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DECISION of 25 April 2001

Case Number:

T 0642/98 - 3.3.3

Application Number:

93908096.6

Publication Number:

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IPC:

B29C 55/12

Language of the proceedings: EN

Title of invention:

Biaxially oriented polyester film

Applicant:

Teijin Limited

Opponent:

Headword:

Relevant legal provisions: EPC Art. 54, 56, 84

Keyword:

"Support (yes)"

"Novelty (yes)"

"Inventive step (yes)"

Decisions cited:

Catchword:



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Boards of Appeal

Chambres de recours

Case Number: T 0642/98 - 3.3.3

DECISION of the Technical Board of Appeal 3.3.3 of 25 April 2001

Appellant:

TEIJIN LIMITED

6-7, Minamihonmachi 1-chome

Chuo-ku

Osaka 541 (JP)

Representative:

Hallybone, Huw George Carpmaels and Ransford 43 Bloomsbury Square London WC1A 2RA (GB)

Decision under appeal:

Decision of the Examining Division of the European Patent Office dated 17 September 1998 issued in writing on 26 January 1998 refusing European patent application No. 93 908 096 pursuant to Article 97(1) EPC.

Composition of the Board:

Chairman:

R. Young

Members:

C. Idez J. De Preter

## Summary of Facts and Submissions

- I. European patent application No. 93 908 096.6, based on International application No. PCT/JP93/00485, filed on 16 April 1993, claiming the priority of the earlier Japanese patent applications No. 96655/92 of 16 April 1992 and No. 155015/92 of 15 June 1992 and published under No. WO 93/20999 (EP-A-0 590 159) on 28 October 1993, was refused by a decision of the Examining Division announced orally on 17 September 1997 and issued in writing on 26 January 1998.
- II. The decision was based on a set of 7 claims of a main request submitted with a letter dated 18 August 1997, and on a set of 7 claims of a first auxiliary request and a single claim of a second auxiliary request, both of the latter having been submitted at the oral proceedings held on 17 September 1997. Independent Claim 1 of the main request read as follows:
  - "A biaxially oriented polyester film which
  - (A) comprises 99.5 to 95 mole % of ethylene-2,6-naphthalenedicarboxylate and 0.5 to 5 mole % of ethylene-2,7-naphthalenedicarboxylate, and
  - (B) has (a) a resistance to dry heat deterioration at 200°C (a time in which 50% of the breaking strength is retained) of at least 2,000 hours,
  - (b) a resistance to wet heat deterioration at 130°C (a time in which 50% of the breaking elongation is retained) of at least 100 hours,
  - (c) a density of 1.355 to 1.370  $g/cm^3$ , and
  - (d) an intrinsic viscosity, as measured at 35°C in o-chlorophenol, of 0.60 to 0.90."

Dependent Claims 2 to 5 related to specific embodiments of the film according to Claim 1. Independent Claim 6 referred to an electrical insulating film comprising the biaxially oriented polyester film set forth in Claim 1 and independent Claim 7 was directed to the use of the biaxially oriented polyester film set forth in Claim 1, for electrical insulation.

Claim 1 of the first auxiliary request differed from Claim 1 of the main request only in that the range of intrinsic viscosity had been restricted to 0.64 to 0.85. Claims 2 to 7 of the first auxiliary request respectively corresponded to Claims 2 to 7 of the main request.

Claim 1 of the second auxiliary request read as follows:

"A method for producing a biaxially oriented polyester film which

- (A) comprises 99.5 to 95 mole % of ethylene-2,6-naphthalenedicarboxylate and 0.5 to 5 mole % of ethylene-2,7-naphthalenedicarboxylate, and
- (B) has (a) a resistance to dry heat deterioration at 200°C (a time in which 50% of the breaking strength is retained) of at least 2,000 hours,
- (b) a resistance to wet heat deterioration at 130°C (a time in which 50% of the breaking elongation is retained) of at least 100 hours,
- (c) a density of 1.355 to 1.370  $g/cm^3$ , and
- (d) an intrinsic viscosity, as measured at 35°C in o-chlorophenol, of 0.60 to 0.90, which comprises subjecting a polyester of composition
- (A) to melt extrusion to form an unstretched film, stretching the unstretched film 2.5 to 5.7 times in the longitudinal direction at 130 to 170°C and then 2.5 to 5.7 times in the transverse direction at 130 to 150°C,

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and then thermosetting the stretched film at 190 to 250°C."

