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(54) **Title:** TRANSPARENT DIFFUSIVE OLED SUBSTRATE AND METHOD FOR PRODUCING SUCH A SUBSTRATE

(57) **Abstract:** The invention is drawn to a transparent diffusive OLED substrate comprising the following successive elements or layers: - a transparent flat substrate made of mineral glass having a refractive index of between 1.45 and 1.65, - a rough low index layer comprising mineral particles, said mineral particles being bonded to one side of the substrate by means of a low index enamel, the mineral particles near, at or protruding from the enamel's surface creating a surface roughness characterized by an arithmetical mean deviation R_a comprised between 0.15 and 3 μm , the mineral particles and enamel both having a refractive index of between 1.45 and 1.65; - a high index planarization layer made of an enamel having a refractive index comprised between 1.8 and 2.1 covering the rough low index layer (b). It is also drawn to a method for preparing such a diffusive OLED substrate.



TRANSPARENT DIFFUSIVE OLED SUBSTRATE AND METHOD FOR PRODUCING SUCH A SUBSTRATE

The present invention is drawn to a new method for producing
5 translucent, light-scattering substrates for organic light emitting diodes
(OLED) and to substrates obtainable by such a method.

OLEDs are opto-electronic elements comprising a stack of organic
layers with fluorescent or phosphorescent dyes sandwiched between two
electrodes, at least one of which is translucent. When a voltage is applied to
10 the electrodes the electrons injected from the cathode and the holes injected
from the anode combine within the organic layers, resulting in light emission
from the fluorescent/phosphorescent layers.

It is commonly known that light extraction from conventional OLEDs is
rather poor, most of the light being trapped by total internal reflection in the
15 high index organic layers and transparent conductive layers (TCL). Total
internal reflection takes place not only at the boundary between the high
index TCL and the underlying glass substrate (refractive index of about 1.5)
but also at the boundary between the glass and the air.

According to estimates, in conventional OLEDs not comprising any
20 additional extraction layer about 60 % of the light emitted from the organic
layers is trapped at the TCL/glass boundary, an additional 20 % fraction is
trapped at the glass/air surface and only about 20 % exit the OLED into air.

It is known to reduce this light entrapment by means of a light
scattering layer between the TCL and the glass substrate. Such light
25 scattering layers have a high refractive index close to the TCL index and
contain a plurality of light diffusing elements.

It is also known to increase out-coupling of light by texturing the
interface between the glass and the high index layers of the OLED.

Both of these "internal" extraction means, also commonly called
30 "internal extraction layers" (IEL), comprise asperities that need to be
planarized before applying the TCL and the organic stack.

WO2011/089343 discloses OLED substrates comprising at least one
textured surface planarized with a high index glass coating. The substrates

are described as being texturized by acid etching. Glass etching using strong acids, in particular HF, is a commonly used method for texturizing glass surfaces. Such a wet chemistry method however is a complicated process when carried out on thin glass (thickness <1 mm). This technique allows only
5 for one of the two faces to be etched per process step as the glass plate has to be kept in a horizontal position during the etching step. Furthermore the roughness profile parameters are difficult to optimize and above all the use of HF results in important security problems for the environment and persons working nearby.

10 The applicant has recently developed an interesting alternative method for roughening one or both sides of the glass substrate, said method comprising mechanical roughening (lapping). This method, described in European application 12306179.8 filed on September 28, 2012, is much less hazardous than chemical etching, allows for better control of the roughness
15 profile and makes it possible to simultaneously roughen both sides of the substrates, thereby producing in a single process step the internal and external extraction layers (IEL and EEL) of a transparent OLED glass substrate.

The present invention is drawn to still another method for producing
20 diffusive low index glass substrates, said method comprising neither a chemical etching step nor a mechanical abrasion step. The idea underlying the present invention is to bond low index mineral particles by means of a low index mineral binder to a low index glass substrate, the amount of mineral binder with respect to the mineral particles being sufficiently low so that the
25 mineral particles protrude from the binder surface or, at least, create significant roughness at the mineral binder surface.

