of features set out in the preamble of Claim 1.

In this document however the amplifier is not said to be a linear voltage to current converter means. Furthermore the output impedance of the amplifier and the impedance of the electrochemical cell are not stated and neither of them can be deduced from the information available. Neither the Appellants unsupported contention that the connection of voltmeter 100 across resistor 98 implies that the latter, (and hence the output of the amplifier), has a high impedance nor their statement that the cell has a low impedance can be followed. The Board is satisfied that the representation of the "Verstärker" 80 indicated that it is a conventional operational amplifier. Such amplifiers are known, for example from document C, to have a low output impedance. This is confirmed by document D in which reference is made to the requirement in potentiostatic operation that the cell is driven by a low impedance source, which in this case is an operational amplifier. Further evidence that the amplifier is, or would be understood by

the skilled man to be, an operational amplifier comes from US-A-3 776 832, which is based on the same United States application as document A, in which the same amplifier is referred to as a conventional operational amplifier.

2.1 The conclusion drawn by the Appellants that the resistor 98 across which the voltmeter 100 is connected will thus have a high value cannot be accepted. In the basic textbook by C.T. Smith (document E) the passage at page 1059 line 33 to page 1060 line 5 indicates that in measuring a current by means of a voltmeter it is a low resistance which is connected in the circuit and that the measurement is inaccurate if the voltmeter itself does not have a very much higher resistance. Again at page 119 of the book by M. Nelkon (document F) where the voltage is measured by a potentiometer a similarly low value of resistor is placed in the circuit. While it is recognised that in low current circuits higher resistances may be needed to generate a voltage drop which is readily measurable by a voltmeter of a particular sensitivity this does not detract from the general principle that the resistor should be low in relation to the resistance of the circuit as a whole, and to that of the voltmeter.

In the absence of any evidence to the contrary regarding the value of resistor 98 it must therefore be assumed to be low.

2.2 Quite apart from this it is questionable whether, even if the resistance were high as the Appellant alleges, it could properly be regarded as constituting part of the output impedance of the amplifier, which performs its role without the aid of the resistor.

2.3 The resistance of the cell itself is also not stated in document A and since the Appellant has also produced no documents or argument to establish its value the argument that it is much lower than the output impedance of the amplifier, the value of which has likewise not been established, has not been made out. The Appellant has likewise failed to show that the amplifier functions as a linear voltage to current converter and the Board itself can find no reason for concluding that it is.

2.4 Fig. 1 of document G and Fig. 6 of document B both show the same circuit diagram but the associated description in the two documents differs. It is apparent from both that the apparatus represented by the diagram has all the features of the preamble of Claim 1 viz. counterelectrode G, reference electrode V, working electrode M and means P to supply current under control of an externally applied voltage U. However, in document B the unit P is not stated to be a linear voltage to current converter and neither its output impedance nor the impedance of the elements in the measurement range selector S nor that of the cell Z is stated. Thus it cannot be deduced from the information provided that the unit P is a linear voltage to current converter with an output impedance much greater than the cell impedance, which is an essential feature of Claim 1.

2.5 The only additional information given in document G is that the output voltage and current of the unit P are 5 volts and 2 milliamps. respectively and are sufficient for potentiostatic measuring purposes. The quoted values indicate an output impedance of 2.5 kilohms as the Respondents admit. In the absence of an indication of the impedance of the cell however it cannot be concluded from this that the impedance relationship condition of Claim 1 is

satisfied, still less that the amplifier produces a current which is linearly proportional to the difference in voltage between its two inputs.

2.6 The Appellants though admitting that the information in documents B and G affords no support for their statement that a measuring cell usually has an impedance much less than 500 ohms e.g. of the order of 100 ohms have produced no other evidence to support it. Following a careful study of the documents the Board finds that the only information regarding cell impedance is to be derived from Fig. 2 on page 20 of document G in which the measured (anode) current is plotted against the potential difference between the measuring electrode and the reference electrode. From the graphs it can be seen that the impedance of the part of the cell between the two electrodes referred to though dependent on hydrazine concentration and current is always well in excess of 2.5 kilohms. In the light of this the impedance of a measuring cell cannot be assumed to be much smaller than the above stated value of the output impedance of potentiostat P.