The Examining Division refused the application on the grounds that the product claims of the main and the first auxiliary requests lacked the support required by Article 84 EPC and that the subject-matter of Claim 1 of the main, the first and the second auxiliary request as well as Claims 2 to 7 of the main and the first auxiliary requests did not meet the requirements of Article 56 EPC in view of documents D2 (FR-A-2 193 853), D3 (FR-A-2 224 505) and D4 (EP-A-0 225 631).

On the issue of support, the decision held that, due to the fact that there was no upper limit for the parameters resistance to dry and wet heat deterioration, the claimed polyester films according to the main and the first auxiliary requests were not obtainable by the teachings of the description.

On the issue of inventive step, the decision stated that any shortcomings in the heat resistance properties of the film disclosed in Run 5 of D2 could have been easily overcome by appropriately adjusting stretching ratios, heat setting conditions and molecular weight, which were technical measures belonging to the general knowledge of the person skilled in the art. Consequently, there was a lack of inventive step in view of D2. Furthermore, the claimed subject-matter represented a selection from the general teaching of D3 and D4 in two respects i.e. the content of 2,7-naphthalene dicarboxylic acid (2,7-NDA) and the combination of certain minimum values for heat stability and certain ranges of density and molecular weight. There was no evidence on file of the criticality of the amount of 2,7-NDA, however, and the minimum values for heat resistance were considered as arbitrarily set. Density, crystallinity and molecular

weight were, moreover, taught explicitly in D3 and D4 to have an influence on the heat resistance. Consequently, the claimed ranges only represented a mere optimisation work in the light of the teachings of D3 and D4, which the skilled person could have carried out without any inventive activity. The same considerations also applied for the stretch ratios and the heat setting temperatures as defined in the second auxiliary request. In short, Claim 1 of all the requests lacked an inventive step in view of any of the documents D2 to D4.

- III. A Notice of Appeal against the decision was lodged on 19 March 1998 by the Appellant (Applicant) together with payment of the prescribed fee. The Statement of Grounds of Appeal was filed on 29 May 1998.
- IV. In response to a communication of the Board annexed to a summons, issued on 13 December 2000, to Oral Proceedings to be held on 25 April 2001, the Appellant submitted, with his letter of 23 March 2001, three sets of claims forming respectively a new main request and two new auxiliary requests. In the communication the Board had raised inter alia the following objections:
  - (i) The novelty of the subject-matter of Claims 1, 3 to 7 of the main request and of Claim 1 of the second auxiliary request could not be acknowledged in view of Run 5 of D2.
  - (ii) The method mentioned in the description for determining the resistance to wet heat was not complete, since the quantity of water used in the autoclave was missing.
  - (iii) The wording "oligomer content" was unclear in the absence of an indication of the upper limit of the molecular weight of these compounds.
- V. During the oral proceedings held on 25 April 2001, a new main request of 6 Claims was submitted. The

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Appellant made the main request submitted with his letter of 23 March 2001 his first auxiliary request and maintained as second auxiliary request the second auxiliary request submitted with the letter of 23 March 2001. Claim 1 of the main request reads as follows:

"A biaxially oriented polyester film which comprises

(A) a polyester composed of 99.5 to 95 mole % of
ethylene-2,6-naphthalenedicarboxylate and 0.5 to 5 mole
% of ethylene-2,7-naphthalenedicarboxylate, and

(B) has (a) a resistance to dry heat deterioration at
200°C (a time in which 50% of the breaking strength is
retained) of at least 2,000 hours,

- (b) a resistance to wet heat deterioration at 130°C (a time in which 50% of the breaking elongation is retained) of at least 100 hours,
  - (c) a density of 1.355 to 1.370 g/cm<sup>3</sup>, and
  - (d) an intrinsic viscosity, as measured at 35°C in o-chlorophenol, of 0.64 to 0.85."

