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Datasheet for the decision of 9 February 2017

Case Number: T 0554/12 - 3.4.03

Application Number: 00904006.4

Publication Number: 1158551

IPC: H01G9/04, H01G9/025

Language of the proceedings: EN

Title of invention:
SOLID ELECTROLYTIC CAPACITOR AND ITS PRODUCTION METHOD

Patent Proprietor:
Murata Manufacturing Co., Ltd.

Opponent:
Heraeus Precious Metals GmbH & Co. KG

Headword:

Relevant legal provisions:
RPBA Art. 12(4)
EPC 1973 Art. 54, 56

Keyword:
Late-filed documents - admitted (yes)
Novelty - main request (no) - first auxiliary request (no)
Inventive step - second auxiliary request (no)

Decisions cited:

Catchword:

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Case Number: T 0554/12 - 3.4.03

D E C I S I O N
of Technical Board of Appeal 3.4.03
of 9 February 2017

Appellant: (Patent Proprietor)	Murata Manufacturing Co., Ltd. 10-1, Higashikotari 1-chome, Nagaokakyo-shi, Kyoto 617-8555 (JP)
Representative:	Strehl Schübel-Hopf & Partner Maximilianstrasse 54 80538 München (DE)
Respondent: (Opponent)	Heraeus Precious Metals GmbH & Co. KG Heraeusstrasse 12-14 63450 Hanau (DE)
Representative:	Herzog, Fiesser & Partner Patentanwälte PartG mbB Immermannstrasse 40 40210 Düsseldorf (DE)
Decision under appeal:	**Decision of the Opposition Division of the European Patent Office posted on 27 December 2011 revoking European patent No. 1158551 pursuant to Article 101(3)(b) EPC.**

Composition of the Board:

Chairman	G. Eliasson
Members:	T. M. Häusser
	T. Karamanli

Summary of Facts and Submissions

I. The appeal of the proprietor concerns the decision of the opposition division to revoke the European patent No. EP-B-1 158 551 (Article 101(3)(b) EPC).

II. The opposition had been filed against the patent as a whole. Grounds of opposition were insufficiency of the disclosure, added subject-matter, and lack of novelty and inventive step (Article 100(a), (b), and (c) EPC and Articles 54 and 56 EPC).

III. Reference is made to the following documents:

D1: EP 0 340 512 A,

D2: EP 0 559 109 A,

D3: EP 0 264 786 A,

D4: US 4 858 078 A,

D10: Encyclopedia Chimica 3, compact edition, 32nd printing, edited by the Editing Committee for Encyclopedia Chimica, 15 August 1989, Kyoritsu Shuppan Co Ltd, p. 699,

D11: English translation of the table in the right-hand column of document D10,

D12: Elias Hans-Georg, *Makromoleküle*, 3rd edition, 1975, Hüthig & Wepf Verlag, p. 357,

D13: Sommer F, *Kautschuktechnologie*, 2nd edition, 2006, Carl Hanser Verlag, first page of chapter 2.1.1,

D14: Odian G, *Principles of Polymerization*, 2nd edition, 1981, John Wiley & Sons, page 35,

D15: *Ullmann's Encyclopedia of Industrial Chemistry*, 6th edition, 2002 electronic edition, Wiley, pages 1-2 of chapter 2.1.1,

D16: Ciesielski A, *An Introduction to Rubber Technology*, 2000, Rapra Technology Ltd, page 120.

IV. At the oral proceedings before the board the appellant (proprietor) requested that the decision under appeal be set aside and that the patent be maintained in amended form on the basis of the claims according to the main request filed on 22 March 2010, or one of the first auxiliary request filed on 22 March 2010 or second auxiliary request filed with the letter dated 28 May 2013.

The respondent (opponent) requested that the appeal be dismissed.

V. The wording of independent claim 1 according to the main request and the first and second auxiliary requests, respectively, is as follows (board's labelling "(a)" and "(b)"):

Main request:

"1. A solid electrolytic capacitor comprising a valve-acting metal (1) having a dielectric film layer (3) formed on the surface thereof, a solid electrolyte polymer layer (4) and an electrically conducting layer (5) which are formed on the dielectric film layer (3), characterized in that
(a) at least said electrically conducting layer (5) contains a rubber-like elastic material."

First auxiliary request:

Claim 1 of the first auxiliary request differs from claim 1 of the main request in additionally comprising the following feature:

(b) "and the solid electrolyte polymer layer (4) has a film-like or lamellar structure".

