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**D E C I S I O N**  
of 25 March 2004

**Case Number:** T 0405/02 - 3.3.5

**Application Number:** 88104002.6

**Publication Number:** 0332717

**IPC:** C03C 17/36

**Language of the proceedings:** EN

**Title of invention:**  
Transparent laminated product

**Patentee:**  
ASAHI GLASS COMPANY LTD.

**Opponent:**  
SAINT-GOBAIN GLASS FRANCE

**Headword:**  
Laminated product/ASAHI

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

**Keyword:**  
"Inventive step - yes"

**Decisions cited:**  
-

**Catchword:**  
-



Case Number: T 0405/02 - 3.3.5

**D E C I S I O N**  
of the Technical Board of Appeal 3.3.5  
of 25 March 2004

**Appellant:**  
(Proprietor of the patent)

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**Decision under appeal:**

Decision of the Opposition Division of the  
European Patent Office posted 6 March 2002  
revoking European patent No. 0332717 pursuant  
to Article 102(1) EPC.

**Composition of the Board:**

**Chairman:** M. M. Eberhard  
**Members:** E. O. Wäckerlin  
J. H. P. Willems

## Summary of Facts and Submissions

I. European patent No. 332 717 based on application No. 88 104 002.6 and relating to a transparent laminated product was granted to the appellant (proprietor) on the basis of five claims. The respondent (opponent) filed a notice of opposition requesting revocation of the patent on the basis of lack of novelty and lack of inventive step. During the opposition procedure the respondent relied *inter alia* on the following documents in support of his arguments:

- A5: US-A-4 179 181
- A6: GB-A-2 126 256
- A7: US-A-4 725 710
- A8: US-A-4 718 932
- A9: US-A-3 682 528
- A10: FR-A-2 273 777
- A12: EP-A-0 219 273
- A16: P.H. Berning: *Principles of design of architectural coatings*, Applied Optics, vol. 22, No. 24, 1983, pages 4127 to 4141.

With decision dated 17 February 1997 the opposition division revoked the patent on the ground that the subject-matter of the claims lacked an inventive step.

II. The appellant lodged an appeal against this decision. He submitted additional experimental data together with the statement of grounds of appeal. Oral proceedings before the board of appeal were held on 9 May 2000. At the oral proceedings the appellant abandoned the product claims and replaced them by process claims 1 to 3 as the main and sole request.

With decision T 484/97 dated 9 May 2000 the board of appeal set aside the decision of the opposition division and remitted the case to the first instance for further prosecution on the basis of the new process claims 1 to 3.

III. Subsequently the opposition division examined the case on the basis of further amended versions of claim 1.

With decision dated 6 March 2002 the opposition division revoked the patent on the ground of lack of inventive step. The decision was based on the amended claims filed on 23 April 2001. The opposition division observed that document A5 disclosed a multilayered structure on a glass substrate consisting of a first layer of a refractory dielectric material, a second layer of silver having a thickness of 11 - 25 nm, and a third layer of a refractory dielectric. In a preferred embodiment this sequence of layers was doubled to give a total of formally six layers. The dielectric material was selected among various refractory materials including zirconium oxide ( $ZrO_2$ ). It was considered that the replacement of zirconium oxide by zinc oxide ( $ZnO$ ) was obvious having regard to, *inter alia*, document A12 where zinc oxide was disclosed as a preferred material in view of its high sputtering rate. The claimed subject-matter was also considered to be obvious if document A9 was used as the starting point instead of document A5. In A9 an article consisting of a substrate, three dielectric layers, two silver layers and four interlayers was described. The preferred dielectric materials were aluminium oxide ( $Al_2O_3$ ), magnesium fluoride ( $MgF_2$ ) or titanium dioxide ( $TiO_2$ ), but it was stated in A9 that the

dielectric material could be changed without a major change appearing in the spectral performance of the filter. In view of this teaching it was obvious to use zinc oxide which was easier to sputter than, for example, titanium dioxide.

IV. The appellant lodged the present appeal against this decision. In the statement of grounds of appeal he relied on documents A5, A9, A7, A8 and A12. Moreover he submitted X-ray diffraction data relating to the crystallinity of silver layers in three layered assemblies. In a letter dated 17 March 2004 the appellant submitted further experimental data relating to the optical properties of various coated glass substrates, X-ray diffraction data and a moisture resistance test.

