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Datasheet for the decision of 30 April 2008

Case Number: T 1392/04 - 3.3.07

Application Number: 99107724.9

Publication Number: 0952241

IPC: C25B 11/04

Language of the proceedings: EN

Title of invention:
Supported Pt-Ru electrocatalyst, and electrodes, membrane-electrode assembly and solid polymer electrolyte fuel cells, using said electrocatalyst

Patent proprietors:
N.E. Chemcat Corporation

Opponents:
Umicore AG & Co. KG

Headword:
-

Relevant legal provisions:
EPC Art. 114(2), 111(1)

Relevant legal provisions (EPC 1973):
-

Keyword:
"Admissibility of fresh ground for opposition (no)"
"Late-filed document - admitted"
"Remittal to first instance (yes)"

Decisions cited:
G 0009/91, G 0004/95, T 0411/98

Catchword:
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Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number: T 1392/04 **-** 3.3.07

D E C I S I O N
of the Technical Board of Appeal 3.3.07
of 30 April 2008

Appellants: (Opponents)
Umicore AG & Co. KG
Rodenbacher Chaussee 4
D-63457 Hanau-Wolfgang (DE)

Representative:
Stellbrink, Axel
Vossius & Partner
Siebertstrasse 4
D-81675 München (DE)

Respondents: (Patent Proprietors)
N.E. Chemcat Corporation
4-1, 2-chome, Hamamatsu-cho
Minato-ku
Tokyo (JP)

Representative:
Wolff, Felix
Kutzenberger & Wolff
Theodor-Heuss-Ring 23
D-50668 Köln (DE)

Decision under appeal:
Interlocutory decision of the Opposition Division of the European Patent Office posted 30 September 2004 concerning maintenance of European patent No. 0952241 in amended form.

Composition of the Board:

Chairman: S. Perryman
Members: F. Rousseau
B. Struif

Summary of Facts and Submissions

I. The appeal by the Opponents ("Appellants") lies against the interlocutory decision of the Opposition Division, posted on 30 September 2004, upholding European patent No. 0 952 241 ("the patent") in amended form.

II. The claims in the application as originally filed and in the patent as granted were identical except for claim 5. Claims 1, 2, 3, 5, 7 and 8 as granted read as follows (the deletion made in claim 5 as filed being indicated by striking-out and the addition made, in bold and underlined):

"1. An electrocatalyst for use in solid polymer electrolyte fuel cells, comprising a cubic platinum-ruthenium solid solution alloy and a hexagonal ruthenium supported together on a conductive carbon carrier.

2. The electrocatalyst according to claim 1, wherein said cubic solid solution alloy and said hexagonal ruthenium each have a crystallite diameter of 10 to 100 Å.

3. The electrocatalyst according to claim 1, wherein the total amount of said cubic solid solution alloy and said hexagonal ruthenium supported ranges from 10 to 80 % by weight based on the electrocatalyst.

5. An electrode for use in solid polymer electrolyte fuel cells, comprising a water repellent-treated support substrate of a conductive and porous

carbon and a catalyst layer containing ~~said~~ **the** electrocatalyst ~~of this invention~~ **according to claims 1-4** and polymer electrolyte particles and formed on one side surface of said support layer.

7. A membrane-electrode assembly for use in solid polymer electrolyte fuel cells, comprising a solid polymer electrolyte membrane, a catalyst layer containing an electrocatalyst and polymer electrolyte particles, and a conductive and porous support substrate;
said catalyst layer and said support substrate having been formed in this order on both side surfaces of said solid polymer electrolyte membrane, wherein one set of the catalyst layer and the support substrate on one side of the solid polymer electrolyte membrane forms anode and another set of the catalyst layer and the support substrate on the other side of the solid polymer electrolyte membrane forms cathode,
wherein at least one of said catalyst layers contains the electrocatalyst as defined in claim 5.

8. A solid polymer electrolyte fuel cell, comprising said membrane-electrode assembly, an anode gas-distribution plate located to the support substrate of the anode, and a cathode gas-distribution plate located to the support substrate of the cathode of said membrane-electrode assembly as defined in claim 7."

III. The patent was opposed and revocation in its entirety asked for on the grounds of lack of novelty and inventive step (Article 100(a) EPC), and of an

extension of the subject-matter of the patent in suit beyond the content of the application as filed (Article 100(c) EPC). ***Inter alia*** the following documents were submitted in the opposition proceedings:

D4 R. Pattabiraman, "Role of reduction methods in the preparation of highly dispersed Pt-Ru electrocatalyst materials for anodes in direct methanol fuel cells (DMFCs)", Proceedings of the first international symposium on new materials for fuel cell systems, New Materials for Fuel Cell Systems 1, 1995, pages 362-374.