Second auxiliary request:

Claim 1 of the second auxiliary request differs from claim 1 of the first auxiliary request in that the first alternative in feature (b), i. e. the expression "film-like or", is deleted.

VI. The parties argued essentially as follows:

(a) Procedural issues - admission of documents D12-D16

The *appellant* requested that the documents D12 to D16 be disregarded for being late-filed. The expression "rubber-like elastic material" had been present in the patent as granted, so that these documents should have been filed earlier.

The *respondent* argued that the appellant had disputed only during oral proceedings before the opposition division that the polyurethane material of document D4 did not constitute the claimed rubber-like elastic material, with reference to documents D10 and D11 which had also been filed during these oral proceedings. The reply to the appeal was therefore the first occasion for the respondent to respond to these allegations by means of documentary evidence. As documents D10 and D11 had been admitted into the opposition proceedings it was only fair to admit documents D12 to D16 into the appeal proceedings. Furthermore, these documents were *prima facie* highly relevant since there were clear reasons to suspect that they would prejudice the maintenance of the opposed patent.

(b) Main request - novelty

The *appellant* argued that it was merely disclosed in document D4 that the utilized polyurethane material was soft and flexible, which did not automatically imply that the material was elastic. Moreover, this material was described as having an elastic modulus of 3 MPa, which was a value outside the range of between 0.1 MPa and 1 MPa stated in document D11 for a rubber-like elastic material. Hence, document D4 did not disclose the feature that the electrically conducting layer contained a rubber-like elastic material.

The *respondent* argued that the value of 3 MPa was within the range disclosed in document D11 when the appropriate accuracy of the stated range was taken into account. Furthermore, the expression "rubber-like elastic material" was broad and documents D12 to D15 showed that a material with an elastic modulus of up to 10 MPa, 20 MPa, 100 MPa, and 300 MPa, respectively, could be regarded as being rubber-like and elastic. Therefore, the polyurethane material described in document D4 could be considered as the claimed rubber-like elastic material.

(c) First auxiliary request - novelty

The *appellant* argued that it was not disclosed in document D4 to what extent the dielectric oxide layer was covered by the composite layer. Only the pores of the oxide layer might be filled by the composite layer.

The *respondent* argued that the solid electrolyte polymer layer of document D4 was at least film-like. The terms "layer", which was used in document D4, and

"film" were synonymous. Moreover, the dipping method used in document D4 for producing the composite layer was also used according to the patent (see Example 1) for producing the film.

(d) Second auxiliary request

(i) Novelty

The *appellant* argued that document D4 did not disclose that the solid electrolyte polymer layer had a lamellar structure. Rather, the dipping process for producing this layer was only performed once. Even the respondent had admitted this in its submission of 17 September 2013 (see page 16).

The *respondent* argued that it was left open in document D4 whether the dipping process for producing the electrolyte layer was performed several times. Such repeated dipping merely led to a thicker layer. In view of this, the layer disclosed in document D4 could be considered as having a lamellar structure.

(ii) Inventive step

The *appellant* argued that the claimed subject-matter differed from the closest state of the art, document D4, in that the solid electrolyte layer had a lamellar structure. The effect of this difference, which was distinct from a laminate structure, was the increased surface area of the solid electrolyte layer, which led to better adhesion of the succeeding layer, i. e. the conducting layer. Moreover, different electric properties were also achieved. The teaching of document D3 was merely to increase the thickness of a polymer layer by means of repeated polymerization. Hence, the skilled

person would not be led to combining documents D3 and D4 in order to solve the posed problem.

The *respondent* argued that the effect of the lamellar structure of the solid electrolyte layer was its increased thickness. It was therefore the objective technical problem to increase the thickness of the solid electrolyte layer. Document D3, in which *in situ* polymerization was used like in document D4, disclosed that repeated polymerization should be used in order to increase the layer thickness (D3, column 5, lines 32-35). It would therefore be obvious for the skilled person to perform such repeated polymerization in order to increase the layer thickness.

Reasons for the Decision

1. Procedural issues - admission of documents D12-D16

1.1 Documents D12 to D16 were cited for the first time by the respondent in its reply to the appellant's letter setting out the grounds of appeal.

The appellant requested that these documents be disregarded for being late-filed.

