

Publication in the Official Journal ~~Yes~~ / No

File Number: T 184/89 - 3.3.2

Application No.: 83 302 904.4

Publication No.: 0 095 349

Title of invention: Laminates of lamellar articles and polyolefins

Classification: B32B 27/32

**D E C I S I O N**  
of 25 February 1992

Applicant: E.I. DU PONT DE NEMOURS AND COMPANY

Headword:

EPC Article 56

Keyword: "Inventive step (yes)"

Headnote



Case Number : T 184/89 - 3.3.2

**D E C I S I O N**  
of the Technical Board of Appeal 3.3.2  
of 25 February 1992

**Appellant :** E.I. DU PONT DE NEMOURS AND COMPANY  
1007 Market Street  
Wilmington  
Delaware 19898 (US)

**Representative :** Jones, Alan John  
CARPMAELS & RANSFORD  
43 Bloomsbury Square  
London WC1A 2RA (GB)

**Decision under appeal :** Decision of Examining Division of the European  
Patent Office dated 28 September 1988 refusing  
European patent application No. 83 302 904.4  
pursuant to Article 97(1) EPC.

**Composition of the Board :**

**Chairman :** A.J. Nuss  
**Members :** M.M. Eberhard  
R.L.J. Schulte

## Summary of Facts and Submissions

- I. European patent application No. 83 302 904.4 (publication No. 0 095 349) was refused by a decision of the Examining Division. The decision was based on nine claims filed on 9 January 1988.
- II. The ground for refusal was that the claimed laminate did not involve an inventive step in the light of the disclosure in EP-A-0 015 556 (D1) and GB-A-1 436 361 (D2).

According to the decision, the closest state of the art was represented by D2 since the latter had more structural features in common with the claimed subject-matter than D1. The non-homogeneous stratified structure of layer (b) of D2 was considered to be equivalent to the claimed structure in which the incompatible polymer was present as parallel and overlapping lamellae. In the Examining Division's view, the only difference between the claimed laminates and those of D2 was that the blend layer contained a condensation polymer as incompatible polymer instead of a saponified ethylene-vinyl ester copolymer. The problem to be solved was thus to find an alternative for the latter copolymer in layer (b) of D2. The Examining Division considered that it was obvious to replace the heterogeneous layer in the laminate of D2 by the one claimed in the present application since both layers, in the form of films, were known from D1 to have good fluid barrier properties.

- III. The Appellant (Applicant) lodged an appeal against this decision. With the Statement of Grounds of Appeal, he filed an amended set of claims as the main request. Claim 1 of this request reads as follows:

"An at least two layer laminate comprising:

- (a) a layer of a polymer comprising a base polyolefin in the form of a continuous matrix phase, a condensation polymer incompatible with the base polyolefin, and a polyolefin which has carboxylic moieties attached thereto either on the backbone itself or on side chains, wherein the condensation polymer is present in the continuous matrix phase as substantially two-dimensional, parallel and overlapping lamellae of material and the polyolefin which has carboxylic moieties attached thereto is present between the lamellae and adheres the lamellae together, and
- (b) a layer of a polyolefin or polyolefin copolymer film, said layer comprising 10 to 90 percent of the thickness of the laminate;

said layers (a) and (b) being melt bonded together such that the molecular networks of the layers at the lamination site are adhered together."

IV. At the oral proceedings held on 25 February 1992 the Appellant handed over an amended set of claims as auxiliary request. During these proceedings he referred to his written submissions as regards inventiveness of the laminate according to Claim 1 of the main request. He particularly stressed that the whole document D2 was centred on the use of a specific saponified ethylene-vinyl ester copolymer in the blend layer and that the skilled man would have had no incentive to dispense with a copolymer which was considered as essential in D2. He further argued that in view of D2 and D1 the skilled man, faced with the problem of providing a laminate with improved barrier properties to hydrocarbons, would have

had at least three alternatives. Thus he could have replaced the polyolefin layer of the laminate of D2 by the heterogeneous film disclosed in D1, or he could have retained the polyolefin layer and substituted the film of D1 for the blend layer of D2, or he could also have contemplated applying the film of D1 on the two-layer laminate of D2, i.e. without omitting any layer. The Appellant emphasised that the teaching of D1 and D2 gave the skilled person no incentive to choose the second alternative. He concluded that the finding in the decision under appeal was based on ex post facto analysis.

- V. The Appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of Claims 1 to 7 filed on 1 February 1989 and Claims 8 and 9 filed on 9 January 1988 as main request, or on the basis of Claims 1 to 7 filed during oral proceedings as auxiliary request, and a description to be adapted.

