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Datasheet for the decision of 15 September 2015

Case Number: T 0461/10 - 3.4.03

Application Number: 98117930.2

Publication Number: 0917166

IPC: H01G9/155

Language of the proceedings: EN

Title of invention:
Electric double layer capacitor and process for manufacturing the same

Patent Proprietor:
JAPAN GORE-TEX, INC.

Opponent:
F. Stemplinger

Headword:

Relevant legal provisions:
EPC 1973 Art. 100(b)

Keyword:
Sufficiency of disclosure - main request (no) - auxiliary request (no)

Decisions cited:
T 2222/09

Catchword:

Case Number: T 0461/10 - 3.4.03

DECISION
of Technical Board of Appeal 3.4.03 of 15 September 2015

Appellant: (Opponent)	F. Stemplinger Lindenstrasse 12 94051 Hauzenberg (DE)
Representative:	Adam, Holger Kraus & Weisert Patentanwälte PartGmbB Thomas-Wimmer-Ring 15 DE-80539 München (DE)
Respondent: (Patent Proprietor)	JAPAN GORE-TEX, INC. 42-5, 1-chome Akazutsumi Setagaya-ku Tokyo-To 156 (JP)
Representative:	Kador & Partner Corneliusstraße 15 80469 München (DE)
Decision under appeal:	**Decision of the Opposition Division of the European Patent Office posted on 30 December 2009 rejecting the opposition filed against European patent No. 0917166 pursuant to Article 101(2) EPC.**

Composition of the Board:

Chairman	G. Eliasson
Members:	V. L. P. Frank
	C. Schmidt

Summary of Facts and Submissions

I. This is an appeal by the opponent against the decision of the opposition division to reject the opposition against European patent EP 917 166 (Article 101(2) EPC).

The patent was opposed in its totality. Grounds of opposition were lack of novelty and inventive step and sufficiency of disclosure (Articles 100(a), 100(b), 54 and 56 EPC 1973).

II. At oral proceedings before the board, the appellant opponent requested that the decision under appeal be set aside and that the patent be revoked.

The respondent proprietor requested in writing that the appeal be dismissed or, alternatively, that the patent be maintained on the basis of claims 1 to 9 of the auxiliary request filed with letter dated 22 September 2010.

III. Independent claim 1 of the main request (patent as granted) reads:

"1. An electric double layer capacitor comprising a plurality of polarizable electrodes disposed in a row arrangement; a metal collector layer laminated to a polarizable electrode layer made from a porous sheet consisting principally of activated carbon; a separator being interposed between said polarizable electrodes; and an electrolyte provided between said polarizable electrodes and said separator;
wherein a carbon-based conductive material is interposed between said collector layer and said

polarizable electrode layer, and
said carbon-based conductive material penetrates into the voids in said polarizable electrode layer and is present in the voids of the polarizable electrode layer in a range of from 0.5 to 10% of the total void volume of the polarizable electrode layer."

Independent claim 1 of the auxiliary request differs from claim 1 of the main request in that the following feature was appended:

" and the polarizable electrode layer has a maximum pore size from 0.5 to 20 µm"

IV. The following documents are cited in this decision:

D1: JP 62 200715 A and the corresponding English translation

D6: http://www.shimadzu-techno.co.jp/technical/hunryutai.html, Shimadzu Techno-Research and the corresponding English translation of the table reproduced on page 12 of the appellant's statement of grounds of appeal.

A2: ASTM E 128-62, "Standard test method for maximum pore diameter and permeability of rigid porous filters for laboratory use"

A4: "A novel mercury free technique for determination of pore volume, pore size and liquid permeability", Jena, A and Gupta, K, P/M Science & Technology Briefs, Vol. 4, No 2, pp. 5-9, June 2002