Dependent Claims 2 to 4 relate to preferred embodiments of the film according to Claim 1. Independent Claim 5 refers to an electrical insulating film comprising the biaxially oriented polyester film set forth in Claim 1 and independent Claim 6 is directed to the use of the biaxially oriented polyester film set forth in Claim 1, for electrical insulation.

- VI. The arguments presented by the Appellant in the written procedure and during the oral proceedings held on 25 April 2001 may be summarized as follows:
  - (i) The subject-matter of the main request was novel over Run 5 of document D2' (GB-A-1 441 304, corresponding to the French patent application D2) in view of the restricted range of intrinsic viscosity mentioned in Claim 1 of the main request.

- (ii) Run 5 was in fact presented as a comparative example in D2' whereas the whole teaching of D2' would lead away from the present application since it suggested to use mixtures of polyethylene-2,6-naphthalate resins with other polyesters instead of copolymers thereof in order to obtained biaxially oriented films with a better heat resistance.
- (iii) D3'(GB-A-1 450 491, corresponding to the French patent application D3) did not disclose examples of a copolyester of ethylene-2,6-naphthalene-dicarboxylate and ethylene-2,7-naphthalenedicarboxylate. There was also no direct relationship in D3' between the thermal stability and the molecular weight of the polyesters.
- D3' merely disclosed polyesters having a (iv) carboxyl group content of not more than 30 eq/ton and an intrinsic viscosity of at least 0.4 and did not teach that polyesters composed of 99.5 to 95 mole % of ethylene-2,6naphthalenedicarboxylate and of 0.5 to 5 mole % of ethylene-2,7-naphthalenedicarboxylate would allow the manufacture of films exhibiting good resistance to delamination in combination with good resistance to the heat deterioration as specified by parameters (B)(a) and (B)(b) in Claim 1. In that respect, Comparative Example 6 of the application in suit showed that the resistance of delamination of a film made of polyethylene-2,6-naphthalate homopolymer was very low.
- (v) Thus, the subject-matter of the main request also involved an inventive step.

- (vi) The fact that there was no upper limit for the parameters (B)(a) and (B)(b), was not relevant, since the invention merely taught that the claimed films must have minimum values in combination with features (B)(c) and (B)(d).
- (vii) It was clear from the description of the test for measuring the resistance to wet heat deterioration that adequate water must be added to the autoclave to ensure a wet environment, i.e. as the skilled person would assume, the presence of saturated vapour.
- (viii) The precise degree of oligomerisation was not material but merely the amount of oligomer in the polyester film as determined by the method indicated in the description of the application in suit.
- VII. The Appellant requested that the decision under appeal be set aside and a patent be granted on the basis of the claims of the main request filed at the oral proceedings, or of the claims of the first auxiliary request filed at the oral proceedings (previously the main request submitted with letter of 23 March 2001; cf. Section IV., above) or of the second auxiliary request filed with letter of 23 March 2001.

#### Reasons for the Decision

The appeal is admissible.

Main request

- 2. Amendments
- 2.1 Claim 1 of the main request differs from Claim 1 as originally filed by the indications that the polyester film "comprises a polyester composed of 99.5 to 95 mole % of ethylene-2,6-naphthalenedicarboxylate and 0.5 to 5 mole % of ethylene-2,7-naphthalenedicarboxylate", and that the intrinsic viscosity is in the range from 0.64 to 0.85.
- 2.3 The amended range of intrinsic viscosity finds its support on page 6, lines 13 to 16 of the description of the application as originally filed.
- 2.4 The composition of the polyester resin is to be found on page 5, lines 6 to 10 of the description of the application as originally filed. There is also no objection to the deletion of the word "substantially" from the expression "substantially comprises", since the polyester film according to Claim 1 remains defined by its essential component (i.e. the polyester resin) and by its essential characteristics (i.e. properties (B)(a), (B)(b), (B)(c), and (B)(d)), as was the case for the polyester film according to original Claim 1.
- Claims 2 to 6 respectively correspond to Claims 3, 5, 6,7, and 8 as originally filed.
- 2.6 Thus, the main request meets the requirements of Article 123(2) EPC.