#### Reasons for the Decision

1. The appeal is admissible.
2. There are no objections under Article 123(2) to the amended claims of the main request. Claim 1 is based upon Claim 1 of the application as originally filed and upon additional features stated in the description thereof. The replacement of the term "alkylcarboxyl-substituted polyolefin" by the sentence "a polyolefin which has carboxylic moieties attached thereto either on the backbone itself or on side chains" is supported by page 9 (lines 26 to 30) of the description. The features that the base polyolefin is "in the form of a continuous matrix phase" and "the condensation polymer is present in the continuous matrix phase as substantially two-dimensional,

parallel and overlapping lamellae" find support on page 3, lines 11 to 17 and page 5, line 8. The dependent Claims 2 to 9 correspond to the original Claims 2 to 9.

3. In reply to the objection of lack of clarity raised by the Examining Division against Claim 1 of 9 January 1988, the Appellant has amended this claim as proposed in point 1 of the decision under appeal. The Board agrees with the Examining Division that Claim 1 thus amended, i.e. Claim 1 of the main request, meets the requirements of Article 84.
  
4. The patent application relates to an at least two-layer laminate exhibiting good fluid barrier properties, which comprises a heterogeneous three-component blend layer and a layer of a polyolefin, these two layers being melt bonded together. The heterogeneous layer comprises a continuous polyolefin matrix and lamellae of a condensation polymer incompatible with the polyolefin.
  - 4.1 Document D2 is considered to represent the closest prior art. It discloses laminates comprising
    - (a) at least one layer of a crystalline film-forming polyolefin and/or at least one layer of a saponified ethylene-vinyl ester copolymer, and
    - (b) at least one blend layer comprising a blend of (A) a crystalline film forming polyolefin, (B) a saponified ethylene-vinyl ester copolymer and optionally (C) a thermoplastic polymer containing carbonyl groups in the main or side chain, for example an acrylic acid-grafted polyethylene.

These two layers are melt bonded together. In the alternative where the laminate is composed of a blend

layer and a polyolefin layer the thickness of the latter is about 9 to 97%, preferably 50 to 96% of the thickness of the laminate. The blend layer (b) may have a non-homogeneous stratified structure (cf. Claims 1, 8, 12, 18, 23; page 4, lines 15 to 55; page 8, lines 3 to 4 and 11 to 23; page 9, lines 25 to 48; page 34, line 8).

These laminates exhibit good gas permeation resistance, in particular a very low permeability to a gas such as oxygen and steam, and a good resistance to delamination. They are useful in containers for aliphatic or aromatic organic solvents since a decrease in the volume of the content can be minimised (cf. page 2, lines 14 to 25; page 3, lines 3 to 11; page 15, lines 1 to 19).

- 4.2 In the light of this prior art, the problem underlying the present application can be seen in providing laminates which exhibit good barrier properties to hydrocarbons such as toluene and which can be produced in the form of shaped articles such as bottles. In this respect, the Board notes that the problem formulated in the decision under appeal partially foreshadows the solution. Such a definition of the problem is not in line with the jurisprudence of the Boards of Appeal (cf. T 229/85, OJ 1987, 237 and T 99/85, OJ 1987, 413).

It is proposed to solve the problem stated above by melt-bonding a three-component heterogeneous layer as defined in Claim 1 to a layer of polyolefin, the layers having the relative thickness indicated in this claim. In view of the permeability tests carried out with toluene and reported in Examples 1 and 2 of the application and of the statement on page 13, second paragraph, it appears to the Board that this problem has been plausibly solved.

5. Novelty of the laminates was not disputed by the Examining Division. After examination of the cited documents the Board has also come to the conclusion that the claimed laminates are novel since D1 does not concern a laminate but a single film and the laminates according to Claim 1 differ from those of D2 by the composition and the structure of the heterogeneous layer.
6. It remains to be examined whether the claimed solution meets the requirement of inventive step.
- 6.1 It is pointed out in D2 that the laminates are useful in containers for aliphatic or aromatic organic solvents, since a decrease in the volume of the content can be minimised. However, the more specific case of permeation resistance to hydrocarbons is not dealt with and D2 itself does not contain any information as to how the known laminates could be modified to achieve this purpose.

Document D1 relates to the manufacture of a single film which is suitable for making shaped articles such as bottles and has improved barrier properties to permeation of fluid, liquid and gaseous materials (cf. page 1, lines 5 to 10, page 3, lines 15 to 19 and examples). This film comprises a heterogeneous blend of a polyolefin, a second polymer incompatible with the polyolefin, and a polyolefin having carboxylic moieties attached thereto either on the backbone itself or on side chains. The "incompatible polymer" may be a polyamide, a polyvinyl alcohol, a copolymer of ethylene and vinyl alcohol or a polyester. It is present as substantially two-dimensional, parallel and overlapping layers embedded in the continuous matrix, these thin layers being adhered together by the polyolefin which contains carboxylic moieties (cf. Claims 7, 8; page 4, lines 7 to 19; page 9, lines 26 to 30). In the Examples 1 to 10, a polyamide was used as the



"incompatible polymer" and bottles were blow moulded with the polymer mixture. The permeation tests carried out in these examples show that the bottles exhibit barrier properties to permeation of unleaded gasoline which are thirty-five times to seventy-five times as great as those of a polyethylene control. The films of Examples 11 to 13 include a polyvinyl alcohol or a copolymer of ethylene and vinyl alcohol as the "incompatible polymer", however only their permeability to oxygen and water vapour was tested so that it cannot be derived from D1 whether or not these films have a good permeation resistance to hydrocarbons.