V. In the decision under appeal, the opposition division found on sufficiency of disclosure essentially that:

- The independent claims required that the carbon-based conductive material was present in the pores of the polarizable electrode layer in a range of from 0.5 to 10% of the total void volume of the electrode layer, ie the *"filling ratio"*. The filling ratio could be determined by comparing the pore volume in the electrode layer after filling of the carbon-based conductive material with the pore volume in the electrode layer before said filling. The skilled person was well aware of methods for determining the total pore volume of a porous material (for example by Nitrogen adsorption/desorption method, BET, Mercury Intrusion Porosimetry...). Those methods were part of the common knowledge of the skilled person and therefore did not need to be included in the patent description. Samples for such measurements could be obtained after lamination of the electrode layer on the collector by cutting or slicing the final product. If the measurement of the pore volume was conducted before and after filling the carbon-based conductive material into the pores of the electrode, the filling ratio could be determined. The skilled person could thus establish whether or not a product characterized by the parameter "filling ratio" specified in the claims was obtained.

VI. The appellant opponent argued on sufficiency of disclosure essentially as follows:

- The European patent should disclose the invention in a manner sufficiently clear and complete for it to be carried out by a person skilled in the art. The opposed patent did not comply with this requirement since both independent claims 1 and 9 required that the carbon-based conductive material penetrated into the voids in the polarisable electrode layer and was present in the voids of that layer in a range of from 0.5 to 10% of its total void volume. These amounts however could not be determined by the skilled person either on the basis of the information given in the opposed patent or on the basis of his or her common general knowledge. In the following discussion the above feature was referred to as the *"filling ratio"*.

- The patent did not disclose any method for measuring the pore volume, since ASTM-E-128-61 (A2) cited in the patent was a standard test method for determining the maximum pore diameter and the permeability of a layer, not its pore volume, and was in any case not capable of measuring closed or blind pores. The methods which in the proprietor’s submission should enable the skilled person to determine the "filling ratio" were based on entirely different physical principles (permeability, gas adsorption/ desorption, mercury intrusion and scanning electron microscopy). According to table D6, the two methods available for pores as large as 18 µm (as mentioned in Example 1 of the opposed patent) were the mercury intrusion method and laser scattering. The latter method was suitable for measuring particle size distribution, but not pore size or volume. Even if these methods could be carried out to determine the "filling ratio" in a meaningful manner different results would be obtained by these four methods. Therefore, the skilled person wishing to measure the "filling ratio" was faced with an undue burden to select or rather guess the correct method for measuring the "filling ratio". The patent did not disclose any method for determining what was considered "the essential feature of the invention", nor did the proprietor disclose any actual method to determine the "filling ratio" during the opposition proceedings. It remained unknown what the "filling ratio" of the samples prepared in the working examples of the opposed patent were. These facts were a strong indication that the proprietor himself was not in a position to measure this obscure parameter.

- The working examples of the opposed patent were entirely silent on how to measure the "filling ratio" and did not disclose what was the actual "filling ratio" of the electrode layer obtained. Even assuming that the "filling ratio" of the electrode layers obtained in the worked examples was within the claimed range of 0.5 to 10%, the examples did not give adequate information how to obtain the claimed "filling ratio”. It was self-evident that the fluidity of the carbon-based conductive material, the pressure of the compression rolls and the temperature of these compression rolls had a bearing on the resulting "filling ratio". As the opposed patent was silent on these conditions, it was not possible to deduce the "filling ratio" from reworking the examples even if one assumed that the skilled person knew how to adjust the porosity and the maximum pore size of the polarisable electrode layer and had information about the depths into which the carbon-based conductive material penetrated the thickness of the electrode layer.

VII. The respondent proprietor argued on sufficiency of disclosure essentially that:

- The basic principles of the EPO decisions on sufficiency of disclosure were that a claim to a product defining the characteristics of the product by parameters should ensure that these parameters could be clearly and reliably determined by objective procedures which were usual in the art. Furthermore, the person skilled in the art reading the specification should be put in the position of being able to carry out the invention in all its essential aspects and of knowing when he was working within the forbidden area of the claims. The "filling ratio" as claimed could be clearly and reliably measured by standard methods which the skilled person was well aware of. Furthermore, all known methods for determining the filling ratio, such as the ethanol bubble point method according to ASTM E-128-61 or the mercury intrusion porosimetry method lead to substantially the same result regarding the filling ratio.