2.7 Apart from all this, although in the circuit of Fig. 6 of document B one input of the potentiostat is connected to the control voltage its other input is connected to the measuring electrode M rather than to the reference electrode V of the cell as required by Claim 1.

3. The subject-matter of Claim 1 and hence that of the appendant Claims 2 to 4 is therefore novel with respect to the documents relied on by the Appellant (Opponent). The Board having reviewed the documents C and D together with those cited on the Search Report is satisfied that these also do not put the novelty of the subject-matter of the

claims in question. Since the Appellant has not relied on any of them it is deemed unnecessary to justify this conclusion here.

4. According to the description of the patent-in-suit, in known potentiostatic control systems of the type set forth in the preamble of Claim 1, the means to supply current to the electrochemical cell is an operational amplifier the output voltage of which is linearly proportional to the difference between its two input voltages. This output voltage is applied across the cell to induce a cell current, hence acting as a current source. Such systems are said to exhibit instabilities due to electrochemical cell impedance, cell current monitoring circuits, and the roll-off characteristics of the operational amplifier. They are therefore operated with a restricted bandwidth to improve stability. They also require an additional high current device to measure the current, which increases the cost and increases the problem of thermal stability. In addition changing from a potentiostatic to a galvanostatic control mode is difficult and requires several other components.

4.1 Many of these limitations are stated to be overcome by the apparatus as claimed in Claim 1 which is said to provide a stable instrument for analyses requiring high frequency, which lends itself naturally to switching between galvanostatic and potentiostatic modes and monitors the cell system without requiring another high current device. The Appellants have not denied that the claimed apparatus provides such advantages, and the Board sees no reason to question this.

5. The question to be answered is, whether in the circumstances set out above, the subject-matter of the claims involves an inventive step.

5.1 As mentioned in 2 above document A discloses an apparatus according to the preamble of Claim 1. On the other hand it contains no discussion of the characteristics of the various circuit elements which are necessary to its successful operation. Nor does it refer to any problems, either those referred to in the patent-in-suit or others, associated with the operation in particular circumstances. It would therefore in the Board's view offer no assistance to the skilled man confronted with such problems and could not lead the skilled man into employing in the circuit in question a linear voltage to current converter having an output impedance much greater than the impedance of the cell.

5.2 In neither of the two documents B and G which describe the circuit shown in Fig. 6 of document B are the characteristics of the element referred to as "Potentiostat mit Transistorverstärker" discussed. The only solid information given in the texts is that relating to the voltage and current output, which enables the output impedance to be calculated as 2.5 kilohms. As already stated this intermediate value does not in itself allow one to conclude that the element is a linear voltage to current converter or that its output impedance is much greater than that of the cell. Still less does it, or anything else in these documents, provide any teaching that the use of such a converter is necessary to successful operation of the circuit or that it would be desirable in certain circumstances to overcome difficulties which might arise. The Appellants have moreover made no attempt to show that these documents contain such a teaching.

5.3 The documents cited in the patent specification which were not relied on during the opposition proceedings likewise give no hint towards the subject-matter of Claim 1. Among these documents US-A-3 855 101, which received a considerable amount of attention during the examination procedure,

also discloses an apparatus according to the preamble of Claim 1. In this however the only element the connections of which are identifiable with those of the linear voltage to current converter of Claim 1, namely the combination of the error signal amplifier and the power amplifier 42 controlled thereby, generates a logarithmic change in current in response to a change in voltage, and the impedance of the cell cannot be seen to be much less than the output impedance of the power amplifier. There is no suggestion whatsoever to modify these circuit elements to provide a linear relationship between input voltage difference and output current.

6. The Board's view therefore is that the subject-matter of Claim 1 would not have been obvious at the priority date from any of the documents relied on in the opposition or appeal proceedings or those cited in the patent specification. Consequently the subject-matter of Claim 1 involves an inventive step within the meaning of Article 56 EPC. The claim can therefore be maintained.

7. The dependent Claims 2 to 4 concern particular embodiments of the apparatus according to Claim 1 and can likewise be maintained.

ORDER

For these reasons it is decided that:

the appeal against the decision of the Opposition Division is dismissed.

The Registrar: The Chairman:

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