The diffusive low index substrate thus obtained is then submitted to a commonly known planarization step using a high index frit, and the resulting planarized diffusive substrate may then be coated with a transparent
30 conductive layer (TCL) and be used as a light-extraction substrate for OLEDs.

The method of the present invention is easy to implement, requiring only rather simple and commonly known equipment. One significant

advantage over the lapping method described in EP 12306179.8 is that it may be used for very large surfaces. Additionally, the lapping method slightly decreases the substrate's mechanical resistance, which is not the case for the method of the present invention.

5 The first subject-matter of the present invention is a transparent diffusive OLED substrate comprising the following successive elements or layers:

- (a) a transparent flat substrate made of mineral glass having a refractive index of between 1.45 and 1.65,
- 10 (b) a rough low index layer comprising mineral particles, said mineral particles being bonded to one side of the substrate by means of a low index enamel, the mineral particles near, at or protruding from the enamel's surface creating a surface roughness characterized by an arithmetical mean deviation R_a comprised between 0.15 and
- 15 3 μm , the mineral particles and low index enamel both having a refractive index of between 1.45 and 1.65;
- (c) a high index planarization layer made of an enamel having a refractive index comprised between 1,8 and 2,1 covering the rough low index layer (b).

20 The present invention also provides a method for preparing a diffusive substrate as defined above.

 The low index layer of the OLED substrate of the present invention is defined by its refractive index (1.45 – 1.65) and by its surface roughness profile, i.e. an arithmetical mean deviation R_a (such as defined in ISO 4287)

25 comprised between 0.15 and 3 μm , said roughness being created by the mineral particles near, at or protruding from the low index enamel's surface. The mineral particles do not necessarily need to protrude from the low index enamel but may be embedded therein as long as it is apparent, for example from a sectional SEM view, that the roughness or waviness of the low index

30 mineral layer can be attributed to the underlying particles, the surface profile closely matching the presence/absence of embedded mineral particles.

 The mineral particles used in the present invention may be crystalline, amorphous or semi-crystalline particles. They may have a random shape

with more or less sharp edges but preferably are rather spherical particles free of sharp edges.

In a preferred embodiment, the mineral particles are solid beads. Such beads are preferred over randomly-shaped sharp-edged particles because
5 they easily spread over the substrate's surface thereby facilitating formation of a thin monolayer of beads, rather than large sized aggregates. Sphere-like particles devoid of sharp edges are also more easily planarized than randomly shaped particles. It is to be understood that hollow beads are not encompassed in the definition of mineral particles of the present invention,
10 because the gas contained therein has a refractive index not comprised between 1.45 and 1.65.

The term "mineral particle", especially when used to describe the method of the present invention, encompasses particles functionalized with organic surface groups, such as trialkylsilyl groups. Said organic surface
15 groups undergo thermal decomposition during the baking or fusing step of the mineral binder or, at the latest, during the formation of the high index enamel layer and consequently are no longer present in the final product.

The mineral particles used in the present invention, whether spherical or not, have an average equivalent spherical diameter (measured by DLS) of
20 between 0.3 μm and 10 μm , preferably of between 0.5 μm and 8 μm , more preferably of between 0.8 μm and 7 μm , the equivalent spherical diameter of the irregularly shaped particles being defined as the diameter of the sphere having the same volume as the mineral particle.

The average equivalent spherical diameter however is not the only
25 size parameter to consider for selecting the mineral particles to be used in the present invention. Advantageously, the mineral particles are essentially free of large sized particles, which would protrude not only from the mineral binder but also from the high index enamel layer, which would then lead to current leakage in the final OLED. The mineral particles used in the present
30 invention consequently are preferably essentially devoid of particles having an equivalent spherical diameter higher than 15 μm , preferably higher than 12 μm .