- 6.2 The Examining Division has considered that it was obvious to the skilled person to replace the stratified blend layer of the laminate of D2 by the polyamide-containing film of D1 since these layers were known from D1 to be equivalent. However the Board cannot share this opinion for the following reasons.

Firstly, it derives from the distributions of the resin components in the blend layer illustrated on Figure 1 of D2 and explained at pages 8 and 13 as well as from the comparison of the methods of manufacture of the film of D1 and of the stratified blend layer of D2, that the latter does not contain parallel, substantially two-dimensional and overlapping lamellae of the saponified copolymer embedded in a continuous matrix and adhered together by means of the third component. Therefore the film of D1 differs from the layer of D2 by its structure. Secondly, D1 does not contain any information allowing to consider that the barrier properties of the polyamide-containing film to unleaded gasoline are equivalent to those of the film comprising the copolymer of ethylene and vinyl alcohol. As already pointed out above the former film was tested for permeation resistance to unleaded gasoline while, in the case of the latter film, only the

permeability to oxygen and water vapour was measured. Thus, it cannot be concluded therefrom that these layers have comparable barrier properties. Under these circumstances the stratified blend layer of D2 cannot be regarded as comparable to the polyamide-containing film of D1 and the findings of the Examining Division cannot be followed by the Board.

6.3 In the Board's opinion, the skilled person faced with the problem defined above would at most have speculated in view of this prior teaching that the incorporation of a polyamide-containing layer of the type disclosed in D1 into the laminates of D2 might favourably influence their barrier properties in respect of hydrocarbons such as toluene, provided the thickness of the layer would be sufficient. However as these laminates comprise not only a blend layer but also a polyolefin layer bonded thereto, there exists several alternatives as to the possible arrangement of the polyamide-containing layer in the laminates of D2 without taking account of the problem of thickness layer which would necessarily arise in connection with such an operation. The blend layer, which is considered as compulsory in D2, could be retained and the polyolefin layer replaced by the polyamide-containing layer of D1, conversely the latter could be substituted for the blend layer and the polyolefin layer retained, or both the blend layer and the polyolefin layer could be retained and the layer of D1 bonded to one of them. Therefore the skilled person was certainly not in a "one-way situation" but had to make a selection between several possible combinations in the absence of any guidance as to which of them could possibly lead to a satisfying solution of the present problem.

6.4 According to D2, the laminates make possible to improve the inherent defects of a polyolefin layer, in particular

its high permeability to gases such as oxygen or carbon dioxide, and the inherent defects of a saponified copolymer layer such as its high steam permeability, while retaining the desirable characteristics of each layer (cf. page 15, lines 1 to 5). D2 is silent about the cause of the improved barrier properties to aliphatic and aromatic organic solvents, however as the polyolefin layer is not compulsory in the laminates, the skilled person would not have deduced that it is responsible for this improvement. Although it is mentioned in D2 that polyolefins have frequently been used as packaging materials for foodstuffs because of their excellent mechanical strength (see page 1, lines 11 to 13), the good mechanical properties of the laminates such as their impact resistance, which is of particular importance in the case of bottles, are mainly attributed to the stratified structure of the blend layer and to the presence of the carbonyl group-containing polymer therein (cf. page 15, lines 6 to 11). On the other hand, D1 teaches that a polyethylene film has not only a lower permeation resistance to unleaded gasoline than the polyamide-containing film but also a lower impact strength when used in the form of bottles (cf. page 15, Table 1). It derives from all this that there is no clear teaching in D1 and D2 which would have led the skilled person to consider the polyolefin layer of the laminates of D2 as the appropriate layer to melt-bond with the polyamide-containing film of D1 in order to obtain laminates which exhibit good barrier properties to hydrocarbons and can be produced in form of bottles. Under these circumstances, the Board can only conclude that the claimed laminate was not obvious in view of D1 and D2.

- 6.5 It results from the preceding that the laminate according to Claim 1 of the main request meets the requirement of inventive step.

7. The dependent Claims 2 to 9 which relate to preferred embodiments of Claim 1 derive their patentability from that of Claim 1.

The main request being allowable, it is not necessary to examine the auxiliary request.

**Order**

**For these reasons, it is decided that:**

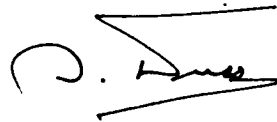
1. The decision under appeal is set aside.
2. The case is remitted to the Examining Division with the order to grant a patent on the basis of Claims 1 to 7 filed on 1 February 1989 and Claims 8 and 9 filed on 9 January 1988, and a description to be adapted.

The Registrar:



P. Martorana

The Chairman:



A.J. Nuss

*John M* 27.3.92

M. Eb