D5 R. Pattabiraman, "Development of Platinum-ruthenium alloy electrocatalysts for direct oxidation of methanol in fuel cells", Bulletin of Electrochemistry 9(5-7), 1993, pages 348-351.

D10 E-TEK, Inc. Gas Diffusion Electrodes and Catalyst Materials, 1995, Catalogue.

IV. The opposition division maintained the patent on the set of claims forming auxiliary request 2 before them. This set of claims is the same as the set of claims forming the Main request in the appeal proceedings. Claim 1 of this set reads as follows:

"1. An electrocatalyst for use in solid polymer electrolyte fuel cells, comprising a cubic platinum-ruthenium solid solution alloy and a hexagonal ruthenium supported together on a conductive carbon carrier, said cubic solid solution alloy and said hexagonal ruthenium each have a crystallite diameter of 10 to 100 Å, and

the total amount of said cubic solid solution alloy and said hexagonal ruthenium supported ranging from 10 to 80 % by weight based on the electrocatalyst."

The wording of claims 3, 5 and 6 of this set corresponds respectively to that of claims 5, 7 and 8 as granted, except that claim 3 now refers back to claims 1 or 2 only, claim 5 refers back to the electrocatalyst of claim 1, and claim 6 refers back to the electrode assembly as defined in claim 5.

V. The reasoning of the Opposition Division in their decision can be summarized as follows:

- The main request, maintenance of the patent as granted, was refused, since claim 1 as granted lacked novelty over document D4.

- The first auxiliary request, with a claim 1 corresponding to a combination of claims 1 and 3 as granted was refused for lack of inventive step. Novelty over D4 and D5 which disclosed a catalyst loading of 5% only was acknowledged. However the range of 10-80% claimed was so broad that an inventive selection over the prior art could not *a priori*, be recognized. The patentee's assertion that the claimed catalyst loading between 10 and 80% yielded an optimum balance between good dispersion and catalyst activity was not found convincing, because it remained undisputed that the selected optimum could not be independent of the specific surface area of the carbonaceous carrier, which was not specified in the claim. In

any event catalyst loadings falling within the claimed range were not unusual in the art (see D6 disclosing a loading of 9.5% Pt and 9.5% Ru).

- The second auxiliary request with a claim 1 corresponding to a combination of claims 1, 2 and 3 as granted, met the requirements of the EPC. It was novel over D4/D5 already because these disclosed a catalyst loading of 5% only. In accordance with D4 and D5 the Ru rich mixed phase had a particle size of above 13nm (see Table 1.) The crystallite size of free Ru was reported in D4 to be 12nm (see page 364). D4 and D5 did not therefore, teach crystallite sizes of free Ru particles within the claimed range.

- It was speculative to argue, as did the opponents, that a reduction with hydrazine instead of sodium formate, applied to a Ru rich 40 Pt/60 Ru mixture would inevitably yield Ru particles smaller than 10nm. The size of the crystallites obtained was not independent from the catalyst loading.

- It was evident that a smaller crystallite size would be beneficial for catalytic activity.

- It could be concluded therefore that the invention defined in claim 1 of the second auxiliary request provided a combination of small crystallite size particles of both the Pt/Ru alloy and the pure Ru particles, at a relatively high catalyst loading. Such a teaching was not found in D4/D5, taken alone or in combination with other prior art.

VI. The Appellants filed a notice of appeal on 8 December 2004 and paid the appeal fee. The grounds of appeal were filed on 9 February 2005, accompanied by experimental evidence purporting to show that use of hydrazine did produce crystallite sizes in the claimed range.

The Appellants further submitted with a letter dated 06 June 2006, ***inter alia*** the following document:

D18 A. Fischer et al., "Research of electrocatalysts for combustion of methanol and CO-contaminated hydrogen in PEMFCs", published in Proceedings of the second international symposium on new materials for fuel cells and modern battery systems, Montreal, Canada, July 6-10, 1997, pages 489-497, published in 1997.

VII. In response to the summons to attend oral proceedings before the Board, the Appellants submitted the following document:

D17a Handbook of Heterogeneous Catalysis, Volume 2, Edited by G. Ertl, H. Knözinger, J. Weitkamp, published by VCH, Weinheim, 1997, pages 446 to 450.