- Although different types of pores existed in different porous materials, the porosity before and after assembling the present electrodes was essentially the same and the passage through the coating rolls did not change the porosity of the already formed porous sheet. A transformation of pore shapes did not occur at usual pressures applied when passing the already formed porous sheet through the coating rolls.

- The appellant opponent argued that the ethanol bubble point method would not be capable of determining the pore volume of blind holes and closed cells, since air could not pass through these types of pores. However, blind holes and closed cells were not substantially present in the porous sheet according to the present invention. According to paragraph [0022] of the patent specification, the average particle size was selected to penetrate into the voids of the polarisable electrode layer. When the particle size was smaller than 0.5 µm, the carbon-based conductive material tended to penetrate far into the pores of the polarisable electrode layer whereas at a particle size of greater than 50 µm, the carbon-based conductive material did not readily penetrate into the voids of the polarisable electrode layer. This clearly indicated that by selecting a suitable average particle size of the carbon-based conductive material, the particles penetrated into the voids/ pores of the porous electrode layer. This would, however, not be possible if closed cells or blind holes formed a substantial part of the pores. For this reason the ethanol bubble point method was fully applicable to measure the pore volume and the maximum pore size of the sheet.

- The appellant opponent further argued that after lamination of the polarisable electrode material sheet onto the collector, one surface of the electrode layer was covered by the collector through which air could not pass. Allegedly it would be impossible to slice off the electrode layer alone but not including any excess of the conductive material remaining on the electrode layer which further added to the incapability of the ethanol bubble point method. Even if it was assumed, for the sake of argument, that the electrode layer could not be sliced off alone, the measurement was still possible before bonding the collector to the electrode layer via the carbon-based conductive adhesive material (bonding the conductive adhesive (3) to the electrode sheet (1) before applying the collector (2) thereto).

- A further allegation of the appellant consisted in the assertion that through holes of the electrode layer were covered by the carbon-based conductive adhesive material so that air could not pass through the specimen. This however was pure speculation, not supported by derivable facts.

- The fact that a feature of a claim may not be measured with absolute accuracy was not a reason alone for denying sufficiency of disclosure (see T2222/09, reasons 3.7). The appellant opponent did not show that an ambiguity in measuring the total void volume of the electrode layer existed (if there was any ambiguity at all), but rather argued that said total void volume was subject to measurement inaccuracies. The specification unambiguously showed that the claimed electric double layer capacitor could be obtained. The process for obtaining a polarizable electrode was disclosed in detail in paragraphs [0025] to [0030] of the patent specification. Specific embodiments according to such a preparation process were given in the working examples 1 and 2. The polarizable electrodes obtained according to the working examples clearly showed that improved internal resistance and low capacitance could be attributed to the claimed filling ratio and that the comparative examples did not show these effects as the carbon-based conductive material either did not adequately penetrate into the voids of the polarizable electrode layers or did not remain in sufficient qualities at the polarizable electrode layer - connector layer interface. The patent disclosed an average particle size of the conductive material of 0.5 to 50 µm, while the maximum pore size of the electrode layer was from 0.5 to 20 µm. This evidenced that the invention could be carried out in these ranges for average particle size of the conductive material and maximum pore size of the polarizable electrode layer.

VIII. With letter dated 10 September 2015 the respondent proprietor withdrew his request for oral proceedings and requested a decision according to the file. He further announced that he would not attend the scheduled oral proceedings.

IX. Oral proceedings were held on the 15 September 2015 in the absence of the respondent proprietor.

Reasons for the Decision

1. The appeal is admissible.

2. *Main request - Sufficiency of disclosure (Article 100(b) EPC 1973)*

2.1 It is common ground that the electric double layer capacitor of claim 1 differs from that disclosed in document D1 only in that the carbon-based conductive material, ie the adhesive joining the polarizable electrode layer and the collector layer, is present in the voids of the polarizable electrode layer in a range from 0.5 to 10% of the total void volume of the polarizable electrode layer (see D1, translation pages 3 and 4, Example 1, Figure 1). This feature may be restated as requiring that the *filling ratio* is in a range from 0.5 to 10%.

