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D E C I S I O N
of 17 March 1998

Case Number: T 0858/94 - 3.3.2

Application Number: 89118002.8

Publication Number: 0363747

IPC: C03C 17/34

Language of the proceedings: EN

Title of invention:
Low reflectance, highly saturated colored coating for monolithic glazing

Applicant:
PPG Industries, Inc.

Opponent:
-

Headword:
Glazing/PPG

Relevant legal provisions:
EPC Art. 56

Keyword:
"Inventive step (yes, after amendment of the claims)"

Decisions cited:
-

Catchword:
-

Case Number: T 0858/94 - 3.3.2

DECISION
of the Technical Board of Appeal 3.3.2
of 17 March 1998

Appellant:
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Representative:
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Decision under appeal: **Decision of the Examining Division of the European Patent Office posted 8 July 1994 refusing European patent application No. 89 118 002.8 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: P. A. M. Lançon
Members: M. M. Eberhard
S. C. Perryman

Summary of Facts and Submissions

I. European patent application No. 89 118 002.8 was refused by the Examining Division. The decision was based on the amended claims 1 to 21 filed with the Appellant's letter of 3 December 1993.

II. The grounds for the refusal were that the subject-matter of claims 1 and 11 lacked an inventive step having regard to the teaching of D1 or D4 in combination with D2, where D1, D2 and D4 are the following citations:

D1: 4ème Colloque international sur les plasmas et la pulvérisation cathodique, Nice, 13-17 September 1982, "High rate sputtering for production of modern architectural glass", S.R. Reineck et al., pages 385 to 395;
D2: EP-A-0 275 474;
D4: GB-A-2 138 026.

The Examining Division held that D1 and D4 represented the closest prior art and disclosed the principle of the present application, ie the variation of the reflected colour by varying the thickness of the metal oxide layer. The claimed subject matter differed from D4 only by using a layer of tin/zinc alloy oxide as antireflective layer. However, as mixed oxide layers and their advantages were well known in the art, e.g. from D2, the skilled person would have used them in a multilayer coating of the type disclosed in D1. In view of the close chemical resemblance of a pure tin oxide coating and a layer of tin/zinc alloy oxide, this substitution would not have been expected to negatively affect the development of the interference colours. The Examining Division considered that the terms "intense saturated colour" were vague. It did not accept the

Appellant's arguments that the use of a layer of Sn/Zn alloy oxide resulted in higher colour saturation, as there was no evidence showing this effect with respect to D4.

III. The Appellant lodged an appeal against this decision. In a first communication from the Board, reference was made to EP-A-0 183 052 (hereinafter D5) cited in the search report.

Oral proceedings were held on 17 March 1998. At the hearing the allowability of the amendments in the claims filed on 29 January 1998 was first discussed. The Board confirmed the preliminary opinion expressed in its communications that the ranges of chromaticity coordinates incorporated into the claims contravened the requirements of Article 123(2). The Appellant abandoned this set of claims and submitted amended claims 1 to 5 at the oral proceedings as sole request. Independent claims 1 and 3 of this request read as follows:

"1. Low transmittance articles of manufacture for the reflectance of solar energy comprising:

a. a glass substrate,
b. a reflective transparent chromium nitride film deposited onto said glass substrate,
c. a transparent zinc/tin oxide film whereby the proportions of zinc and tin of the cathode used to deposit the film by sputtering are between 30 and 70 percent tin and the balance zinc in a thickness sufficient for the article to exhibit interference colors; and
d. a second reflective transparent chromium nitride film deposited onto said zinc/tin oxide film, resulting in one of the colors with chromaticity coordinates

Y	x	y	Color
9.28	.2325	.2460	deep blue
13.7	.2437	.2633	pale blue
10.7	.2246	.2397	blue
9.2	.2236	.2342	deep blue
10.5	.2356	.2464	blue
13.6	.2592	.2745	pale blue
11.2	.2261	.2459	green-blue
7.5	.2259	.2260	red-blue
6.5	.2325	.2249	red-blue
5.9	.3339	.3065	purple-pink
11.7	.3714	.3766	orange-yellow
9.2	.3802	.3788	dark orange-yellow."

"3. A method of making solar energy reflecting coated articles of low transmittance comprising the steps of:

a. sputtering onto a surface of a glass substrate a reflective transparent chromium nitride film, and

b. sputtering onto the chromium nitride film a transparent zinc/tin oxide coating, whereby the proportions of zinc and tin of the cathode are between 30 and 70 percent tin and the balance zinc in such a thickness that the article exhibits color by interference effects, and

c. sputtering over said zinc/tin oxide film a second reflective transparent chromium nitride film, resulting in one of the colors with chromaticity coordinates ... (see the coordinates and colors stated in claim 1)."