As already specified above, the glass substrate, the mineral particles and the mineral binder, i.e. the low index enamel, all have about the same refractive index, comprised between 1.45 and 1.65, preferably between 1.50 and 1.60.

5 In a preferred embodiment of the present invention, the mineral particles are selected from silica particles.

In order to obtain diffusive substrates from ingredients all having about the same refractive index, it is necessary to create and control the surface roughness of the low index mineral layer. As mentioned above, the low index
10 mineral layer should have an arithmetical mean deviation R_a comprised between 0.15 and 3 μm , preferably between 0.2 and 2 μm .

The arithmetical mean deviation R_a is defined in ISO 4287. It may be measured by scanning electron microscopy (SEM) of cross sections of the sample, by surface profile measurement or by 3D laser microscopy.

15 To obtain a mineral low index layer having both a suitable surface roughness and a satisfactory mechanical resistance, it is important to appropriately select the amount of low index glass frit with respect to the amount of mineral particles. If one uses too high amounts of glass frit, the mineral particles will be completely embedded in the resulting low index
20 mineral binder matrix and will not create the required surface roughness (R_a) of between 0.15 and 3 μm . On the other hand, in case the amount of mineral binder is too low with respect to the mineral particles, the bonding strength of the enamel binder is too weak and the resulting mineral layer will be excessively brittle and easily damaged when handled.

25 The applicant found that a weight ratio of mineral particles to the glass frit comprised between 0.2 and 4, preferably between 0.4 and 3, leads to a suitable surface roughness and mechanical resistance of the low index layer.

The final low index mineral layer may also be characterized by the volume ratio of the mineral particles to the low index enamel which is
30 preferably comprised between 0.3 and 3, preferably between 0.5 and 2 and more preferably between 0.7 and 1.5.

The high index enamel (c) on the low index mineral layer (b) should be thick enough to completely cover and planarize the roughness profile

thereof.

The thickness of the high index layer is advantageously comprised between 3 μm and 20 μm , preferably between 4 μm and 15 μm and more preferably between 5 μm and 12 μm . The thickness of the high index layer is
5 the mean distance between the mean lines (defined as in ISO 4287, 3.1.8.1) of the roughness profile of the low index layer and the roughness profile of the high index layer.

The surface roughness of the high index layer should be preferably as low as possible and the high index enamel advantageously has an
10 arithmetical mean deviation R_a of less than 3 nm, more preferably less than 2 nm and most preferably less than 1 nm.

The high index layer is preferably essentially free of diffusive elements dispersed therein, especially free of diffusive solid particles dispersed therein. As a matter of fact such solid diffusive particles could
15 undesirably protrude from the surface of the high index layer and cause leakage currents in the final OLED.

The resulting flat glass substrate carrying the low index mineral layer (low index mineral particles + low index enamel) planarized by the high index glass frit generally has a haze comprised between 75 and 98 %, preferably between 85 and 97 %, and more preferably between 87 and 95 %. Haze value can be measured by optical spectrophotometers like PE
20 Lambda 950 or Varian Carry 5000, but also by faster and cheaper dedicated devices like BYK Hazemeter.

In a preferred embodiment, the transparent diffusive OLED
25 substrate of the present invention further comprises a transparent electro-conductive layer (d) preferably directly in contact with the high index enamel layer (c). Such transparent conductive layers that may be used as anodes for OLEDs are well known in the prior art. The most commonly material used is ITO (Indium Tin Oxide). The transparent conductive layer should have a light
30 transmission of at least 80 %, and a refractive index (at $\lambda = 550 \text{ nm}$) of between 1.7 and 2.2. Its total thickness is typically comprised between 50 and 400 nm.