1.2 According to Article 12(4) RPBA, everything presented by the parties under Article 12(1) RPBA, in particular in the reply to the grounds of appeal (Article 12(1)(b) RPBA), shall be taken into account by the board if and to the extent it relates to the case under appeal and meets the requirements of Article 12(2) RPBA, the board having the power to hold inadmissible facts, evidence or requests which could have been presented or were not admitted in the first instance proceedings.

1.3 The respondent cited documents D12 to D16 in relation to the meaning of the expression "rubber-like elastic material", in particular in the context of the assessment of the novelty of the subject-matter of claim 1 of the main request in view of document D4.

The above expression had already been present in claim 1 as granted. Moreover, claim 1 of the main request and document D4 had been presented in the course of the proceedings before the opposition division. Documents D12 to D16 could therefore have been presented during the first instance proceedings.

Consequently, the board has the power not to admit these documents under Article 12(4) RPBA. Hence, the question arises whether the documents should be admitted into the proceedings or not.

1.4 During the course of the opposition proceedings the focus in the assessment of novelty and inventive step of the subject-matter of claim 1 of the main request shifted from documents D1, D2, and D3 to document D4. The latter document was submitted by the opponent (present respondent) about two months prior to the oral proceedings before the opposition division. The contentious issue concerning document D4 was whether the polyurethane material disclosed in this document could be considered the claimed "rubber-like elastic material". The value of Young's modulus of the polyurethane material being provided in document D4, the proprietor (present appellant) submitted at the oral proceedings before the opposition division documents D10/D11, in which a range of values of Young's modulus corresponding to rubber-like materials was indicated.

Documents D12 to D16 were submitted to provide further evidence in relation to the values of Young's modulus of rubber-like materials. In particular, by submitting these documents the respondent attempted to show that materials having higher values of Young's modulus than the upper limit of the range provided in documents D10/D11 might also be considered rubber-like. Furthermore, the board accepts that the respondent, having failed to provide such evidence during first-instance proceedings, submitted these documents at least at the earliest occasion after the submission of documents D10/D11, namely with its reply to the grounds of appeal. The appellant was therefore in a position to assimilate the documents and to react to them if deemed necessary.

The documents are also easy to comprehend and do not render the appeal proceedings more complex.

1.5 For these reasons the board, exercising its power under Article 12(4) RPBA, decided to admit documents D12 to D16 into the appeal proceedings.

2. Main request - novelty

2.1 In the decision under appeal the opposition division held that the subject-matter of claim 1 of the main request was not new over document D4 (see point 7.1 of the Reasons).

2.2 Document D4 discloses (see column 1, lines 6-9; column 4, lines 13-21; examples 7, 9 and 10) a solid electrolytic capacitor, wherein a conductive polymer compound is used as the solid electrolyte.

Example 10 is described with reference to examples 7 and 9 and relates to an aluminum capacitor, in which a

composite layer of polypyrrole and polyvinyl alcohol doped with toluene sulfonic acid ions is formed on a dielectric oxide layer. A layer comprising polyurethane and fine nickel particles (average particle size: 5 µm) is formed on the polyvinyl alcohol/doped polypyrrole composite layer. The polyurethane material is described as being a flexible, soft polymer compound and having a modulus of elasticity after drying of 0.3 kg/mm^2, while the modulus of elasticity of the polyurethane/Ni powder composite layer is 1.8 kg/mm^2.

2.3 There is agreement between the parties that document D4 discloses the preamble of claim 1 of the main request. Indeed, using the wording of that claim, document D4 discloses a solid electrolytic capacitor comprising a valve-acting metal (aluminum foil) having a dielectric film layer (dielectric oxide layer) formed on the surface thereof, a solid electrolyte polymer layer (polyvinyl alcohol/doped polypyrrole composite layer) and an electrically conducting layer (polyurethane/Ni powder composite layer) which are formed on the dielectric film layer (dielectric oxide layer).

2.4 The contentious issue between the parties is whether document D4 discloses the characterizing feature (a) of claim 1 of the main request, i. e. that at least the electrically conducting layer contains a rubber-like elastic material.

2.4.1 The opposition division held in the decision under appeal (see points 4.3, 4.4 and 7.1 of the Reasons) that the value of the elastic modulus stated in document D4, namely 0.3 kg/mm^2 corresponding to 3 MPa, implied that the polyurethane disclosed in D4 was elastic and rubber-like. This was also confirmed by document D11 which merely provided orders of magnitudes

for the lower and upper limits of the range of values of the elastic modulus of rubber-like elastic materials.

