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D E C I S I O N
of 14 December 1993

Case Number: T 0380/91 - 3.3.2

Application Number: 83305528.8

Publication Number: 0104870

IPC: C03C 17/36

Language of the proceedings: EN

Title of invention:

Low emissivity coatings on transparent substrates

Patentee:

Pilkington Plc

Opponent:

- 01) Leybold Aktiengesellschaft
- 02) Vegla Vereinigte Glaswerke GmbH
- 03) Interpane Entwicklungs- und Beratungsgesellschaft mbH & Co.

Headword:

Low emissivity coatings/PILKINGTON

Relevant legal norms:

EPC Art. 54, 56

Keyword:

"Novelty (yes) - no unambiguous disclosure in the prior art"
"Inventive step (no) - obvious alternative process"

Decisions cited:

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Catchword:

-

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Decision under appeal: Decision of the Opposition Division of the
European Patent Office dated 25 October 1990,
posted on 12 March 1991 revoking European patent
No. 0 104 870 pursuant to Article 102(1) EPC.

Composition of the Board:

Chairman: P.A.M. Lançon
Members: I.A. Holliday
S.C. Perryman



Case Number: T 0380/91 - 3.3.2

D E C I S I O N
of the Technical Board of Appeal 3.3.2
of 14 December 1993

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Summary of Facts and Submissions

- I. European patent No. 0 104 870 was granted on the basis of 24 claims contained in European patent application No. 83 305 528.8. Claim 1 as granted reads as follows:
- "A process for the production of a low emissivity coating on a transparent substrate of glass or plastics material by cathode sputtering comprising, in sequence,
- (i) sputtering a layer of silver from 5 to 30 nm thick onto the transparent glass or plastics substrate
 - (ii) sputtering an additional metal or metals other than silver in an amount equivalent to a layer 0.5 to 10 nm thick onto the silver and
 - (iii) reactively sputtering, in the presence of oxygen or an oxidising gas, an anti-reflection metal oxide layer or layers over the silver and additional metal, under conditions which, in the absence of the additional metal or metals, would lead to substantial loss of the low emissivity properties of the resultant product."
- II. Three oppositions were filed against the granted patent. Of the numerous documents cited, the following remain relevant to the present decision:
- (1) EP-A-0 035 906
 - (2) US-A-4 336 119
 - (3) EP-A-0 024 925
 - (4) Münz et al., *Moderne Beschichtungstechnologien von Architekturglas*, Vacuum-Technik, 1981, 8, 236 to 246.
- III. The Opposition Division revoked the patent on the grounds of lack of inventive step. The closest prior art

was considered to be document (1) since the process thereof involved the use of an additional metal layer when applying an oxide coating to a metallic silver layer. However, the process of (1) did not involve reactive sputtering. The Opposition Division considered that document (2), which was concerned with essentially the same layered structure except for the additional metallic layer, would have led the skilled person to use reactive sputtering.

The Opposition Division considered that the product Claims 14 to 24 also lacked inventive step in the light of the disclosure of (1).

- IV. An appeal was lodged against the decision of the Examining Division. Oral proceedings took place on 14 December 1991.

- V. In the Statement of Grounds of appeal, the Appellant announced that the product claims would not be pursued. At the oral proceedings, the Appellant admitted that the products obtained by the process of the patent in suit could only derive novelty by their method of preparation and were in fact the same as the products of document (1).

The Appellant argued that the correct starting point should be document (2) since this related to a process of reactive sputtering and involved a similar problem, namely the degradation of a silver layer when subjected to reactive sputtering. Document (1), on the other hand, was concerned with the long term stability of the multi-layer coating. However, starting from (1), there are a number of alternative methods available for preparing the layered products therein disclosed, e.g. wet deposition and sputtering from oxide sources. Document (1) itself has no example involving reactive sputtering.

Quoting the text-book Thin Film Phenomena by Chopra and a paper by Münz presented at the IPAT Conference in February 1984, the Appellant argued that the skilled person would choose conditions under which the silver layer would not be destroyed.

The oxygen partial pressures of the oxygen/argon mixtures used according to the patent in suit were an order of magnitude greater than those employed in document (2) and in DE-A-3 027 256 which has also been referred to. Contrary to the assertions of Respondent (01), the Appellant assured the Board at the oral proceedings that the process of the patent in suit can be operated with pure oxygen as well as oxygen/argon mixtures. It is particularly to be noted that the process of document (1) employs very low oxygen pressures which accounted for the insignificant change in emissivity even in the absence of a barrier layer. In no way could the ion plating process of document (1) be considered equivalent to the vigorous conditions employed according to the patent in suit.