Oral proceedings before the board of appeal were held on 25 March 2004. At the oral proceedings the appellant submitted amended claims 1 to 3. Claim 1 reads as follows:

"A process for producing a transparent laminated product consisting of a transparent substrate and a coating consisting of transparent oxide and silver layers alternately laminated in a total of  $(2n + 1)$  ( $n \geq 2$ ) layers with the innermost and outermost layers being transparent oxide layers and having a surface resistance of not higher than  $6 \Omega/\text{sq.}$  and a visible ray transmission of at least 60 %, wherein each transparent oxide layer is a ZnO layer, including the steps of adjusting the thickness of each silver layer within the range of 6 to 12 nm (60 to 120 Å), the thickness of the innermost and outermost transparent oxide layers within

the range of from 20 to 60 nm (200 to 600 Å), and the thickness of the other transparent oxide layers within the range of 40 to 120 nm (400 to 1200 Å) to obtain the desired colour of reflection."

- V. The appellant's written and oral arguments can be summarised as follows:

Starting from document A5 as representing the closest prior art, the technical problem underlying the present invention was to provide a transparent electrically conductive laminated product having an adequately low surface resistance, an adequately high visible ray transmission and a visible ray reflectance at a level of a usual transparent glass sheet, while concurrently avoiding the problem of "island formation" usually encountered when sputtering thin silver layers on a dielectric oxide. The use of zinc oxide as the material for the dielectric layers gives rise to thin silver layers of good quality. The X-ray diffraction pattern shows that the crystallinity of the silver layers in the system Ag/ZnO is unexpectedly higher than in other systems such as Ag/TiO<sub>2</sub>, Ag/SnO<sub>2</sub> or Ag/ZrO<sub>2</sub>. The high crystallinity has a remarkable influence on the electric properties and the durability of the product obtained by the process according to claim 1.

Another important aspect of the invention consists in that the thickness of each layer of ZnO can be varied independently. This allows a wide range of the colour of reflection, in contrast to the 6-layered structure of A5 where the thicknesses of the dielectric layers are restricted by the "doubling" of the basic 3-layered structure.

The problem of poor reproducibility of thin silver layers is addressed in document A9. According to A9 the problem is solved by providing precoat layers of nickel or chromium. There is no teaching in A9 which would lead the skilled person to consider using ZnO as the dielectric material instead of providing Ni or Cr precoats.

The disclosure contained in documents A7, A8 and A12, respectively, does not lead to the invention either. Doubling the 3-layered structures disclosed in each of documents A7, A8 and A12 allows less freedom in the choice of the reflection colour and mainly provides for a violet type reflection only.

VI. The submissions of the respondent made orally and in writing may be summarised as follows:

Example III of document A5 discloses a transparent substrate coated with double period coatings, each period consisting of a thin silver layer having a thickness of 11 nm, and two layers of zirconium oxide having a thickness of 25 nm each. In total this corresponds to a sequence of five layers, viz.  $ZrO_2(25\text{ nm})/Ag(11\text{ nm})/ZrO_2(50\text{ nm})/Ag(11\text{ nm})/ZrO_2(25\text{ nm})$ . Table I of A5 shows that the visible ray transmission of the system is higher than 60%.

Although the surface resistance is not indicated explicitly, a simple calculation leads to the conclusion that it must be below 6  $\Omega/sq$ . According to document A6 the surface resistance of the system  $TiO_2(35\text{ nm}) / Ag(15\text{ nm}) / TiO_2(35\text{ nm})$  was 3.3  $\Omega/sq$ . Since the surface

resistance is inversely proportional to the thickness of the layer, a silver layer of 11 nm must have a surface resistance of 4.5  $\Omega$ /sq. If there are two silver layers of the same thickness, as in Example III of A5, the resulting surface resistance is approximately half the value of a single layer, i.e. 2.25  $\Omega$ /sq. (see page 4 of the respondent's letter dated 4 April 2000).