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- 3. Clarity and support
- The fact that no upper limit has been indicated in 3.1 Claim 1 for the resistance to dry heat deterioration at 200°C and for the wet heat deterioration at 130°C does not lead in the present case to a lack of support. In fact, present Claim 1 only seeks to embrace values which should be as high as can be attained above specified minimum levels (i.e. 2000 hours for the resistance to dry heat deterioration and 100 hours for the resistance to wet heat deterioration), given the other parameters (intrinsic viscosity and density) of the claimed film. Consequently, the finding in the decision under appeal that the claims lacked support to the extent that they covered polyester films which were not obtainable by the teachings of the description is not pertinent. On the contrary, there is no lack of support in the sense of Article 84 EPC.
- 3.2 The disclosure of the application in suit gives details of how to determine the resistance to dry heat deterioration (cf. page 10, lines 16 to 28) and the amount of oligomers (cf. page 10, line 29 to page 11, line 4). Although the description of the method for determining the resistance to wet heat deterioration (cf. page 9, line 27 to page 10, line 15 of the application as originally filed) does not mention the amount of water in the autoclave, it is credible to the Board in view of the test described in Examples 11 and 12 of D3' that, as submitted by the Appellant, the person skilled in the art would understand that enough water should be present in order to carry out the test in presence of saturated vapour. Thus, the Board takes the view that the parameters (B)(a) and (B)(b) and oligomer content are sufficiently defined so that the requirements of Article 84 EPC are fulfilled in this respect also.

- 4. Closest prior art, the technical problem and its solution
- 4.1 The patent application in suit relates to biaxially oriented polyester films.
- 4.2 Whilst there is a conspicuous absence of any reference to a specifically identified relevant prior art document in the introductory description of the application in suit, such films are described in documents D2', D3' and D4.
- 4.3 The documents D2', D3' and D4 may be summarized as follows.
- D2' refers to a biaxially oriented polyester film, 4.3.1 which comprises a blended mixture of (1) a first polyester resin whose dicarboxylic acid units are derived from naphthalene-2,6-dicarboxylic acid (2,6-NDA) and optionally one or more other dicarboxylic acids and whose dihydric alcohol units are derived from ethylene glycol and optionally one or more dihydric alcohols, the relative proportions of the units being such that the sum of x, the mol % relative to the total dicarboxylic acid component, of units derived from dicarboxylic units other than 2,6 NDA, and y, the mol % relative to the total dihydric alcohol component, of units derived from dihydric alcohols other than ethylene glycol is lower or equal to 10, and (2) 0.5 to 10% by weight based on the first resin of a second polyester resin whose dicarboxylic acid units are derived from a principal dicarboxylic acid and optionally one or more other subsidiary dicarboxylic acids and whose dihydric alcohol units are derived from a principal dihydric alcohol and optionally one or more subsidiary dihydric alcohols, the relative proportions of the units being such that the sum of p, the mol % relative to the total dicarboxylic acid component, of