2.4.2 The appellant argued that it was merely disclosed in document D4 that the utilized polyurethane material was soft and flexible, which did not automatically imply that the material was elastic. Moreover, this material was described as having an elastic modulus of 3 MPa, which was a value outside the range of between 0.1 MPa and 1 MPa stated in document D11 for a rubber-like elastic material.

2.4.3 The board notes first of all that the expression "rubber-like elastic material" in feature (a) is intended to denote an elastic material which has the elasticity of a rubber-like material (see page 16, lines 25-26 of the opposed patent, which relates to the description of a conducting carbon layer comprising such an elastic material).

In relation to the disclosure in document D4 the board emphasizes the fact that it is stated in this document that the elastic modulus of the polyurethane material has a certain value. This modulus is a measure of a material's resistance to being deformed elastically, i. e. non-permanently, when a force is applied to it. It is defined as the slope of the material's stress-strain curve in the elastic deformation region. The mere fact that an elastic modulus is provided in document D4 for the polyurethane material is therefore considered to imply that it is an elastic material.

Moreover, it is appropriate to consider its value of the elastic modulus for deciding whether the polyurethane material of document D4 has the elasticity of

a rubber-like material. In particular, after surveying the values of the elastic modulus for various rubber materials it may be determined whether the value of the elastic modulus of the polyurethane material falls within the determined range of values. For example, documents D10/D11 disclose that the elastic modulus of rubber-like materials is between 0.1 MPa and 1 MPa; document D12 discloses that the elastic modulus of vulcanized rubber is between 1 MPa and 10 MPa; document D13 discloses that the elastic modulus of rubbers is between 0.1 MPa and 100 MPa. From these documents alone, but also from the skilled person's common general knowledge in relation to the elastic modulus of various materials it follows that the elastic modulus of 3 MPa provided in document D4 for the polyurethane material is well within the range values of the elastic modulus characterizing the elasticity of a rubber-like material.

2.4.4 Consequently, document D4 discloses - using the wording of claim 1 of the main request - that at least the electrically conducting layer (polyurethane/Ni powder composite layer) contains a rubber-like elastic material (polyurethane).

2.5 The subject-matter of claim 1 of the main request is therefore not new over document D4 (Article 52(1) EPC and Article 54(1) EPC 1973).

3. First auxiliary request - novelty

3.1 Claim 1 of the first auxiliary request differs from claim 1 of the main request in additionally comprising feature (b) according to which the solid electrolyte polymer layer has a film-like or lamellar structure.

The opposition division held in the decision under appeal that the additional feature was also disclosed in document D4 and that the subject-matter of claim 1 of the first auxiliary request was thus not new over document D4 (see point 7.2 of the Reasons).

3.2 The appellant argued that it was not disclosed in document D4 to what extent the dielectric oxide layer was covered by the composite layer. Only the pores of the oxide layer might be filled by the composite layer.

3.3 The board notes that the polyvinyl alcohol/doped polypyrrole composite layer of example 10 of document D4 serves as the second electrode (cathode) of the solid electrolytic capacitor, the first electrode (anode) being the aluminum foil (see D4, column 1, lines 6-9; column 8, lines 38-51). Moreover, in order to form the polyvinyl alcohol/doped polypyrrole composite layer the capacitor element is immersed in a pyrrole monomer solution for 5 minutes, then taken out from the monomer solution and immersed in an aqueous solution and kept at 0°C for 1 hour for polymerization (*ibid.*).

Hence, in view of the purpose of the polyvinyl alcohol/doped polypyrrole composite layer and its method of formation it is evident that the composite layer cannot merely fill the pores of the dielectric oxide layer but covers the oxide layer as a thin contiguous layer, which is considered to have the claimed film-like structure.

3.4 Consequently, the subject-matter of claim 1 of the first auxiliary request is not new over document D4 (Article 52(1) EPC and Article 54(1) EPC 1973).

4. Second auxiliary request

4.1 Novelty

4.1.1 Claim 1 of the second auxiliary request differs from claim 1 of the main request in additionally comprising the feature that the solid electrolyte polymer layer has a lamellar structure.

The respondent argued that it was left open in document D4 whether the dipping process for producing the electrolyte layer was performed several times, which could therefore be considered as having a lamellar structure.

4.1.2 The board notes that it is not mentioned in document D4 that the polyvinyl alcohol/doped polypyrrole composite layer of example 10 has a lamellar structure. Moreover, there is no indication in D4 that the dipping process for producing this layer is performed more than once. It is therefore neither explicitly nor implicitly disclosed in document D4 that this layer has a lamellar structure.