The product of Example III of A5 corresponds therefore to the product obtained by the process according to the patent in suit, except with regard to the material of the dielectric layers. To replace  $ZrO_2$  by ZnO is obvious in particular in view of the teaching of A12 which discloses that ZnO can be easily deposited.

Concerning the possibility of varying the colour of reflection, claim 1 does not define any specific colour. Furthermore, it was known from A10 and A16 that the colour of reflection could be changed by varying the thickness of the dielectric layers.

The argument according to which the use of ZnO gives rise to a higher crystallinity of the silver layers and thus the avoidance of "island formation" is misconceived. In the application as filed no link between the selection of the dielectric material and the degree of "island formation" was disclosed. Moreover there is no disclosure of a link between "island formation" and crystallinity. According to the patent in suit there exists no problem of "island formation", whenever silver layers of a thickness of at least 6 nm are used (see page 4, lines 20 to 22).



The respondent concludes that the appellant's argumentation regarding crystallinity and "island formation" amounts to a reformulation of the technical problem which cannot be accepted, because the alleged effects are not derivable from the application as filed.

The experimental data provided by the appellant with his letter dated 17 March 2004 are in any case late and must be disregarded.

- VII. The appellant requested that the decision under appeal be set aside and that the patent be maintained on the basis of claims 1 to 3 filed during the oral proceedings.

The respondent requested that the appeal be dismissed.

### Reasons for the Decision

1. The appeal is admissible.
2. *Allowability of the amendments*

There are no objections under Article 123(2) EPC with respect to the amended claims 1 to 3 submitted on 25 March 2004. A basis for these claims can be found in the application as originally filed (see claims and page 11, lines 9 to 11; page 8, lines 16 to 21; page 9, line 7; page 10, line 9; , page 12, line 18; page 8, line 26 to page 9, line 7; page 11, line 25 to page 12, line 3; page 25, lines 6 to 8 and examples).

There are also no objections under Article 123(3) EPC: The amendments made to the claims consist in (a) replacing the product claims as granted by process claims, (b) lowering the surface resistance to values not higher than  $6 \Omega/\text{sq.}$ , (c) selecting ZnO as the transparent oxide, (d) narrowing down the ranges of the thickness of the oxide layers, (e) introducing the step of adjusting the thickness of each silver and oxide layer to obtain the desired colour of reflection, and (f) replacing the term "comprising" by "consisting of". These amendments constitute a limitation of the extent of protection conferred by the patent.

3. *Late filed evidence*

The board has considered the late-filed evidence submitted by the appellant in the letter dated 17 March 2004. In the exercise of its discretion under Article 114(2) EPC the board decided not to allow the late filed comparative tests into the proceedings as they had been submitted only about one week before the oral proceedings and the respondent did not have sufficient time to assess them or to provide counter-experiments. In this context the board observes that the moisture resistance tests themselves require at least 17 days (humidity bath 3 days + samples left in a room for 2 weeks, see page 6 of the comparative tests). Furthermore the respondent expressed doubts as to whether or not the relatively thick layer of Zr (i.e. 17 nm) in samples B and C, respectively, had been completely oxidised, and pointed out that due to the very short time between receipt of the comparative tests and the oral proceedings it had not the opportunity to discuss this point with its own experts.

4. Novelty

4.1 Example III of A5 relates to a transparent infra-red reflecting product consisting of a transparent substrate, viz. polycarbonate, and what is called a "double period coating" consisting of the following sequence of transparent oxide and silver layers:  $ZrO_2$  (having a thickness of 25 nm) / Ag (11 nm) /  $ZrO_2$  (25 nm) /  $ZrO_2$  (25 nm) / Ag (11 nm) /  $ZrO_2$  (25 nm). Formally the number of layers is 6, but in view of the fact that the two inner layers consist of the same material, i.e.  $ZrO_2$ , they may be regarded as being technically equivalent to one single layer of  $ZrO_2$  having a thickness of 50 nm. It cannot be inferred from A5 that these two layers were deposited using different operating conditions, or that the two layers had different properties.

The use of ZnO as the material of the transparent oxide layers is not disclosed in A5.

It was not disputed that the visible ray transmission is more than 60% (see Table I of A5), with values varying between 64% at the wavelength of 380 nm, 74% at 600 nm, 67% at 700 nm, and 56% at 750 nm.