2.2 The appellant opponent argued essentially that the opposed patent did not disclose any method for determining the total void volume of the polarizable electrode layer, in particular after laminating the polarizable electrode layer on the collector layer, and furthermore, as different porosimetry methods gave different results, it was left to the skilled person to find out which method should be employed.

2.3 The respondent proprietor argued essentially that all methods for measuring the total void volume gave substantially the same result and that measuring porosity was within the general knowledge of the skilled person. Hence there was no need to describe these well known methods in the patent. This was also the view of the opposition division.

2.4 The opposed patent discloses that

(a) the porosity of the polarizable electrode layer should range from 40-90%, preferably 60-80% ([0018]),

(b) the maximum pore size of the polarizable electrode layer should be from 0.5-20μm ([0018]),

(c) the carbon particles of the carbon-based conductive material have an average particle size of 0.5-50µm ([0022]),

(d) the carbon-based conductive material present in the voids of the polarizable electrode layer should fill up 0.5-10%, preferably 1-5%, of the total void volume of that layer ([0023]), and

(e) the carbon-based conductive material should penetrate 0.15-30%, preferably 0.25-15%, of the thickness of the polarizable electrode layer, as determined by scanning electron microscopy (SEM) ([0024]).

In working example 1, referring to the polarizable electrode layer, it is disclosed that *"the sheet had a pore volume of 66% and 18µm maximum pore size (measured according to ASTM-E-128-61 using the ethanol bubble point)"* ([0035]).

2.5 The only measurement method disclosed in the patent, namely ASTM-E-128-61 using the ethanol bubble point (ie document A2), is however not a method for measuring the pore volume, but is a standard test method for measuring maximum pore diameter and permeability. It is neither disclosed in A2 nor in the opposed patent how to derive the total void volume from the maximum pore diameter and the permeability values obtained by applying the ethanol bubble point method of A2. Hence the board agrees with the appellant opponent that the patent does not disclose any measurement method for determining the pore volume of the polarizable electrode layer.

2.6 It now has to be assessed whether the skilled person would be able to determine the pore volume of the polarizable electrode layer before and after laminating this layer onto the collector layer on the basis of his common general knowledge which includes, as the respondent proprietor argued, general porosity measurement methods.

2.7 Pores in a layer may be classified in three types according to the number of surfaces they connect, as argued by the appellant opponent. *Through pores* go from one major surface of the layer to the opposite one, ie the connect two surfaces. *Blind pores* are open only to one surface, while *closed cells* are not connected to any surface. Some porosimetry measurement methods only detect through pores while other methods can measure through pores and blind pores. Closed cells may be measured by SEM in transverse sections through the layer.

2.8 The problems arising due to the existence of different pores types in a sample is illustrated in document A4. In the example of this document a sheet having a median pore diameter of 21µm was used (middle of page 4, Figure 4), ie a value comparable to the upper limit specified in the patent. The pore volume measured by liquid extrusion porosimetry was 2.834cm^3/g while the pore volume measured by the mercury intrusion porosimetry technique was 3.352cm^3/g. This difference in pore volume arises from the fact that the former method measures only the through pores while the latter measures through pores and blind pores. Hence the assertion of the respondent proprietor that all porosimetry methods deliver substantially the same results is not correct, since different methods may measure different kinds of pores.

2.9 According to the patent, a polarizable electrode layer 1 is laminated onto the collector layer (eg an aluminium foil) by coating a layer 3 of the carbon-based adhesive material on one surface of the collector layer, superimposing the electrode layer on the adhesive layer and passing the assembly through compression rolls 23, 24 to produce a 3-layer laminated sheet in which a portion of the adhesive is forced into the pores of the surface region of the sheet 1 ([0034]-[0037]; Figure 4).

2.10 While it may be argued that for the initial electrode layer 1 the total pore volume could be measured by the mercury intrusion method, it is not evident how this method could be applied once the polarizable electrode layer is laminated on the collector layer, since in that situation all the blind pores which were open to the surface now in contact with the collector layer are now covered by the collector and are hence no longer accessible. Hence the remaining free volume of a part of the blind pores after being partially filled with the adhesive cannot be measured using this method.