IV. The Appellant's arguments in support of the inventive step of the claimed subject-matter can be summarised as follows:

The Appellant argued that D4 represented the closest prior art. Starting from D4, the technical problem to be solved by the present invention was to provide a low transmittance coated article for reflecting solar energy which exhibited selected reflected colours. The inclusion of the chromium nitride layer between the dielectric layer and the glass substrate allowed a more appropriate adjustment of the interference colours. D2 and D5 disclosed articles having a high transmittance in the visible range and a low luminous reflectance, the coatings being preferably essentially colourless. The claimed combination of layers was not derivable from these documents, nor could it be inferred therefrom that this combination would have led to the specific reflected colours defined in claims 1 and 3. It was common general knowledge that mixed oxides had a structure differing from that of pure oxides. Furthermore, the use of primer layers in D2 showed that the adhesion of the tin/zinc alloy oxide on a surface might be critical. The transfer of the teaching from D2 to D4 or D1 was based on a hindsight approach ignoring essential parts of these documents and their different objects.

V. The Appellant requested that the decision under appeal be set aside, that a patent be granted on the basis of the set of claims submitted at the oral proceedings on 17 March 1998, and that the case be remitted to the first instance for adaptation of the description.

Reasons for the Decision

1. The appeal is admissible.

2. The amended claims 1 to 5 filed at the oral proceedings meet the requirements of Article 123(2) EPC. Claim 1 is based on the combination of the features disclosed in claims 1, 2, 5, 6 and 9 as filed with additional features indicated in the original description. In particular, the proportions of zinc and tin of the

cathode used to deposit the tin/zinc oxide film by sputtering is disclosed on page 7, lines 8 to 11 of the description. The chromaticity coordinates and colours recited in claims 1 and 3 are supported by examples I to XII. In view of the transmittance values indicated in these examples (17 to 22%) and of the light transmittance of 20% mentioned on page 5, lines 7 to 21, it is also directly derivable from the original description that the coated article has a low transmittance. The process claim 3 is based on the combination of original claims 11 and 17 and on the passages of the original description indicated above as regards the proportions of tin and zinc, the chromaticity coordinates, colours and low transmittance. The sequence of deposition of the chromium nitride film, zinc/tin oxide film and second chromium nitride film is disclosed in the examples. Dependent claims 2, 4 and 5 correspond to original claims 10, 12, 16 and 21.

3. None of the documents cited in the search report discloses an article for the reflectance of solar energy comprising a glass substrate, a reflective transparent chromium nitride film deposited onto said glass substrate, a transparent zinc/tin oxide film in a thickness sufficient for the article to exhibit interference colours, and a second reflective transparent chromium nitride film deposited onto said zinc/tin oxide film. Therefore, the article and the process as defined in claims 1 and 3 are new over the cited prior art.

4. D4 represents the closest prior art. This document discloses a solar-control glazing having a transmission of visible light in the range of 5 to 40% and heat-reflection properties. It comprises a glass substrate, an oxide layer having an optical thickness of 20-280 nm located directly on the glass substrate and a chromium

nitride layer with a geometric thickness of 10-40 nm deposited onto the oxide layer. The oxide layer preferably consists of tin oxide, titanium oxide or aluminium oxide having a refractive index of 2, 2.3 and 1.7 respectively. The layers are deposited by cathodic sputtering. Deposition of the tin oxide layer is carried out using a tin target and an oxygen-containing atmosphere while for the chromium nitride layer a chromium target is sputtered in an atmosphere consisting of an inert gas and nitrogen.

The layer system and the process disclosed in D4 make it possible to adjust the appearance of the reflecting colour by appropriate choice of the optical thickness of the oxide layer, the colour characteristic extending from silver through bronze and blue to green if the optical thickness is gradually increased from 20 to 280 nm. Transmission in the visible range can be varied by varying the thickness of the chromium nitride layer. D4 further teaches that chromium nitride exhibits excellent mechanical resistance to grinding or scratching and is also extremely resistant to chemical agents such as chlorine, sulphur, acids and lyes. The mechanical and chemical resistance can further be improved by applying a third layer consisting of a dielectric, preferably one of the above mentioned oxides (see page 1, lines 27 to 45 and 52 to 56; page 2, lines 1 to 19 and 35 to 42; page 3, lines 6 to 9, examples 1 to 4; claims 1, 3, 4 and 7).