As mentioned above the present invention is also drawn to a method for preparing the OLED substrate of the present invention, said method
35 comprising the following successive steps:

- (1) Providing a transparent flat substrate made of mineral glass having a refractive index of between 1,45 and 1,65;
- (2) Applying onto one side of said substrate a low index glass frit mixed with mineral particles having a glass transition temperature (T_g) or a fusion temperature at least 50 °C higher than the T_g of the glass frit, both the glass frit and the mineral particles having a refractive index of between 1.45 and 1.65;
- (3) Heating the resulting glass frit layer to a temperature allowing fusion of the glass frit without fusion of the mineral particles, resulting in an rough low index layer comprising mineral particles bonded to the substrate by means of a low index enamel;
- (4) Applying onto said rough low index layer a layer of a high index glass frit having a refractive index of between 1.8 and 2.1;
- (5) Drying and fusing said high index glass frit so as to obtain a high index enamel having a refractive index comprised between 1.8 and 2.1 covering the transparent rough low index layer.

The flat glass substrates provided at step (1) advantageously have a thickness of between 0.1 and 5 mm, preferably of between 0.3 and 1.6 mm.

At step (2) the glass frit particles and mineral particles are mixed and suspended in a conventional organic vehicle comprising an organic solvent and an organic polymer. The suspension is then applied according to known techniques such as screen printing or slot coating. The mineral particles may be amorphous, crystalline or semi-crystalline. They should not fuse or be substantially softened during the subsequent fusion step (4) of the glass frit. That's why the fusion point of the crystalline particles or the T_g of the amorphous fraction of the particles must be significantly higher than the T_g of the glass frit, i.e. at least 50 °C, more preferably at least 100 °C higher than the T_g of the glass frit.

Low index glass frits that may be used in the present invention for bonding the mineral particles to the glass substrates are well known in the art.

Preferred low index glass frits have the following composition:

SiO₂: 10 – 40 wt%

Al₂O₃: 1 – 7 wt%

B₂O₃: 20 – 50 wt%

Na₂O+Li₂O+K₂O: 5 – 30 wt%

ZnO: 3 – 35 wt%

At step (3) the frit-coated substrate is submitted to firing at a
5 temperature sufficiently high to effect fusion of the glass frit. To obtain
complete fusion of the glass frit, it is generally necessary to heat the
substrate to a temperature at least 100 °C higher than the T_g of the glass frit
and to maintain this temperature for a duration of about 2 to 30 min.

In a typical embodiment, the glass frit and mineral particles
10 (70 – 80 wt%) are mixed with 20 – 30 wt% of an organic vehicle (ethyl
cellulose and organic solvent). The resulting paste is then applied onto the
glass substrate for example by screen-printing or slot-coating. The resulting
layer is dried by heating at a temperature of 120 – 200 °C. The organic
binder (ethyl cellulose) is burned out at a temperature of between 350
15 – 440 °C, and the firing step resulting in the final enamel is carried out at a
temperature of between 510 °C and 610 °C, preferably between 520 °C and
600 °C.

At step (4) the high index glass frit is then applied onto the low index
rough layer by any suitable method such as screen printing, spray coating,
20 bar coating, roll coating, slot coating and spin coating, of an aqueous or
organic suspension of glass particles. A description of suitable high index
glass frits and methods for coating and firing them can be found for example
in EP 2 178 343.

The glass frit should be selected to have a melting point comprised
25 between 450 °C and 570 °C and should lead to an enamel having a refractive
index of 1.8 to 2.2.

Preferred high index glass frits have the following composition:

Bi₂O₃: 55 – 75 wt%

BaO: 0 – 20 wt%

30 ZnO: 0 – 20 wt%

Al₂O₃: 1 – 7 wt%

SiO₂: 5 – 15 wt%

B₂O₃: 5 – 20 wt%

Na₂O: 0.1 – 1 wt%

CeO₂: 0 – 0.1 wt%

In a typical embodiment, the glass frit particles (70 – 80 wt%) are mixed with 20 – 30 wt% of an organic vehicle (ethyl cellulose and organic solvent). The resulting frit paste is then applied onto the diffusive coated glass substrate by screen printing or slot coating. The resulting layer is dried by heating at a temperature of 120 – 200 °C. The organic binder (ethyl cellulose) is burned out at a temperature of between 350 – 440 °C, and the firing step resulting in the final enamel is carried out at a temperature of between 510 °C and 610, preferably between 520 °C and 600 °C.