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units derived from the subsidiary dicarboxylic acids, and q, the mol % relative to the total dihydric alcohol component, of units derived from the subsidiary dihydric alcohols is lower or equal to 10 with the proviso that if said principal dicarboxylic acid is 2,6 NDA, said principal dihydric alcohol is not ethylene glycol, said film having a softening point at least 1°C higher than the equilibrium softening point of the mixture of components (1) and (2). The film exhibits a good resistance to thermal degradation, as indicated by its break elongation after having been exposed at 200°C for 400 hours. The polyester resin (1) preferably has an intrinsic viscosity of 0.35 to 0.80, while that of the second resin is preferably greater than 0.25 (cf. D2', Claim 1; tables 1 and 2; page 2, lines 111 to 127). In a comparative example (Run 5) D2' discloses a biaxially oriented polyester film obtained by extruding a copolyester of ethylene-2,6-naphthalate containing 2% by mole of ethylene-2,7-naphthalate having a intrinsic viscosity of 0.62 to form an unstretched film having a thickness of 335 microns, stretching the unstretched film 3.5 times in the longitudinal direction at 140°C, then 3.7 times in the transverse direction at 130°C and heat setting the stretched film for 15 seconds at 235°C. The elongation at breakage after heat degradation of this film is lower than that of a film obtained from a blend comprising 98% by weight of polyethylene-2,6-naphthalate and 2% by weight of polyethylene-2,7-naphthalate (cf. D2', Table 1, Runs 2 and 5).

4.3.2 D3' relates to a biaxially oriented film comprising a linear polyester which contains units derived from 2,6 NDA, units derived from ethylene glycol and optionally units derived from one or more dicarboxylic acids other than 2,6 NDA and/or units derived from other dihydric alcohols other than ethylene glycol, the sum of molar percent of said other dicarboxylic acids (based on the

total dicarboxylic acid component) and said other dihydric alcohols (based on the total dihydric alcohol component) being not more than 10 percent, the polyester having an intrinsic viscosity of at least 0.40 and a concentration of free carboxyl groups of not more than 30 equivalents/metric tonne. The film retains high tensile strength and high elongation at break in particular when exposed to wet heat atmosphere at an elevated temperature and to a dry heat atmosphere at an elevated temperature and is suitable as insulating material for electric appliances (cf D3', Claim 1; page 3, lines 68 to 85 and 104 to 111). Although the use of 2,7-NDA as comonomer is mentioned in D3' (cf. page 1, line 75), the examples only refer to homopolymers of ethylene-2,6-naphthalate (cf. Examples 1 to 6, and 9 to 12) or to copolymers comprising ethylene terephthalate units (cf. Examples 7 and 8). In particular, Example 9 of D3' discloses a film obtained from polyethylene-2,6-naphthalate homopolymer having an intrinsic viscosity of 0.69.

D4 discloses polyethylene-2,6-naphthalate biaxially 4.3.3 oriented films having a Young's modulus of not smaller than 600 kg/mm<sup>2</sup> both in the machine direction and in the transverse direction and a degree of crystallinity not smaller than 40% (i.e. a density of at least 1.3578 g/cm³). The polyethylene-2,6-naphthalate polymer may be modified with less than 10% of a third component such as 2,7-NDA but only films obtained from the homopolyester are exemplified. The intrinsic viscosity of the polyethylene-2,6-naphthalate polymer is preferably between 0.6 and 0.9. The films are used as plastic film capacitors and exhibit, in view of their high degree of crystallinity, a good dimensional stability and good resistance at soldering temperature (cf. D4, page 6, line 5, to page 7, line 11; page 7, line 25 to page 8, line 4; page 14, lines 5 to 14; Examples 1 to 3).

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- As indicated in the introduction of the application in suit, biaxially oriented polyester films which are used for electrical insulation should exhibit good resistance to dry heat and wet heat deterioration and not give rise to delamination (interlaminar peeling).
- Consequently, D3', which relates to the use of biaxially oriented polyester films on the basis of polyethylene-2,6-naphthalate as insulating materials and which, alone of the documents referred to, deals with the improvement of the resistance of these polyester films to dry heat and wet heat deterioration, in particular Example 9 thereof, qualifies, in the Board's view, as the closest state of the art.
- 4.6 Thus, starting from Example 9 of D3', the technical problem may be seen in the provision of biaxially oriented polyester films having further useful properties for application as electrical insulation films in addition to resistance to dry heat and wet heat deterioration.
- The solution proposed according to the application in suit is to provide a film a priori fulfilling certain minimum requirements of resistance to wet heat and dry heat deterioration and comprising a copolymer composed of 99.5 to 95 mole % of ethylene-2,6-naphthalene-dicarboxylate and 0.5 to 5 mole % of ethylene-2,7-naphthalenedicarboxylate instead of a polyethylene-2,6-naphthalate homopolymer, and having a density of 1.355 to 1.370 g/cm³ and an intrinsic viscosity of 0.64 to 0.85, as set out in Claim 1.
- In view of Examples 1 to 5 and comparative Example 6 of the application in suit, which show that the claimed films have an improved resistance to delamination in combination with the required good resistance to dry heat and wet heat deterioration, the Board is satisfied