The subject-matter of claim 1 of the second auxiliary request is therefore new over document D4 (Article 52(1) EPC and Article 54(1) EPC 1973).

4.2 Inventive step

4.2.1 Closest state of the art / distinguishing features

Both parties argued in relation to inventive step starting from document D4. Indeed, this document discloses subject-matter that is conceived for the same purpose as the claimed invention, namely for providing

a solid electrolytic capacitor, and has the most relevant technical features in common with it, as detailed above. Document D4 is therefore regarded as the closest state of the art.

The subject-matter of claim 1 of the second auxiliary request differs from the device of document D4 (example 10) in comprising the feature that the solid electrolyte polymer layer has a lamellar structure (see points 2 and 4.1 above).

4.2.2 Objective technical problem

The appellant argued that it was the effect of the distinguishing feature, which was distinct from a laminate structure, that the surface area of the solid electrolyte layer was increased, which led to better adhesion of the succeeding layer, i. e. the conducting layer. Moreover, different electric properties were also achieved.

The board notes that there is no indication in the opposed patent concerning the purpose of the lamellar structure of the solid electrolyte polymer layer.

Regarding the method of production of the lamellar structure and the configuration of the lamellae it is mentioned in the patent with respect to example 9 (see paragraphs [0062]-[0063]) that the oxidized aluminum foil is dipped into an isopropanol solution, removed from the solution and air-dried, dipped into an aqueous solution, removed and allowed to stand at 60°C for 10 minutes to thereby carry out oxidation polymerization. The procedure from dipping into the isopropanol solution to oxidation polymerization is performed 25 times. Thereafter, the foil is washed with water and

dried to thereby form the solid electrolyte layer which exhibits a lamellar structure on the inner surfaces of micro-pores of the dielectric oxide film on the aluminum foil so as to cover the surfaces, space portions existing between the lamellar structure. The thickness of the solid electrolyte layer structure formed on the outer surface of the micropore structure is approximately 5µm and the thickness of one layer forming the lamellar structure is approximately 0.1-0.3µm.

Hence, the lamellae of the solid electrolyte layer are essentially stacked one on top of the other, so that only the outermost lamella is in contact with the succeeding conducting layer. In this respect there is no difference between the resulting lamellar structure and a laminate structure. It is therefore not considered to be an effect of the lamellar structure to improve the adhesion between the solid electrolyte layer and the succeeding conducting layer. Rather, the board agrees with the respondent in that the effect of the lamellar structure is merely to increase the thickness of the solid electrolyte layer. The objective technical problem is therefore to achieve this effect.

4.2.3 Obviousness

Document D3 concerns - like document D4 - a solid electrolytic capacitor and would thus be considered by the skilled person when attempting to solve the posed technical problem.

In particular, document D3 discloses (see column 2, lines 17-25; column 5, lines 18-38) a capacitor comprising an oxidized aluminum plate 1 surrounded by a polypyrrole layer 2, which is in turn covered by a

graphite layer 3 and a silver layer 4. The element is covered by an epoxy resin housing. Electrical connection can be established via contact terminals 5 and 6. In order to produce the polymer layer the oxidized surface of the metal plate is brought into contact with a monomer and a suitable oxidizing agent. Depending on the desired thickness of the polymer layer the coating process can be repeated several times.

The relevant teaching of document D3 is therefore to use repeated polymerization in order to produce a polymer layer of a desired thickness.

In order to solve the posed problem of increasing the thickness of the solid electrolyte layer, the skilled person would apply this teaching to the device according to the closest state of the art document D4, i. e. the capacitor of example 10 of that document, by repeatedly performing the process steps for forming the polyvinyl alcohol/doped polypyrrole composite layer, which are described under point 3.3 above. Such repeated polymerization corresponds essentially to the manner in which the lamellar structure of example 9 of the patent is produced (see point 4.2.2 above).

The board is therefore of the opinion that the skilled person would arrive in this way without exercising any inventive activity at a polyvinyl alcohol/doped polypyrrole composite layer having a lamellar structure.

Therefore, the subject-matter of claim 1 of the second auxiliary request does not involve an inventive step (Article 52(1) EPC and Article 56 EPC 1973).

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar: The Chairman:

S. Sánchez Chiquero G. Eliasson

Decision electronically authenticated