The surface resistance of the laminated product is not specified in A5. Moreover A5 is silent on any steps which have to be taken in order to obtain a specific reflection colour.

In view of the distinguishing features set out above the process according to claim 1 of the patent in suit is novel with regard to the disclosure of A5.

4.2 The respondent's argument according to which it was clear that in the case of Example III of A5 the surface resistance was as low as 2.25  $\Omega$ /sq., cannot be accepted. In fact even if the basic assumptions made by the respondent, namely (i) that the relationship between the surface resistance and the thickness of the silver layer is inversely proportional, and (ii) that two silver layers of identical thickness have half the surface resistance of one of these layers alone, appear to be plausible, the respondent admitted that they are only an approximation (see, in this respect, Example 2 and comparative Examples 1 and 2, which do not fully confirm the basic assumptions). Furthermore no evidence has been presented that the surface resistance data contained in A6, which are based on the system  $\text{TiO}_2 / \text{Ag} / \text{TiO}_2$ , are representative for Example III of A5, where a different dielectric material (i.e.  $\text{ZrO}_2$ ) and a different method of deposition were used. In this respect the board observes that the burden of proof lies with the respondent for its allegation concerning the surface resistance in Example III of A5. In the absence of sufficient evidence, the argument brought forward by the respondent must fail.

4.3 The claimed process is also novel over the teaching of the other cited prior art. This was not in dispute in the appeal proceedings, so that further considerations in this respect are not necessary.

5. *Inventive step*

5.1 In agreement with the position taken by the opposition division and the parties, the board considers that A5

represents the closest prior art. The transparent interference-coated articles of A5 reflect infrared light and selectively transmit visible light. A sharp cut-off between reflected and transmitted light is obtained by using double period interference coatings. The coated article of Example III exhibits a high transmittance in the visible region and a relatively low visible ray reflectance (see abstract, column 1, lines 35 to 44; column 2, lines 24 to 28; Figure 2; Table I, Example III).

With respect to A5 the technical problem underlying the claimed process can be seen in providing a process for producing a transparent laminated product having a low surface resistance, which makes it possible to vary the colour of reflection fairly freely, while maintaining a high visible ray transmission, a high reflectance in the infrared region, a sharp rising of the reflectance in the near infrared region and a visible ray reflectance at the level of a normal glass sheet.

- 5.2 It is proposed to solve this problem by the combination of features defined in claim 1, which differs from A5 in particular by using zinc oxide (ZnO) as the material of the transparent oxide layers, and by the step of adjusting the thicknesses of each of the silver layers and the oxide layers within the ranges stated in claim 1 to obtain a specific colour of reflection within a broad range of different colours. In view of the results reported in the examples of the patent in suit (see, in particular, Examples 1 to 4, 9, and reference Examples 6, 7), it is credible that the problem indicated above has actually been solved by the method defined in the claims.

- 5.3 Document A12 discloses the use of zinc oxide (ZnO) as a transparent oxide layer in transparent products including dielectric-metal-dielectric layers suitable for windshields or other applications. These products have a high transmission in the visible region and a low emissivity (see e.g. claims 6 and 11; page 7, Table 3; page 9, line 19 to page 10, line 3; page 2, lines 22 to 26; Figure 1). The surface resistance of the coated layers is less than 5  $\Omega$ /sq. (see page 3, lines 21 to 22; claim 8). According to page 3, lines 15 to 17 of A12 "the intermediate index material is preferably zinc oxide because this material can be relatively quickly and inexpensively deposited by reactive sputtering".
- 5.4 The respondent argued that in view of this disclosure it was obvious to a skilled person to combine the teachings of A5 and A12, i.e. to substitute  $ZrO_2$  used in Example III of A5 by ZnO.
- 5.5 The board is not convinced by this argument for the following reasons:
- According to A12 ZnO is used as an "intermediate index material" in combination with  $TiO_2$ , not as a single dielectric material (see e.g. page 3, lines 5 to 15; page 4, lines 6 to 22). Thus the ZnO layer does not simply act as a substitute for the  $TiO_2$  layer in  $TiO_2$  / Ag /  $TiO_2$  coatings referred to on page 2, lines 10 to 12.
  - According to A12 the  $TiO_2$  layer deposited between the silver layer and the ZnO layer is mandatory. A12