In rebutting the novelty objection *vis-à-vis* document (3), elaborated by Respondent (01) at the oral proceedings, the Appellant argued that many selections would have to be made from the disclosure of (3) in order to arrive at the subject-matter of the patent in suit.

In response to a question by the Board, the Appellant indicated that no prejudice would have existed at the earlier priority date of the patent in suit against reactive sputtering on a gold layer.

The Statement of Grounds of appeal was accompanied by a declaration of Dr R.P. Hawson of the University of

Technology, Loughborough, together with a number of literature references.

According to the Appellant's main request, maintenance of the patent was requested on the basis of a main claim filed on 12 July 1991, which corresponded to Claim 1 as granted. According to the first auxiliary request, also filed on 12 July 1991, the oxide layer under (iii) was restricted to an oxide of indium, tin and/or zinc. The second auxiliary request, submitted at the oral proceedings, restricted the said oxide layer to an oxide of indium, tin or zinc.

VI. The arguments of the Respondents, both in the written procedure and during the oral proceedings, may be summarised as follows.

Respondent (01) endorsed the decision of the Opposition Division arguing that there was an analogy between the long term ageing of the silver layer which was the aim of document (1) and the effects of harsh conditions such as those encountered in the reactive sputtering process of the patent in suit. The Respondent doubted whether the process of the patent in suit would work with oxygen alone, arguing that the presence of the heavier argon molecule was necessary to initiate reactive sputtering.

Starting from document (2), the Respondent argued that it would be an obvious modification of a continuous gas coating plant to introduce a further module for sputtering of a metallic barrier layer (cf. document (4)).

The Respondent developed a novelty objection based on document (3). This document, although introduced into the opposition proceedings by Respondent (03), was only discussed shortly before and during the oral

proceedings. Respondent (01) pointed to a specific reference to reactive sputtering in (3) and further argued that the disclosure of (3) included the preparation of layered structures falling within the ambit of Claim 1 of the patent in suit.

Respondent (02) generally supported the arguments outlined above and mentioned at the oral proceedings that it was only Claim 1 of the patent in suit which was opposed, not the specific examples.

The Respondent argued that document (1) was not only concerned with long term stability but also referred to resisting degradation by heat. It was also pointed out that the long term stability must obviously depend upon the individual layers retaining their integrity during coating.

The arguments of Respondent (02) were supported by declarations from Prof S. Schiller of Dresden and Mr François of the St Gobain company together with copious references to the literature.

Respondent (03) also supported the arguments of the other respondents specifically mentioning that reactive sputtering was a well known process at the priority dates, pointing to the disclosure of document (3). The Respondent also argued that not only did document (3) destroy the novelty of the patent in suit but the same also applied to document (1).

VII. The Appellant requested that the decision under appeal be set aside and that the patent be maintained on the basis of the main request or first auxiliary request filed on 12 July 1991 or of the second auxiliary request submitted at the oral proceedings on 14 December 1993.

The Respondents requested that the appeal be dismissed.

Reasons for the Decision

1. The appeal is admissible.
2. Claim 1 according to the main request corresponds to the granted form of the claim. The functional limitation, which was not present in the originally filed claim, i.e. specifying conditions "which, in the absence of the additional metal or metals, would lead to substantial loss of the low emissivity properties of the resultant product" finds support on page 3, lines 1 to 4 of the printed patent specification (page 4, lines 13 to 18 of the original disclosure).
 - 2.1 Restriction of the metal oxide coating to indium, tin or zinc oxide (second auxiliary request) is supported by the paragraph from line 52 to line 61 on page 2 of the printed patent (page 7, line 22 to page 8, line 14 of the original disclosure).
 - 2.2 The final sentence of the same paragraph provides a basis for the use of more than one metal oxide layer (and/or) according to the first auxiliary request.
 - 2.3 The requirements of Articles 123(2) and 123(3) are accordingly satisfied.
3. Having regard to the objections of the Respondents, the novelty of Claim 1 of the patent in suit *vis-à-vis* documents (1) and (3) must be considered.
 - 3.1 Since document (3) contains on page 10, lines 18 to 22 a specific reference to reactive sputtering, its

disclosure will be considered first. Document (3) relates to coatings, on a plastics or glass substrate (A) of a transparent thin layer (B) having a high refractive index in contact with one or both surfaces of a transparent heat-wave reflective thin layer of an electroconductive metal (C). According to Claim 1 of (3), the metal layer (C) must contain, by weight, at least 50% Ag, 3 to 30% Au and 0.5 to 30% Cu, i.e. Au must always be present. The layer (B) may be an oxide, e.g. of Ti, Bi or Zn.