The resulting enamels have been shown to have a surface roughness with an arithmetical mean deviation R_a (ISO 4287) of less than 3 nm when measured by AFM on an area of 10 μm x 10 μm.

The amount of the high index glass frit coated onto the roughened surface is generally comprised between 20 and 200 g/m², preferably between 25 and 150 g/m², more preferably between 30 and 100 g/m², and most preferably between 35 and 70 g/m².

The high index glass frit used in the present invention and the enamel resulting therefrom preferably are substantially devoid of solid scattering particles such as crystalline SiO₂ or TiO₂ particles. Such particles are commonly used as scattering elements in high index scattering layers but generally require an additional planarization layer, thereby increasing the total thickness of the high index coating.

The diffusive substrates planarized with high index enamel are particularly useful as substrates for bottom-emitting OLEDs. A transparent conductive layer has to be applied on top of the high index enamel before application of the stack of organic light emitting layers.

In a preferred embodiment, the method of the present invention therefore further comprises an additional step of coating the high index enamel resulting from step with a transparent conductive layer, preferably a transparent conductive oxide. Formation of such a TCL may be carried out according to conventional methods such as magnetron sputtering.

Example 1

A low index frit (20 parts by weight) is mixed with spherical SiO_2 particles (10 parts by weight) having an average equivalent diameter of 6 μm . The resulting powder is dispersed in 70 parts by weight of an organic medium using a 3-roll milling process.

5 The low index frit used has the following composition: 28.4 wt% of SiO_2 ; 3.6 wt% of Al_2O_3 ; 39.5 wt% of B_2O_3 ; 15.9 wt% of alkali oxides (Na_2O , Li_2O , K_2O); 12.6 wt% of ZnO . It has a refractive index of 1.54 and a T_g of 484 °C.

10 The resulting slurry is coated by screen-printing on a soda-lime glass substrate (0.7 mm) and then submitted to drying at 150 °C. The dried coating is fired at 600 °C for 20 minutes in an IR furnace.

Figure 1 shows the SEM micrograph of the rough low index layer after firing and before planarization with the high index frit.

15 The resulting low index rough layer was then coated by screen-printing with a slurry of a high index frit having a refractive index of 1.90.

The coating was dried at 150 °C and was submitted to firing for 10 minutes at 545 °C in an IR furnace.

20 Figure 2 shows the SEM micrograph of the planarized substrate (glass substrate carrying mineral particles bonded by low index enamel in grey; high index planarization layer in white).

REVENDECATIONS

- 5 1. A transparent diffusive OLED substrate comprising the following successive elements or layers :
- (a) a transparent flat substrate made of mineral glass having a refractive index of between 1.45 and 1.65,
- 10 (b) a rough low index layer comprising mineral particles, said mineral particles being bonded to one side of the substrate by means of a low index enamel, the mineral particles near, at or protruding from the enamel's surface creating a surface roughness characterized by an arithmetical mean deviation R_a comprised between 0.15 and 3 μm , the mineral particles and enamel both having a refractive
- 15 index of between 1.45 and 1.65;
- (c) a high index planarization layer made of an enamel having a refractive index comprised between 1,8 and 2,1 covering the rough low index layer (b).
2. The substrate according to claim 1, wherein the mineral particles
- 20 have an average equivalent spherical diameter of between 0.3 μm and 10 μm , preferably of between 0.5 μm and 8 μm , more preferably of between 0.8 μm and 7 μm .
3. The substrate according to claim 1 or 2, wherein the mineral particles are solid beads.
- 25 4. The substrate according to any of the preceding claims, wherein the mineral particles are essentially free of particles having an equivalent spherical diameter higher than 15 μm , preferably higher than 12 μm and more preferably higher than 10 μm .
5. The substrate according to any of the preceding claims, wherein the
- 30 refractive index of the substrate, low index enamel and mineral particles is comprised between 1.50 and 1.60.
6. The substrate according to any of the preceding claims, wherein the thickness of the high index layer is comprised between 3 μm and 20 μm , preferably between 4 μm and 15 μm and more preferably between 5 μm and
- 35 12 μm .