teaches that the metallic layer (14) has a thickness of 10 to 50 nm. However, a silver layer having a thickness of 15 to 20 nm is preferred (see page 4, lines 13 to 15; claim 4), i.e. a thickness lying outside the claimed range. The preferred embodiment has a surface resistance of 4 to 5  $\Omega$ /sq. In the examples of A12 a thickness of 18 nm is used for the silver layer in combination with one or two thin TiO<sub>2</sub> layers and two ZnO layers. A12 contains no information suggesting that, by using ZnO instead of ZrO<sub>2</sub> as the dielectric material in combination with a silver layer thinner than the preferred range stated in A12, while omitting the mandatory TiO<sub>2</sub> layer, it would still be possible to maintain the desired optical properties obtained in A5 for a layer stack comprising two silver layers.

- It can also not be inferred from A12 that the use of a thinner silver layer in combination with a ZnO dielectric layer and without the TiO<sub>2</sub> layer would result in the desired low electrical surface resistance.
- Furthermore neither A5 nor A12 deals with the problem of how to obtain a wide range of reflection colours while maintaining the other desired optical and electrical properties.
- It is further observed that the technical advantage disclosed in A12 regarding the sputtering rate of ZnO is only achieved if reactive sputtering is used as the coating method. A5 does not disclose that reactive sputtering is a suitable method for

preparing the 6- layered structure of Example III having the optical properties stated in A5.

It follows from the above that in view of the teaching of A12 the skilled person would not have been prompted to replace the  $ZrO_2$  dielectric layers of Example III of A5 by ZnO layers while omitting the  $TiO_2$  layer considered as mandatory in A12, since he could not expect these measures to result in a multilayered structure having the desired combination of optical and electrical properties. In the board's judgment the respondent's arguments appear, therefore, to be based on an *ex post facto* analysis of the case.

Therefore the board holds that it cannot be derived from A12 that the technical problem stated above can be solved by using ZnO instead of  $ZrO_2$  as the transparent oxide in the interference coating of Example III of A5.

- 5.6 As far as the variation of the reflection colour is concerned, document A10 teaches that the colour "can be modified by using a supplementary dielectric layer having a thickness which corresponds to the desired colour" (see claims 8, 14 and 17; page 2, lines 23 to 25; page 8, lines 16 to 19). The additional layer consisting of a transparent oxide forms the outermost or innermost surface of the coated substrate (see Figure 2, reference number 31; Figures 4 and 5, reference number 77; page 8, lines 16 to 24; page 17, lines 1 to 4). ZnO is not mentioned. A10 states that "the whole range of colours" can be obtained by adjusting the thickness of this outer layer (see page 9, lines 3 to 10; page 13, lines 26 to 35). According to the examples the "colouration layer" 31 of silicium



oxide is relatively thick, namely 180 nm and 360 nm, respectively (see page 11, Table III; page 12, Table IV).

5.7 Although A10 discloses the possibility of varying the colour of reflection by adjusting the thickness of a so called "colouration layer", the concept differs from the present method according to which the reflection colour can be varied by adjusting the thickness of each separate layer of the product within the narrow ranges indicated in claim 1. Moreover neither silver nor ZnO are mentioned in A10 as suitable materials for the metallic layer and the dielectric layers, respectively. Therefore this document cannot give the skilled person an incentive to replace the ZrO<sub>2</sub> layers in the product according to Example III of A5 by ZnO layers in order to solve the technical problem stated above.

5.8 Document A16 deals also with the question of colour variation through the use of optical interference (see pages 4138 to 4140). It teaches that a great variety of reflection colours can be produced in multilayer metal/dielectric coatings by utilizing appropriate choices of component materials along with a broad range of dielectric layer thickness (see page 4139, left hand column, lines 10 to 16). Figure 27 shows a model calculation for the system Ni (thickness 4 nm) / dielectric material having a refraction index of 2.30 (of variable thickness from about 30 to 359 nm) / Ni (18 nm). On the basis of the resulting curve the conclusion is drawn that all dominant wavelengths of colour can be realised. However it can also be derived from A16 that this conclusion is only theoretical, and that practically speaking the colour production by

interference means is limited in the light of the current technology (see page 4139, left hand column, lines 23 to 27; page 4140, left hand column, lines 23 to 27). A16 is mainly concerned with theoretical considerations and contains no reference to specific oxides which may be used as transparent oxide layers. As in the case of A10, ZnO is not mentioned.