- 3.1.1 In the vast majority of the worked examples of (3), a TiO_2 layer is deposited by a wet coating method, e.g. from a solution of tetrabutyltitanate. In one example (37), TiO_2 is applied by sputtering using a TiO_2 target. This is clearly not a reactive sputtering process in which either oxygen or an oxygen/argon mixture would be directed towards a metallic titanium target. The metal layer (C) is usually applied by sputtering but since this relates to deposition of metal, reactive sputtering in the presence of oxygen is not involved.

- 3.1.2 The only worked examples of (3) in which two distinct layers of metal are applied is Example 38. In this case a layer of TiO_2 is first applied by wet coating to a plastics substrate followed by a coating of Ag/Au (85/15 by weight) applied by sputtering. A copper layer is next applied from a solution of cupric acetylactonate. Finally, a further TiO_2 layer is applied by wet coating. Thus, although the layers are superficially similar to the product of the process of the patent in suit, the method of preparation is quite different. The same reasoning applies to Examples 11 to 13 in which three distinct layers Au, Ag, Cu are applied by vacuum deposition.

3.1.3 During the oral proceedings, Respondent (01) presented a diagram of a layered structure based on the disclosure of (3) showing composite metal layers (C) sandwiched between two oxide layers B and B', the whole being coated on a substrate. The structure could correspond to Example 38. Although the description of (3) (page 10, lines 21 to 22) does mention the possibility of applying a TiO₂ coating by reactive sputtering, there is not a single worked example in which such a method is used. Moreover, there is also no specific disclosure of applying a **silver** layer by sputtering, each example, having regard to the requirements of Claim 1 of (3), being a composite layer of Ag, Au and Cu. Furthermore, although the said composite layers are applied by sputtering, there is no worked example in which a further metal layer is applied by sputtering (cf. Example 38 noted above).

3.1.4 Although the various features which define the process of Claim 1 of the patent in suit may be found within the broad disclosure of document (3), it is not permissible to combine them unless the document itself suggests such a combination (cf. T 305/87, OJ EPO 1991, 429; Reasons, point 5.3). This is clearly not the case. Novelty over the disclosure of (3) must accordingly be recognised.

3.2 In arguing that document (1) related to a process involving reactive sputtering, the Respondent (03) relied on a cross-reference in (1) on page 2, line 9 to document (3). Document (1) itself refers merely to "cathode sputtering" as a method for applying a barrier layer (C). Certain worked examples of (1) employ a low temperature sputtering with a TiO₂ target. Other examples are based on solution coating. Thus, although the products of (1) may be the same as those of the patent in suit, there is no disclosure in (1) of the various process features of the patent in suit in

combination. Novelty over the disclosure of (1) must also be recognised.

4. The patent in suit concerns a process for the production of a transparent low emissivity coating on a glass or plastics substrate. At first glance, it might seem that the problem to be solved would be to modify or improve glass or plastics substrates with low emissivity coatings as known in the prior art e.g. as described in documents (2) and (4).

4.1 However, having regard to the Appellant's admission at the beginning of the oral proceedings that the products obtained according to the process of the patent in suit are in fact the same as those described in document (1), it is the Board's opinion that document (1) must be considered to be the closest prior art.

4.1.1 Document (1) relates to a selectively light transmitting laminated structure composed of: (A) a transparent layer, e.g. of plastics (page 6, line 27 to page 7, line 3) or glass (page 7, lines 4 to 7); (D) an infrared reflective layer of thickness 500 to 3000 nm comprising metallic silver; a transparent thin layer (B_1) having a high refractive index between layers (A) and (D) or a similar high refractive index layer (B_2) on layer (D); the layers (B_1) and (B_2) may be oxides of Ti, In, Zn or Sn (Claim 3); the structure of document (1) is characterised by the presence of a barrier layer (C) having a thickness of 30 to 1000 nm, which consists of Ti, Zr, In, Si, C, Co or Ni, interposed between layers (D) and (B_2), i.e. on the opposite side of layer (D) in relation to (A).

4.1.2 In the worked examples of (1), the layer (D) may be of pure Ag; other examples use alloys of Ag and Cu and Ag and Au. The second metallic layer (C) or barrier layer

is formed from Ti, Ni or Co as well as C and Si in the worked examples. Layer (B₂), i.e. the metal oxide layer applied on top of the barrier layer (C) is usually TiO₂. Examples relate to its application by coating from solution, e.g. from tetrabutyl titanate in isopropyl alcohol, by low temperature sputtering from a TiO₂ target and also by ion plating using a high frequency power source.

4.2 In the light of the admitted lack of novelty of the products obtained by the process of the patent in suit, the problem to be solved can be seen as the provision of an alternative process for preparing known multi-layer products.

4.3 The problem is solved by using reactive sputtering to apply the metal oxide anti-reflection layer. Having regard to the examples of the patent in suit, the Board is satisfied that the problem has been solved in a plausible manner.