4.1 Starting from this prior art, the technical problem underlying the present application can be seen in providing other low transmittance coated articles for reflecting solar energy, which exhibit a variety of specific reflected colours with a minimum of layers and materials, as well as mechanical and chemical resistance.

In the absence of evidence showing that the claimed products exhibit a higher colour saturation than those of D4, this unproved effect cannot be taken into consideration for the formulation of the problem.

The problem stated above is solved by an article comprising the kind and sequence of layers as defined in claim 1. The claimed coated articles differ from the coated solar-control glazing of D4 by the oxide layer being a layer of zinc/tin alloy oxide with the composition defined in claim 1 and by the presence of an additional reflective transparent chromium nitride film on the glass substrate. The question whether or not all the reflected colours recited in claim 1 differ from those obtained in the examples of D4 can remain open since the outcome of the present decision does not depend on this issue. In view of the examples of the description, it is credible in the absence of evidence to the contrary that the problem defined above has been solved by the claimed articles.

4.2 D4 itself discloses the addition of a top layer onto the chromium nitride layer in order to further improve the mechanical and chemical resistance, the top layer consisting of a dielectric, preferably one of the oxides used for the first layer, ie tin, titanium or aluminium oxide (see page 2, lines 40 to 42, and claim 3). This teaching does not suggest to replace the tin oxide layer by a tin/zinc oxide layer and to deposit an additional transparent chromium nitride layer on the glass substrate in order to solve the problem stated above.

4.3 D5 discloses a high transmittance, low emissivity coated product comprising a glass substrate, a first transparent layer of zinc/tin alloy oxide deposited on the substrate, a transparent highly reflective metallic film such as a silver film deposited on the first oxide layer, a second transparent film of tin/zinc alloy oxide on the silver layer, and optionally a primer layer between the silver layer and the tin/zinc oxide layer, the layers being deposited by sputtering a cathode target. The coated article of examples II and III, which comprise one primer layer of zinc/tin alloy and two copper primer layers respectively, exhibit a transmittance in the visible wavelength of 87% and 83.2%. These products have a slightly bluish reflectance from both sides. It can be inferred from D5 that the articles are preferably essentially colourless (see paragraph bridging pages 2 and 3 and examples). D5 teaches that while various metal alloys may be sputtered to form the oxide films, alloys of tin and zinc are preferred in order to produce a high transmittance, low emissivity multiple layer film, in particular an alloy having a tin/zinc weight ratio from 46:54 to 50:50. The improved adhesion between the silver film and the films of zinc/tin alloy oxide results from the use of the primer layer(s) (see claims 1 to 4, 5, 7, 11, 19 to 24; page 5; page 7, first paragraph; paragraph bridging pages 8 and 9; page 9, third paragraph; examples II and III; page 13, second paragraph).

Thus, this document teaches the possibility of using two layers of tin/zinc alloy oxide instead of tin oxide layers in coated articles comprising a silver film sandwiched between metal oxide layers and exhibiting a high transmittance in the visible wavelength and a low emissivity in the IR-wavelength. However, such articles which are usually used to keep heat radiation energy inside a building do not belong to the same kind of coated architectural glass as solar control glazings, which exhibit a comparatively low transmission over the visible wavelength and reduce energy transfer from the solar radiation into the building's interior (see in this respect D1, in particular page 385 and page 386, first to fourth paragraphs).

Taking into account (i) that D4 and D5 belong to two different kinds of coated architectural glass, (ii) that D5 does not give any information about the influence of the tin/zinc alloy oxide on the reflected colours and does not discuss the problem of achieving a variety of specific reflected colours, (iii) that the improvement of adhesion between the films of tin/zinc alloy oxide and the silver film does not result from the use of the tin/zinc alloy oxide but from the presence of the primer layer(s), and (iv) that neither D4 nor D5 discloses the inclusion of an additional chromium nitride layer on the glass substrate, let alone the effect of such an inclusion on the properties of the coated articles, the Board is not convinced that the teaching of D4 and D5 would have given the skilled person an incentive to replace the tin oxide layer by a layer of tin/zinc alloy oxide in the solar control glazing of D4 and to deposit an additional chromium nitride layer on the glass substrate in order to solve the problem stated above.