- 5.9 On the basis of the evidence outlined above the board concludes that none of the documents A12, A10 and A16 provided the skilled person with an incentive to use ZnO as the transparent oxide in a multilayered coating as disclosed in Example III of A5, and to adjust the thickness of each separate layer within the ranges set out in claim 1 in order to solve the technical problem stated above. Therefore the process according to claim 1 involves an inventive step.
- 5.10 The same conclusion is reached if A9 is taken as the starting point of the analysis of inventive step. A9 discloses a transparent infra-red reflecting interference filter comprising typically two silver layers and three layers of a dielectric material (see Figure 3), for example  $\text{Al}_2\text{O}_3$ ,  $\text{MgF}_2$  or  $\text{TiO}_2$  (see column 4, lines 1 to 2). A9 is silent on the surface resistance of the product. According to the example outlined in column 6, lines 18 to 33 the thickness of each silver layer is 7 nm, whereas the three dielectric layers have a thickness of 50 nm, 114 nm and 50 nm, respectively. Thus the thicknesses of the silver layers and dielectric layers fall within the claimed ranges, but ZnO is not disclosed as a possible material for the dielectric layers, as in the case of Example III of A5. Generally speaking the preceding considerations

concerning the combination of the teaching of A5 with the disclosure of A10, A12 and A16 apply in an analogous manner when A9 is taken as the closest prior art. Nevertheless the teaching of A9 includes a further element, namely the need to use very thin precoat layers of a metal such as Ni or Cr, or a suitable dielectric material as a "nucleation layer" which enables the fabrication of the thin silver layers (see column 2, line 60 to column 3, line 7; column 3, lines 40 to 43; column 4, lines 31 to 38). The process according to claim 1 of the patent in suit does not use intermediate layers. In this respect the board notes that the wording of claim 1 with the term "consisting of" clearly excludes the presence of intermediate layers.

- 5.11 The respondent's affirmation that in view of the high sputtering rate the skilled person would have automatically selected ZnO as the transparent oxide, irrespective of any other considerations, has been contested by the appellant. The board is not convinced by the argument either. Although it cannot be denied that the sputtering rate is indeed an important factor whenever sputtering methods are used for depositing the dielectric layers, there are other technical factors of at least equal importance which must also be taken into consideration, in particular the need to obtain the combination of desired optical and electrical properties. It is in particular essential to use a dielectric material with a suitable refraction index. In this respect the board observes that the statement in A9, column 4, lines 39 to 42, according to which the dielectric material utilized in the filter can be changed without a major change appearing in the

spectral performance of the filter is somewhat misleading. On the one hand it is explained in A9 that the refractive index does, in fact, have an impact on the transmission passband and the cut-off (see column 4, lines 43 to 46), and on the other hand no attention is paid in A9 to the reflection colour.

- 5.12 In view of the foregoing the board cannot accept the respondent's conclusion that the skilled person would have automatically selected ZnO because of its high sputtering rate.
- 5.13 The other documents cited during the opposition and the appeal proceedings do not contain any additional information which could hint at the claimed solution, when considered alone or in combination with the teachings of A5, A9, A10, A12 or A16.
- 5.14 It follows from the above that the subject-matter of claim 1 meets the requirement of inventive step laid down in Articles 52(1) and 56 EPC. The same applies to the dependent claims 2 and 3. Therefore claims 1 to 3 as submitted during the oral proceedings on 25 March 2004 are allowable.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The case is remitted to the opposition division with the order to maintain the patent with claims 1 to 3 submitted during the oral proceedings on 25 March 2004, the drawings as granted and the description to be adapted.

The Registrar:



A. Wallrodt

The Chairman:



M. M. Eberhard