7. The substrate according to any of the preceding claims, wherein the surface roughness of the high index layer has an arithmetical mean deviation R_a of less than 3 nm, more preferably less than 2 nm and most preferably less than 1 nm.

5 8. The substrate according to any of the preceding claims, wherein the high index layer is essentially free of diffusive elements dispersed therein, especially free of diffusive solid particles dispersed therein.

9. The substrate according to any of the preceding claims, wherein the mineral particles are selected from silica particles.

10 10. The substrate according to any of the preceding claims further comprising a transparent electro-conductive layer on the high index enamel layer.

11. The substrate according to any of the preceding claims, wherein the volume ratio of the mineral particles to the low index enamel is comprised
15 between 0.3 and 3, preferably between 0.5 and 2 and more preferably between 0.7 and 1.5.

12. A method for preparing a transparent diffusive substrate according to any of claims 1 - 11, comprising the following steps :

20 (1) Providing a transparent flat substrate made of mineral glass having a refractive index of between 1,45 and 1,65;

(2) Applying onto one side of said substrate a low index glass frit mixed with mineral particles having a glass transition temperature (T_g) or a fusion temperature at least 50 °C higher than the T_g of the glass frit, both the glass frit and the mineral particles having a refractive index of
25 between 1.45 and 1.65;

(3) Heating the resulting glass frit layer to a temperature allowing fusion of the glass frit without fusion of the mineral particles, resulting in an rough low index layer comprising mineral particles bonded to the substrate by means of a low index enamel;

30 (4) Applying onto said rough low index layer a layer of a high index glass frit having a refractive index of between 1.8 and 2.1;

(5) Drying and fusing said high index glass frit so as to obtain a high index enamel having a refractive index comprised between 1.8 and 2.1 covering the transparent rough low index layer.

13. The method according to claim 12, wherein the mineral particles have an average equivalent spherical diameter of between 0.3 μm and 10 μm , preferably of between 0.5 μm and 8 μm , more preferably of between 0.8 μm and 7 μm

5 14. The method according to claim 12 or 13, wherein the weight ratio of the mineral particles to the glass frit is comprised between 0.2 and 4, preferably between 0.4 and 3.

10 15. The method according to any of claims 12 to 14, wherein the fusing of the high index glass frit is carried out at a temperature comprised between 510 $^{\circ}\text{C}$ and 580 $^{\circ}\text{C}$, preferably between 520 $^{\circ}\text{C}$ and 580 $^{\circ}\text{C}$.