4.4 D2 discloses a heatable article having a high transmittance in the visible wavelength and a low emissivity in the infra-red wavelength. It comprises a glass substrate, a first transparent anti-reflective metal oxide film comprising zinc, preferably a film of tin/zinc alloy oxide deposited on the glass substrate, a transparent IR-reflective metallic film such as a silver film deposited on the said oxide layer, a primer layer deposited on the silver film and containing Ti, Zr, Cr, or a Zn/Sn alloy, or mixture thereof, and a second transparent anti-reflective metal oxide layer comprising zinc, preferably a layer of a tin/zinc alloy oxide, deposited on the primer film. Optionally an additional primer layer is deposited between the first tin/zinc oxide film and the silver film. The layers are formed by sputtering. The article of example 1, which is coated with six layers and comprises two layers of zinc/tin alloy oxide and two primer layers of titanium, has a transmittance of 80-85% after high temperature processing. The primer layers provide the high-temperature resistance to the coating and improve adhesion between the silver and the zinc oxide or zinc/tin oxide films (see claims 1 to 4, 6, 8 to 10, and 13; column 4, line 11 to column 5, line 12; column 5, line 37 to column 6 line 8; example 1).

The objective of D2 is to overcome the drawbacks of the known multiple-layer, low emissivity, high transmittance films, which are sufficiently durable for architectural applications in multiple glazed window units, but not sufficiently temperature resistant to withstand high temperature processing, such as tempering or bending, or to function as heating elements, for example, as de-icing, defrosting and/or defogging coating for windshields (see column 3, line 56 to column 4, line 18). Thus, D2 deals with a technical problem which is completely different from that of D4 and the present application. It does not discuss the problem of obtaining a variety of specific reflected colours and the colour of the exemplified articles is either not mentioned or visually neutral. Furthermore, the improvement in adhesion between the silver layer and the zinc/tin oxide layers is not due to the tin/zinc alloy oxide but to the titanium primer layers. D2, like D5, also does not suggest the inclusion of a second chromium nitride layer in solar control glazings. In these circumstances, the skilled person confronted with the problem stated above would not have been prompted in view of the teaching of D2, D4 and D5 to replace the tin oxide layer by a layer of zinc/tin alloy oxide in the solar control glazing of D4 and to introduce an additional chromium nitride layer on the glass substrate in order to solve the said problem.

4.5 D1 already mentioned above discloses solar control films for solar control glazings having a low transmittance in the visible wavelength range. They consist of a tin oxide film deposited on the glass substrate and a titanium carbide film deposited on the tin oxide film. Chromium, stainless steel or semi-oxidised chromium can be used instead of TiC. D1 also teaches that by varying the thickness of the tin oxide layer the colour in reflection of the coated window pane can be changed from brown, green, blue to violet or even red (see page 386, 1st and 4th paragraphs; page 388, 1st and 4-6th paragraphs; page 389, 1st paragraph). As D1 discloses only two-layer solar control films and neither mentions chromium nitride nor zinc/tin alloy oxide as material for these layers, this document would not have given a hint at the claimed solution even in combination with D2, D4 and D5.

4.6 The remaining pre-published document cited in the search report also does not contain additional information pointing towards the claimed solution.

4.7 It follows from the above that the products as defined in claim 1 meet the requirement of an inventive step set out in Articles 52(1) and 56 EPC over the cited prior art.

5. As regards the independent process claim 3, the Board observes that sputtering a cathode target of a tin/zinc alloy for depositing the tin/zinc oxide layer and depositing a chromium nitride layer by sputtering a chromium target in a nitrogen atmosphere might be obvious methods of deposition in view of the cited

prior art. However, as the reasons given above in connection with the inventive step of the product claim 1 apply likewise to the product containing the kind and sequence of layers recited in claim 3, the process claim 3 derives its patentability from that of the product.

6. Claims 1 and 3 being allowable, the same applies to the dependent claims 2, 4 and 5 whose patentability is supported by that of the main claims.

7. At the oral proceedings the Appellant's representative has requested that the case be remitted to the first instance for adaptation of the description with the amended set of claims. Taking into account that, in the present case, additional information and instructions from the Appellant appear to be necessary for amending some passages of the description, the Board finds it appropriate, in accordance with Article 111(1) EPC, to remit the case to the Examining Division for adaptation of the description.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the first instance with the order to grant a patent on the basis of the set of claims submitted at the oral proceedings on 17 March 1998 and a description to be adapted.

The Registrar:

P. Martorana

The Chairman:

P. A. M. Lançon

H. Eb
I.P.