VIII. The Proprietors ("Respondents") in response to the summons, submitted eight sets of claims as main and first to seventh auxiliary requests for consideration by the Board. The main request corresponded to the version underlying the decision under appeal. The claims of the first auxiliary request corresponded to the claims of the main request, in which it was further

specified that the crystallite diameter was measured by XRD. The Respondents also submitted the following documents:

D22 Lee *et al.* "Effect of Drying Conditions of Au-Mn Co-Precipitates for Low-Temperature CO Oxidation", Journal of Catalysis 200, pages 298-308, 2001.

D23 Declaration from Mr. Akito Takayama dated 3 December 2007, on the relation between crystallite sizes measured by TEM and XRD respectively.

IX. Oral proceedings were held on 30 April 2008.

X. The submissions of the Appellants relevant to the present decision can be summarized as follows:

- The experimental evidence provided showed that the use of hydrazine as a reducing agent did indeed produce crystallites of a size in the claimed range, and was not a mere supposition.

- Claim 1 of the main request did not define any characterizing method for determining the diameter of the crystallites. Different techniques would lead to different values as evidenced by document D23.

- As shown by document D17a, even the use of X-Ray Diffraction line broadening analysis would provide different results depending on whether the Scherrer formula was employed or a complete line profile analysis was performed.

- Moreover, the lower limit defined in the patent in suit for the diameter of crystallite was lower than the extreme limit of size, i.e 1.5 to 2 nm, which can be according to document D17a measured by X-Ray Diffraction.

- In the absence of any statement of the method for measuring the diameter of the crystallites in the claim and relevant details for its application, claim 1 of the main request lacked clarity and insufficiency of disclosure arose.

- The electrocatalyst disclosed in document D18 contained only a small amount of pure platinum, as the (111) diffraction peak of the cubic platinum-ruthenium solid solution alloy was almost shifted at $2\theta = 40^{\circ}$, which meant that the total amount of the cubic solid solution alloy and hexagonal ruthenium was close to the total amount of platinum and ruthenium employed, i.e. was above 10 % by weight as required by claim 1 of the main request.

- Further, the present claims did not exclude the presence of unalloyed platinum.

- As the diameter of the crystallite had no clear meaning, it should be disregarded when assessing novelty and inventive step.

- Consequently, document D18 anticipated claim 1 and was therefore prima facie highly relevant.

XI. The submissions of the Respondents relevant to the present decision can be summarized as follows:

- An objection of lack of clarity of the feature defining a crystallite diameter of 10 to 100 Å could not be raised in appeal proceedings, as this feature was already contained in granted claim 2.

- An objection to lack of sufficiency of disclosure could not be introduced in the appeal proceedings, without the consent of the patent proprietor. This consent was expressly refused.

- Late-filed document D18, which was acknowledged to belong to the state of the art according to Article 54(2) EPC, was not highly relevant for the assessment of novelty.

- As far as the crystallite diameter was concerned, document D18 did not directly and unambiguously disclose that the cubic solution alloy had a crystallite diameter of 10 to 100 Å. The presence of unalloyed ruthenium in document D18 was only a supposition. The crystallite diameter of about 10 nm as determined by transmission electron microscopy exclusively referred to the platinum-ruthenium alloy.

- Further, the expression "about 10 nm" in document D18 did not disclose a particle size of 10 to 100 Å. The diameter of the crystallites was measured in the patent in suit by XRD, which technique was known from document D22 and as demonstrated by document D23, to provide larger values than that

obtained measured by transmission electron microscopy.

- In order to determine the diameter of those crystallites, which must be of course understood as the mean value, the skilled person would naturally apply the Scherrer formula.

- The electrocatalyst of document D18 contained clusters of pure unalloyed platinum on the conductive carbon in an unknown amount and consequently the total amount of cubic platinum-ruthenium solid solution alloy and hexagonal ruthenium did not meet the amount the requirements of claim 1, of from 10 to 80 % by weight based on the electrocatalyst.

- For considering inventive step, document D18 was less relevant than documents D4 and D5, because D18 taught that hexagonal ruthenium was detrimental to the catalytic activity.

XII. The Appellants requested that the decision under appeal be set aside, that document D18 be admitted into the proceedings and that the patent be revoked or that the matter be remitted to the first instance for further prosecution.

XIII. The Respondents requested that document D18 not be admitted into the proceedings and that the appeal be dismissed, or if document D18 is admitted into the proceedings that the matter be remitted to the first instance for further prosecution.

XIV. At the end of the oral proceedings the decision of the Board was announced.

Reasons for the Decision

1. The appeal is admissible.

Objections of lack of clarity and sufficient disclosure

2. Claim 1 of the main request requires that the crystallite diameters of the cubic platinum-ruthenium solid solution alloy and the hexagonal ruthenium are each from 10 to 100 Å, but is silent as to the method by which this parameter should be measured.