5. It remains to investigate whether or not Claim 1 of the main request satisfies the requirements of Article 56 EPC in respect of inventive step.

5.1 The Appellant has argued that document (1) relates to improving the long term storage properties whilst the patent in suit relates to the stability of a thin silver reflective layer during the deposition of an anti-reflection metal oxide layer. In the opinion of the Board, the two effects must be related. On the one hand, such a layered product could not be regarded as having been made at all if the layers broke down during the actual coating process. On the other hand, such a product which retained its structural integrity during coating would be useless for window glass if it did not possess long term stability.

- 5.2 The essential difference between the process of the patent in suit and that of document (1) lies in the use of reactive sputtering instead of the methods outlined above when applying a metal oxide layer to a coated silver layer. Apart from the reference to the European application number corresponding to document (3) on page 2 of (1), there is no hint in (1) itself which might have given an incentive to the skilled person to investigate reactive sputtering.
- 5.3 Document (3), however, gives a clear reference to using reactive sputtering employing a titanium target (page 10, lines 21 to 22). At the foot of page 5, document (3) refers to the Tamman law, according to which a bulk alloy containing at least 50 atomic% of a noble metal together with a less noble metal has chemical resistance which is almost the same as that of the original noble metal. Contrary to expectations, the coatings according to (3) have similar properties of chemical resistance to those of gold (page 6, lines 24 to 34). At the oral proceedings before the Board, the Appellant acknowledged that at the earlier priority date of the patent in suit there would have been no prejudice against using reactive sputtering when applying an oxide coating to a gold layer. Since the layers of (3) have similar chemical resistance to gold, the same argument must apply. It is to be noted that document (3) is not restricted to alloys of Ag, Au and Cu but that separate layers of the said metals may be applied and similar chemical resistance obtained, cf. Examples 11 to 13 and 38. As pointed out by Respondent (01) at the oral proceedings such a combination of layers fulfils the requirements of Claim 1 of the patent in suit. Accordingly, the skilled person would have had an incentive to use reactive sputtering when applying a metallic oxide to a silver layer overcoated with copper and gold (Example 12 of (3)).

- 5.4 The multi-layer structures known from both the patent in suit and from document (1) are analogous to the layers noted above, especially that of Example 12 of (3). It is to be noted that both Au and Cu are amongst the preferred metals according to the patent in suit for overcoating the Ag layer (cf. page 3, lines 35 to 40 and 45 to 47). If, as acknowledged above, the final coated products known from (1) correspond to those of the patent in suit, it must follow that the substrate/oxide/silver/other metal structure prior to applying the final oxide layer must also correspond. Accordingly, the same incentive to use reactive sputtering for the application of the oxide layer must also apply to the structures known from (1).
- 5.5 Document (2) also provides an incentive to use reactive sputtering. The Appellant has argued that the oxygen pressures (or partial pressures) used in the patent in suit are an order of magnitude higher than those used in the reactive sputtering process of document (2), quoting, for example, the value of 2.5×10^{-3} torr in Example 1 of the patent in suit and comparing it with a value of 1.5×10^{-4} torr for the oxygen partial pressure calculated from Example 1 of (2). It is, however, to be noted that document (2) in column 5, lines 60 to 66, although preferring partial pressures of less than 10^{-4} torr, mentions as possible, values less than about 10^{-1} torr, thus including the values referred to in the patent in suit. It is thus the Board's view that the expression "under conditions which, in the absence of the additional metal or metals, would lead to substantial loss of the low emissivity properties of the resultant product" is both insufficient to define the vigorous conditions used in the patent in suit and insufficient to distinguish them from the reactive sputtering conditions known from the prior art, especially from document (2).

- 5.6 The Board can only conclude that reactive sputtering was a well known process at the earlier priority date and that its use represents an obvious alternative to the procedures known from the prior art. Accordingly, Claim 1 of the patent in suit (main request) lacks the required inventive step (Article 56 EPC).
6. The Board notes that the oxides of indium, tin and zinc, referred to in the auxiliary requests were well known materials for use in antireflective layers (cf. (1), page 9, lines 25 to 34; (2) column 2, lines 4 to 9 and column 10, lines 32 to 49, and (3), page 10, lines 1 to 4). The restriction to these oxides is the only difference between Claim 1 of the main request, and the respective Claims 1 of the auxiliary requests. Such a restriction leaves the reasoning above set out in relation to Claim 1 of the main request equally applicable to the Claim 1 of each of the auxiliary requests, which claims thus also lacks the required inventive step.
7. As each of the requests put forward by the Appellant contains a claim which fails to comply with the patentability requirements of the EPC, the appeal must be dismissed.

Order

For these reasons, it is decided that:

The appeal is dismissed.

The Registrar:

The Chairman:

P. Martorana

P.A.M. Lançon