2.11 On the other hand, the liquid extrusion porosimetry method can also not be employed, since all through pores are now closed by (a) the adhesive in the pores, (b) the adhesive layer between the electrode and collector layer and (c) the collector layer itself. Hence, even if it could be argued that the collector layer could be sliced off and removed from the electrode/collector laminate, as argued by the respondent proprietor, it is not evident how the remaining pore volume could be measured, since the pores would still be closed by the adhesive inside the pores. This is not a pure speculation, as argued by the respondent proprietor, but a direct consequence of having the carbon-based adhesive penetrate into the pores of the polarizable electrode layer, as disclosed in the opposed patent (page 6, lines 24-25).

2.12 The opposed patent does not discuss any of these issues nor does it provide sufficient guidance to the skilled person that would enable him to determine the filling ratio of the polarizable electrode layer without having to embark on a research program.

2.13 It has now to be assessed whether the working examples of the patent disclose the necessary steps for manufacturing a polarizable electrode assembly having a filling ratio from 0.5 to 10%. If this would be the case, the skilled person would be in a position to manufacture an electric double layer capacitor according to claim 1 without having to be able to measure exactly the filling ratio of the electrode layer. The board finds however that the working examples of the patent do not provide sufficient information to the skilled person to that effect, since they do not disclose all the essential parameters, so that when attempting to repeat them, it is by no means certain that a so obtained electrode/collector laminate would have a filling ratio falling within the claimed range.

2.14 Although Example 1 discloses the composition of the carbon-based conductive adhesive, it does not disclose the pressure applied to the compression rolls 23,24 during the step of forming the 3-layer laminated sheet polarizable electrode/adhesive/collector in which a portion of the carbon-based conductive adhesive is forced into the pores of the surface region of the polarizable electrode layer ([0037]). The amount of carbon-based adhesive forced into the pores of the polarizable electrode layer depends directly on the pressure applied to the compression rolls and, hence, the filling ratio in the final product would depend on which pressure had been applied to the compression rolls. Hence, it is not certain that an electrode assembly having a filling ratio from 0.5-10% may be obtained by repeating the working examples of the patent.

2.15 Summarizing, the respondent proprietor did not present any explanation as to how the pore volume of the polarizable electrode layer could be measured before and after being laminated on the collector layer. He merely pointed to different porosimetry methods and argued that measuring the porosity was within the common general knowledge of the skilled person. The patent itself is silent on any porosity measurements in the general description of the invention and also when presenting its working examples. As already mentioned, the working examples do not disclose all the essential parameters that would enable the skilled person to reproduce them in order to obtain a capacitor with the claimed filling ratio.

2.16 The board judges, for these reasons, that the patent does not disclose the invention as claimed, ie the double layer capacitor and the corresponding manufacturing method, respectively, in a manner sufficiently clear and complete for it to be carried out by a person skilled in the art (Article 100(b) EPC 1973).

3. Auxiliary request - Sufficiency of disclosure (Article 100(b) EPC 1973)

3.1 Claim 1 of the auxiliary request differs from claim 1 of the main request in that the following feature was appended:

> "and the polarizable electrode layer has a maximum pore size from 0.5 to 20 µm".

3.2 Claim 1 of the auxiliary request however still comprises the same feature of claim 1 of the main request that the carbon-based conductive material is present in the voids of the polarizable electrode layer in a range from 0.5 to 10% of the total void volume of the polarizable electrode layer.

3.3 The board judges, for the reasons put forward in relation to the main request, that the invention claimed in the auxiliary request has not been disclosed in a sufficiently clear and complete manner (Article 100(b) EPC 1973).

4. In consequence, none of the requests of the respondent proprietor is allowable.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The patent is revoked.

The Registrar: The Chairman:

Beschwerdekammern des Europäischen Patentamts
Boards of Appeal of the European Patent Office
Chambres de recours de l'Office européen des brevets

S. Sánchez Chiquero G. Eliasson

Decision electronically authenticated