3. Claim 1 is a combination of granted claims 1, 2 and 3 as granted. The feature relating to the crystallite diameters was the subject of dependent claim 2 as granted, so that any lack of clarity of this feature was already present in the granted claims and does not arise out of the amendments made at the opposition/appeal stage. It is established case law that since an objection of lack of clarity under Article 84 EPC is not a ground of opposition, an objection of lack of clarity cannot be raised if the lack of clarity already existed, as here, in the granted claims, and has not been introduced by amendment. An objection of lack of clarity under Article 84 EPC against claim 1 of the main request thus cannot be considered in these appeal proceedings.

4. Fresh grounds of opposition may not be introduced at the appeal stage without the explicit consent of the

proprietor(see G 9/91 OJ EPO 1993, 408, point 18 of the decision). The ground of insufficiency of disclosure under Article 100(b) EPC had not been raised during the opposition proceedings here, and the respondents have expressly refused to consent to its introduction on appeal. The Board follows this Enlarged Board of Appeal decision, and accordingly refuses to consider this ground in these appeal proceedings.

Admissibility of late filed document D18

5. As stated in decision G 4/95 of the Enlarged Board of Appeal (OJ EPO 1996, 412) (see Reasons, point 4) "While the filing of facts and evidence by parties to opposition and opposition appeal proceedings is not precluded at any stage of such proceedings, the admissibility of facts and evidence filed at a late stage in such proceedings is always a matter of discretion for the EPO (see Article 114(2) EPC)". An important criterion in favour of admission is if such new material is ***prima facie*** highly relevant.

6. Document D18 was submitted only during the appeal proceedings, and then only after the four months period stipulated in Article 108 EPC for filing the statement setting the grounds of appeal. Its admission is thus a matter for the discretion of the Board. As the parties are both asking for remittal of the case to the first instance if it should be admitted into the proceedings, it is inappropriate for the Board to make detailed comments on document D18 if it decides to admit it into the proceedings and remit the case, in order not to pre-empt the decision to be taken by the first instance.

Disclosure of document D18

7. Document D18 concerns an investigation of ternary electrocatalysts for anodic methanol combustion in membrane fuel cells, in which the activity of ternary formulations of Pt/Ru/Sn and Pt/Ru/WO_x are compared with that of a Pt/Ru formulation as standard. The standard Pt/Ru catalyst is prepared from a commercial (E-TEK)Pt catalyst, which contains 10 wt.% Pt on active carbon, by reductive deposition of ruthenium from aqueous $RuCl_3$ solutions, using Na-formate as reducing agent (page 490, first paragraph of the experimental parts). For the standard, the total amount of platinum and ruthenium on the carbon support appears to be computable as 14,4% by weight based on the electrocatalyst.

8. The standard catalyst is characterized in document D18 by X-ray diffraction. The X-Ray diffraction of catalyst 1 is said (page 491, first paragraph) to reveal mainly reflexes of Pt (figure 2a, page 495) with a lattice constant, which differs somewhat from that of the pure Pt, in agreement with the formation of a Pt/Ru alloy. According to the same passage, Figure 2 of the same document allows one to distinguish at $2\theta = 44°$ the (101)-reflex of ruthenium.

9. The Respondents have disputed the existence of clusters of pure unalloyed ruthenium, arguing, citing page 492, lines 32 to 33, that it was merely "supposed", i.e. hypothetical. But it is a supposition based on XRD much as in the patent in suit.

10. The Respondents have also referred as distinctions to the fact that there will be some free platinum, and

that the effect of the free Ruthenium in D18 appears to be to lessen the activity of the catalyst. These possibilities do not appear excluded by claim 1.

11. Furthermore, document D18 discloses that the particle size obtained for the "self-made" catalyst is about 10 nm as can be determined by transmission electron micrographs (page 492, point 7. "Discussion").

12. There is thus a ***prima facie*** argument that this standard catalyst of D18 meets all the requirements of claim 1, and is thus more relevant than documents D4 and D5 already in the proceedings, in which the total amount of platinum and ruthenium on the carbon support is only about 5%, far below the requirements of the claim.