that this technical problem is effectively solved by the combination of features according to Claim 1.

### 5. Novelty

Lack of novelty was not a ground for the refusal of the application. Furthermore, in view of the restricted range of intrinsic viscosity of 0.64 to 0.85 mentioned in Claim 1, the novelty objection raised in the communication of 13 December 2000 of the Board in view of Run 5 of D2 cannot be maintained. The subject-matter of Claim 1 is therefore novel (Article 54 EPC).

#### 6. Inventive step

It remains to be decided whether the claimed subjectmatter is obvious in view of the cited prior art.

- 6.1 There is no mention in D3' of delamination resistance, let alone a combination of this property with specific levels of resistance to wet heat and dry heat deterioration. While it is disclosed in D3' that the polyethylene-2,6-naphthalate polyester might be modified by comonomers such as 2,7-NDA in an amount of up to 10% by mole (cf. D3', page 1, lines 17 to 75), there is no indication in this document of whether a film comprising such copolymer might exhibit delamination resistance. Even less is there any suggestion that the further conditions of having a density of 1.355 to 1.370 g/cm<sup>3</sup> and an intrinsic viscosity of 0.64 to 0.85 would contribute to attaining the relevant goal. Thus, D3' itself cannot suggest the solution of the technical problem.
- 6.2 D2' refers to the problem of improving the heat resistance of films based on polyethylene-2,6-naphthalate resins, useful as insulating materials.

  Although D2' discloses in a comparative example (Run 5)

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a biaxially oriented film comprising a copolymer of ethylene-2,6-naphthalate and ethylene-2,7-naphthalate which still shows a good breaking strength after exposure to dry heat (cf. D2', table 1), D2' will in fact lead the person skilled in the art away from the solution of the technical problem, since it teaches to use mixtures of polyethylene-2,6-naphthalate with other polyesters instead of copolymers thereof in order to improve the heat resistance of the films. Furthermore, D2' is totally silent on the resistance to wet heat deterioration and to delamination. Thus, D2' cannot provide any assistance in solving the technical problem.

- Although D4 relates to polyethylene-2,6-naphthalate films having a good heat resistance and useful for the manufacture of capacitors, there is no reference to delamination resistance or to resistance to wet heat deterioration, let alone a specific combination of these qualities. In particular, there is no suggestion further to modify the polyethylene-2,6-naphthalate polyesters with 0.5 to 5 mole % of ethylene-2,7-naphthalate for such a purpose. Thus, D4 contains no hint to the solution of the technical problem.
- 6.4 It follows that the solution of the technical problem does not arise in an obvious way from the cited state of the art. Hence, Claim 1 meets the requirements of Article 56 EPC.
- 7. Thus, Claim 1 of the main request is allowable. The patentability of Claims 2 to 6 is supported by the patentability of Claim 1 to which they refer, and these claims are therefore also allowable.
- 8. Since the main request can be granted, there is no need to consider the auxiliary requests submitted by the Appellant.

#### Order

# For these reasons it is decided that:

- 1. The decision under appeal is set aside.
- 2. The case is remitted to the Examining Division with the order to grant a patent on the basis of the claims of the main request filed during the oral proceedings after any necessary consequential amendment of the description.

The Registrar:

The Chairman: