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Datasheet for the decision of 8 November 2024

Case Number: T 2472/22 - 3.3.05

Application Number: 15851413.3

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IPC: C01B32/05, B01J23/42, H01M4/86, H01M4/96, H01M8/10, H01M8/0234, H01M8/0243, H01M8/0245, H01M4/88

Language of the proceedings: EN

Title of invention:
CARBON SHEET, GAS DIFFUSION ELECTRODE BASE MATERIAL, AND FUEL CELL

Patent Proprietor:
Toray Industries, Inc.

Opponent:
Carl Freudenberg KG

Headword:
Electrode Base Material/TORAY

Relevant legal provisions:
EPC Art. 54, 56
RPBA 2020 Art. 11, 12(4), 12(6)

Keyword:
Late-filed evidence - circumstances of appeal case justify admittance (yes)
Novelty - main request (yes)
Inventive step - main request (no) - obvious modification
Remittal - special reasons for remittal (yes)

Decisions cited:
T 0651/91, T 0210/05

Catchword:

Europäisches Patentamt
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Beschwerdekammern

Boards of Appeal

Chambres de recours

Boards of Appeal of the
European Patent Office
Richard-Reitzner-Allee 8
85540 Haar
GERMANY
Tel. +49 (0)89 2399-0

Case Number: T 2472/22 - 3.3.05

D E C I S I O N
of Technical Board of Appeal 3.3.05
of 8 November 2024

Appellant: (Opponent)	Carl Freudenberg KG Höhnerweg 2-4 69469 Weinheim (DE)
Representative:	Steglich, Gregor Banse & Steglich Patentanwälte PartmbB Herzog-Heinrich-Straße 23 80336 München (DE)
Respondent: (Patent Proprietor)	Toray Industries, Inc. 1-1, Nihonbashi-Muromachi 2-chome Chuo-ku Tokyo 103-8666 (JP)
Representative:	Kador & Partner Part mbB Corneliusstraße 15 80469 München (DE)
Decision under appeal:	**Decision of the Opposition Division of the European Patent Office posted on 7 October 2022 rejecting the opposition filed against European patent No. 3228588 pursuant to Article 101(2) EPC.**

Composition of the Board:

Chair	G. Glod
Members:	T. Burkhardt
	S. Fernández de Córdoba

Summary of Facts and Submissions

I. The opponent's (appellant's) appeal lies from the opposition division's decision to reject the opposition against European patent No. 3 228 588 B1.

II. Of the documents discussed at the opposition stage, the following are relevant to the present decision:

E1 DE 11 2005 002 039 T5

E2 WO 2014/030553 A1

E2a EP 2 889 939 A1 (family member of E2 in English)

E8 M. Mathias et al., "Chapter 46: Diffusion media materials and characterisation" in "Handbook of Fuel Cells - Fundamentals, Technology and Applications, Volume 3: Fuel Cell Technology and Applications", John Wiley & Sons, 2003, pages 1-21

E10 P.K. Das et al., "Liquid-Water-Droplet Adhesion-Force Measurements on Fresh and Aged Fuel-Cell Gas-Diffusion Layers", Journal of The Electrochemical Society, 159 (5), 2012, pages B489-B496

E17 Affidavit by Mr. Masamichi Utsunomiya, 22 June 2021

E24 "Toray Carbon Fiber Paper 'TGP-H'", technical data sheet

III. With its grounds of appeal, the appellant additionally submitted *inter alia* the following evidence:

E30 Reproduction of Example 3 of E1

IV. With its reply, the patent proprietor (respondent) submitted *inter alia* the following evidence:

E32 Declaration by H. Sugaya comprising Attachment 1 "Freudenberg's E30", 21 June 2023

V. Independent claim 1 of the main request (version as granted) reads as follows:

Feature:

1 "A gas diffusion electrode substrate, comprising

2 a carbon sheet (1), the carbon sheet (1) being a porous carbon sheet comprising

3 a carbon fiber and a binding material, and

3.1 a hydrophobic material is included in the binding material, wherein said hydrophobic material is a fluorine-based polymer,

4 wherein when among layers obtained by dividing the carbon sheet in a through-plane direction thereof into three equal parts within a section (10) extending from a surface (5) having a 50% average fluorine intensity, which is closest to one surface (2) of the carbon sheet, to a surface (6) having a 50% average fluorine intensity, which is closest to the other surface (3) of the carbon sheet, one of a layer (7) close to one surface (5) has the largest average fluorine intensity, is a layer X, the other one of a layer (9) close to the other surface (6) has a smaller average fluorine intensity than that of the layer X (7), is a layer Y, and a layer (8) between the layer X (7) and the layer Y (9) is a layer Z, and wherein when a surface of the carbon sheet closest to the layer X is a surface X(2),

5 the gas diffusion electrode substrate has a microporous layer on the surface X side of the carbon sheet (1),

4.1 characterized in that the average fluorine intensity of the layer decreases in the order of the layer X (7), the layer Y (9) and the layer Z (8) as determined by the method defined in the description."

The feature numbering is that of point II.2 (page 4) of the decision under appeal.

VI. The arguments put forward by the appellant during the appeal proceedings which are relevant to the present decision can be summarised as follows:

The main request did not meet the requirements of Article 54 EPC in view of both the general disclosure of E1 and Example 3 of E1 when considering the results shown in E30.

The main request did not meet the requirements of Article 56 EPC in view of Example 3 of E1 in combination with E30 and E2/E2a.

The case should not be remitted to the department of first instance.

VII. The arguments put forward by the respondent during the appeal proceedings which are relevant to the present decision can be summarised as follows:

Document E30 was not to be considered.

The hatched areas in Figure 2b and Figure 2c of E1 were outside the carbon sheet.

The patent as granted met the requirements of the EPC.

The case should be remitted to the department of first instance to deal with the auxiliary requests.

VIII. Oral proceedings were held on 8 November 2024 as an in-person hearing.

IX. The appellant requested that the decision under appeal be set aside and that the patent be revoked.

The respondent requested that the appeal be dismissed. As an auxiliary measure, they requested that the patent be maintained in amended form on the basis of one of auxiliary requests 1 to 12 submitted with the reply to the grounds of appeal or auxiliary requests 1A to 12A, 1B to 10B or 12B submitted with the letter dated 18 September 2024.

Reasons for the Decision

1. Admission/consideration of document E30

1.1 The respondent requests that experimental evidence **E30**, which aims at reproducing Example 3 of E1, not be admitted since it should already have been filed before the opposition division (Article 12(6) RPBA).

This was even more the case since Example 3 of E1 had not even been mentioned in the notice of opposition.

1.2 However, for the reasons set out below E30 is considered.

In response to the opposition division's preliminary opinion, which acknowledged that **E1** discloses Feature 4 and Feature 4.1, the now respondent, two months before the oral proceedings, submitted new, different feature mapping, according to which the left side of the left peak and the right side of the right peak of Figure 2c of E1 could not be construed as parts of the "carbon sheet" of claim 1.

Compared with the preliminary opinion, the opposition division changed its view in the decision under appeal and accepted the new feature mapping (see item II.6.4).

In contrast to the qualitative Figure 2c of E1, the reproduction of Example 3 of E1 in E30 allows a quantitative assessment of the fluorine intensity profile through the carbon sheet.

Under these circumstances, E30 merely further develops the novelty objection against the subject-matter of claim 1 over E1 and is a fair reaction to the feature mapping used in the impugned decision.

In this specific context, it is of no relevance that Example 3 of E1 had not been mentioned in the notice of opposition.

As E30 was moreover submitted at the beginning of the appeal proceedings it is admitted (Article 12(4) RPBA).

2. Main request: novelty over E1

For the reasons set out below, neither Example 3 of **E1** nor its general disclosure anticipates the subject-matter of claim 1 (Article 54 EPC).

2.1 Example 3 of E1 with due consideration of E30

2.1.1 **E1** discloses in Figures 2a to 2c a gas diffusion electrode substrate comprising a porous carbon sheet, which comprises a carbon fibre and a binding material (see for example paragraphs [0001], [0006], [0015] and [0020]). The binding material includes a fluorine-based polymer (paragraph [0015] "erstes Fluorkohlen-wasserstoffpolymer").

Contrary to the respondent's view, the "carbon sheet" of claim 1 not only extends between reference numerals "10" and "11" of Figures 2a to 2c of E1 but also encompasses the hatched regions above and below in Figure 2b.

Because of the presence of protruding carbon fibres, the surface of the carbon sheet of E1 is not as even as shown in schematic Figures 2a to 2c, but has a certain roughness. Consequently, the carbon sheet does not end precisely at 10 and 11 but extends into the regions left of "10" and right of "11" in Figure 2c.

This is in line with the decreasing fluorine intensities in these regions. Because of the protruding fibres, the density of fibres in these outer regions is lower than in the centre of the carbon sheet and decreases towards the outside. As explained in paragraph [0040] of E1, polymer and solvent initially contained in the centre of the carbon sheet move

towards the outer surfaces upon heating. The solvent evaporates and the fluorine-comprising polymer remains attached to the carbon fibres in the outer regions. As the appellant submitted during the oral proceedings, this was shown in Figure 1 of **E10**. As there are fewer fibres to which fluorine can attach in the outer regions of the carbon sheet, fluorine intensity progressively decreases towards the outside, to the left to "10" and to the right to "11", in Figure 2c of E1.

In this regard it is noted that a fluorine intensity distribution measured in an uncompressed, fibrous carbon sheet cannot be plotted directly (i.e. without conversion of the scale) as a function of the measured thickness because thickness is measured while the carbon sheet is being compressed (as explained in paragraph [0027] of the patent in suit, for example).

If, by contrast, a portion of the fluorine polymer really left the carbon sheet of E1 upon heating and was deposited on the outer surface of the carbon sheet (as argued by the respondent and as apparently suggested by Figures 2a and 2b of E1), fluorine intensity in these polymer bulks in the outer regions in Figure 2c would be higher than in the central portion, not lower. Moreover, in view of the low polymer concentration in the solution in Example 3, i.e. 3% PTFE, deposition outside on the surface of the carbon sheet is not credible, and the respondent has failed to provide experimental evidence for such manifestly impossible behaviour.

The fact that the peak at "10" in Figure 2c is higher than that at "11" is a consequence of the asymmetric process of E1, with heating only being provided from

above (Figure 2a). This results in increased evaporation on the upper side of the carbon sheet (i.e. on the left side in Figure 2c) (see also E1: paragraph [0040], lines 14 to 21).

While the fluorine intensity profile of Figure 2c of E1 does not allow a quantitative evaluation (just like Figure 2 of the patent in suit), it allows the conclusion:
- that there has to be a surface having a 50% average fluorine intensity in the region left of "10"
- that there has to be a surface having a 50% average fluorine intensity in the region right of "11"
- that it is possible to define, from the left to the right in Figure 2c, a section with layers X, Z and Y having a decreasing average fluorine intensity in the order X > Y > Z.

Thus a layer X containing the peak at "10", a layer Y containing the peak at "11" and a layer Z in-between can be defined.

2.1.2 These findings are confirmed by **E30**, in which Example 3 of E1 is reproduced and in which a fluorine intensity profile resembling that of Figure 2c of E1 is obtained (see Figure 2 of E30).

The respondent argued that E30 was not a reliable reproduction of Example 3 of E1. According to expert declaration **E32**, infrared heating used in Example 3 of E1 and convection heating used in E30 were not comparable. The influence of the heating was confirmed by paragraph [0046] of E1. Moreover, in E30 only one sample was used, i.e. sample 9 ("Probe 9: LV22/11-7"), while paragraph [0109] of the patent in suit (and hence claim 1) required 50 samples.

However, these assertions are not convincing. Paragraph [0046] of E1 indicates that it is the heating *rate* (and thus the energy input) which is important, not the type of heating. Paragraph [0032] of E1 even mentions convection and infrared heating as alternatives.

With regard to the number of samples, the use of a single sample in E30 is perhaps not as reliable as the use of 50 samples. However, the respondent has provided no quantifiable distribution of the fluorine intensity to counter these findings.

2.1.3 On the other hand, the microporous layer of Example 3 of E1 is applied to the surface Y side of the carbon sheet, not to its surface X side as required by Feature 5 of claim 1.

In Example 3 of E1, while infrared heating is applied from above, a paste comprising a PTFE dispersion is applied to the bottom side. According to paragraph [0058]
- the paper is placed onto a scaffold ("Das Papier wird aus der Lösung entnommen und auf ein Gestell aus rostfreiem Stahl gelegt"),
- infrared heating is applied from above ("Eine Infrarotwärme wird über dem eingeweichten Kohlefaserpapier von einer Infrarotflächen-heizeinrichtung [...] aufgebracht" [...]), and
- a paste comprising a PTFE dispersion is applied to the opposite side, i.e. the bottom side towards the scaffold ("[...] wird eine Paste, die [...] PTFE-Dispersion [...] enthält, auf die Rückseite (d.h. die während des Trocknens gegen das Gestell gehaltene Seite) des Kohlefaserpapiers [...] stangen-beschichtet)".

The appellant did not contest this, but argued that an integral layer of the carbon sheet of Example 3 of E1 could be construed as the microporous layer of Feature 5. The pores of carbon fibre Toray TGP-H060 were in the micrometre range (see scales in the figures on page 1 of **E24** and at the top of Figure 1 of E30).