13. Document D18 also appears arguably more relevant than documents D4 and D5 in relation to inventive step. A stated object of the patent in suit is to provide an electrocatalyst having an excellent poisoning resistance to carbon monoxide when used as an anode in reformed gas fuel cells (see paragraphs [0007] and [0008]). Document D18 also relates to the poisoning resistance to carbon monoxide of platinum-ruthenium electrocatalysts in anodic oxidation of hydrogen (see page 490, lines 6 to 8, page 491, paragraph 6, "Current voltage curves" and page 496, Figure 3), whereas documents (4) and (5), considered in the contested decision in the evaluation of inventive step, only focus on different methods to provide platinum-ruthenium catalysts and their catalytic activity in direct oxidation of methanol, without addressing the question of carbon monoxide poisoning. Thus it needs to be considered whether document D18 is not a more

suitable starting point for evaluating inventive step than the documents so far considered.

14. The Board concludes that document D18 is so relevant that it needs to be admitted into the proceedings, and remittal to the first instance in accordance with the wishes of both parties in this situation is appropriate. The Board also considers that remittal is appropriate because the approach of neither the parties nor the first instance to the difficult issues involved has been wholly on the proper legal basis.

Guidance on legal approach to be adopted

15. The general rule is that the legal burden of proof is on the party who has to establish a fact.

16. In the case of lack of novelty the legal burden of proof is on the opponent. Claimed subject-matter would lack novelty only if it were derivable as a whole directly and unambiguously from a prior art disclosure and if a "clear and unmistakable teaching" of the combination of all claimed features (and not only the essential ones) could be found in said disclosure (see decision T 0411/98, point 4.1).

17. Where as in this case for the feature of crystallite size or the feature of the total amount of cubic platinum-ruthenium solution alloy and hexagonal ruthenium if this cannot simply be taken as the total amount of platinum and ruthenium (cf. paragraph [0020] and claim 3 of the patent), the claim is silent as to how a particular feature is to be measured, it is open to the opponent to adopt any method of measurement

which can be said to be standard for a skilled person in the art concerned, and show that on this method of measurement the claimed feature is already known from a particular piece of prior art.

18. If the proprietor wishes to argue that a feature in a claim needs to be measured in a particular way mentioned in the description, then for this argument to be accepted it is first necessary to limit the claim to this method of measurement by amendment, assuming this can be done meeting the requirements of Article 123(2) EPC. It is not enough to argue that the claim should be read in a particular way, when the wording of the claim does not require this. Article 69 EPC and the protocol thereto cannot be relied on to read a claim in a restricted sense which is not required by the wording of the claim itself. Article 69 EPC and its protocol were intended to allow a claim to be read by a court, considering an alleged infringement, in a broader way than might have been suggested by the strict wording of the claim, and were not intended to allow a claim to be read in a more restricted way than that suggested by its wording so as to avoid a novelty objection.

19. When it comes to considering inventive step the legal burden of proof of showing that at least part of the claimed subject matter can be derived in an obvious manner from the state of the art is on the opponent.

20. However, where the proprietor alleges that the problem to be solved by the subject matter claimed over the closest prior art is to obtain an improvement of some particular property of this closest prior art, the legal burden of proof that there is an improvement is

on the proprietor. A mere allegation that there is an improvement is not sufficient, there should be at least some experimental evidence that the particular property of the prior art is improved, when this closest prior art is modified in the minimum way necessary to fall under the subject matter now claimed. Experimental evidence is required to show that the improvement is necessarily attributable to the difference between the claimed subject matter and the closest prior art. The instances of the EPO should also be satisfied that this evidence makes it plausible that the problem has been solved over the whole range of the subject matter claimed.

21. It is of course not necessary that the claimed subject matter achieve an improvement. If the subject matter claimed provides an alternative to the closest prior art this may suffice for inventive step to be recognized if there are no reasons why the skilled person would modify the state of the art in the direction of what is claimed.

22. The treatment (see point V. above) in the decision under appeal of inventive step of the subject-matter of claim 1 of the second auxiliary request (corresponding to the main request in these appeal proceedings), does not state expressly what is being as treated closest prior art or what problem is to be solved in relation to this, nor is it clear whether an improvement is being acknowledged. The patent (see paragraph [0008]) refers to an object of the invention being to provide an electrocatalyst having excellent poisoning resistance to carbon monoxide for use in solid polymer electrolyte cells, but the patent provides no evidence

of the type outlined in point 20. above. The identification of the closest prior art, the problem to be solved, the evidence that this problem can be regarded as solved, and what the skilled person would or would not derive from the prior art, are all issues on which the evidence and arguments of the parties should focus.

Order

For these reasons it is decided that:

1. Document D18 is admitted into the proceedings.

2. The decision under appeal is set aside.

3. The case is remitted to the first instance for further prosecution.

The Registrar The Chairman

C. Eickhoff S. Perryman