1/1

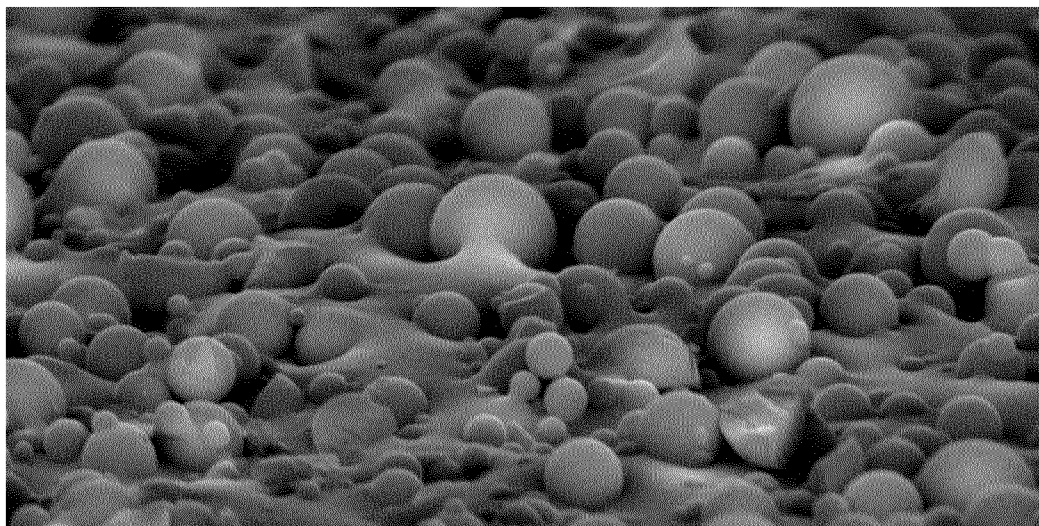


Figure 1

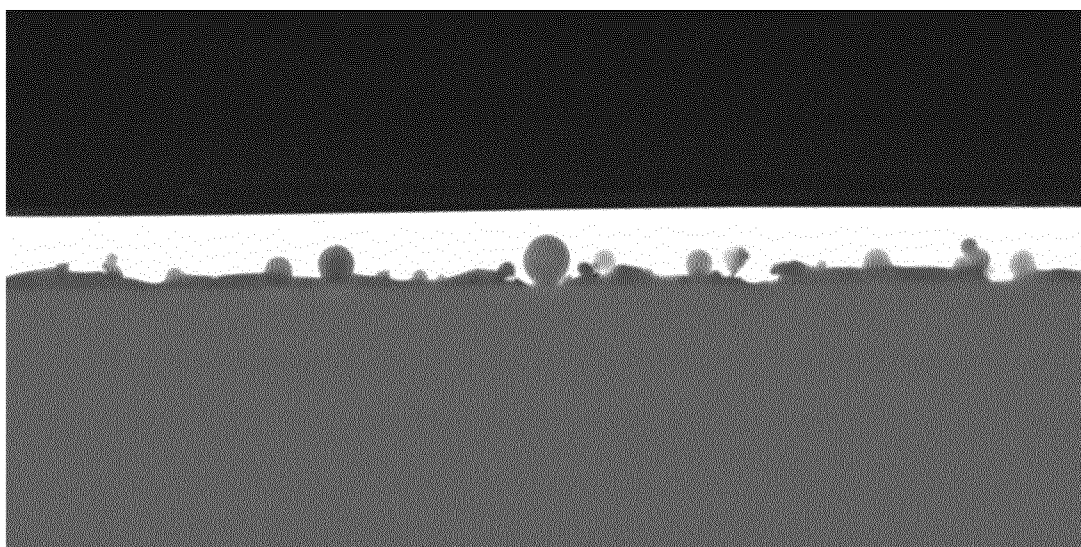


Figure 2

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2014/058737

A. CLASSIFICATION OF SUBJECT MATTER

INV. C03C3/066 C03C8/02 C03C17/34
ADD.

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
C03C

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	FR 2 963 705 A1 (SAINT GOBAIN [FR]) 10 February 2012 (2012-02-10) claims 1,7,9 page 19 - page 21 -----	1-15
Y	US 2004/156110 A1 (IKEYAMA AKIHIRO [JP]) 12 August 2004 (2004-08-12) claim 1 -----	1-15
A	WO 2007/032977 A1 (FERRO CORP [US]; SAKOSKE GEORGE E [US]; CLAUD MICHEL [FR]; LARONZE HER) 22 March 2007 (2007-03-22) the whole document -----	1-15



Further documents are listed in the continuation of Box C.



See patent family annex.

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Date of the actual completion of the international search

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INTERNATIONAL SEARCH REPORT

Information on patent family members

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