This is not convincing. **E8**, which illustrates the common general knowledge of the skilled person in the field of diffusion media, confirms that such a microporous layer has much smaller pores, i.e. pores having diameters between 100 nm and 500 nm (page 9, right-hand column, first paragraph).

Consequently, a layer of the carbon sheet of E1 closest to layer X cannot be construed as the microporous layer of Feature 5 of claim 1 of the patent in suit.

Consequently, Feature 5 is a distinguishing feature over Example 3 of E1.

2.2 General disclosure of E1

The appellant also argued that the general disclosure of E1 anticipated the subject-matter of claim 1.

This is not convincing either. Paragraph [0006] of E1 discloses a microporous layer on a desired side of the carbon sheet ("eine gewünschte Seite"). Although there are only two possible sides of the carbon sheet, this does not necessarily anticipate Feature 5 of claim 1 (see T 651/91, catchword). In addition, an example constitutes a specific embodiment that cannot be combined with other information of the description (T 210/05, Reasons 2.3).

Moreover, the general part of E1 does not disclose in a direct and unambiguous manner the claimed order of fluorine intensities in layers X, Y and Z (Feature 4.1). A merely probable disclosure, e.g. due to heating from only one side, is not sufficient. An explicit disclosure of the *combination* of features as claimed is not present in the general part of E1.

3. Main request: inventive step over E1

For the reasons set out below, Example 3 of **E1** in combination with E2/E2a renders the subject-matter of claim 1 obvious (Article 56 EPC).

3.1 The invention relates to a gas diffusion electrode substrate.

3.2 E1 also deals with a gas diffusion electrode substrate and also aims at improving the water management (paragraphs [0001] and [0006]).

Since E1 relates to the same technical field as the invention and since in particular Example 3 has numerous features in common with the subject-matter of claim 1, as set out above, this example is a suitable starting point for assessing inventive step.

3.3 According to the patent in suit, the problem to be solved is to provide a gas diffusion electrode substrate having high electrical conductivity for extracting generated electric current in addition to excellent gas diffusivity and water removal performance (paragraph [0022]).

3.4 It is proposed that this problem be solved by the gas diffusion electrode substrate of claim 1 being characterised by a microporous layer on the surface X side of the carbon sheet.

3.5 The respondent submitted comparative data **E17** to prove the benefits of the invention.

In contrast to inventive Example 2 of the patent in suit, comparative example 3 of E17 has the microporous layer on the surface Y side of the carbon sheet. This results in an undesirable decrease in output voltage from 0.43 V (last line of Table 1 of the patent) to 0.29 V (last line of Table 3 of E17). Consequently, a microporous layer on the surface X side would result in improved output voltage and thus in improved anti-flooding characteristics.

3.6 However, even if it were accepted that the problem posed has been successfully solved, inventive step cannot be acknowledged.

E2/E2a also relates to a gas diffusion electrode substrate (paragraph [0001]). Paragraph [0080] teaches that by arranging the microporous layer on the surface X side, i.e. the side having a higher fluorine intensity, the "cell performance of a fuel cell is more improved" and "drying of the electrolyte membrane is more inhibited by the back-diffusion of generated water".

In view of this incentive, the proposed solution is clearly taught in E2/E2a and the subject-matter of claim 1 does not involve an inventive step (Article 56 EPC).

The respondent argued that the arrangement of the microporous layer on the surface Y side in Example 3 of E1 was "teaching away".

This is not persuasive. Since paragraph [0006] of E1 indicates that the microporous layer is applied to a desired side ("auf eine gewünschte Seite") and as E1 discloses no reason why the microporous layer should not be applied to the surface X side of the carbon sheet, there is no "teaching away".

3.7 Therefore the requirements of Article 56 EPC are not met and the main request is not allowable.

4. Remittal to the opposition division

The appellant requested that the case not be remitted.

In accordance with Article 11 RPBA, a case shall not be remitted to the department of first instance unless there are special reasons.

In the case at hand, document **E30**, in which Example 3 of E1 was reproduced, was filed only during appeal proceedings and is admitted (see point 1. above). As explained in points 2.1 and 3., consideration of E30 is decisive for the negative assessment of inventive step, thereby leading to a change in the case compared with the impugned decision.

In addition, the auxiliary requests are not dealt with in the decision under appeal.

These are special reasons within the meaning of Article 11 RPBA, and in this context it is appropriate to remit the case to the opposition division.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the opposition division for further prosecution.

The Registrar: The Chair:

C. Vodz G. Glod

Decision electronically authenticated