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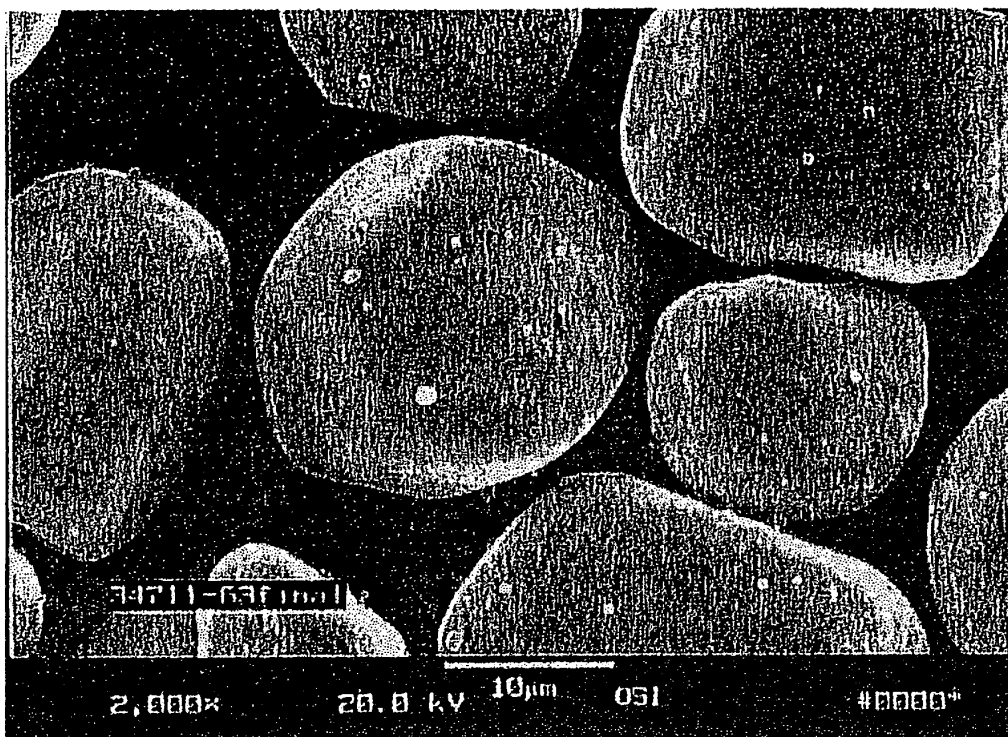
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(54) **Manufacturing method of light-emitting material**

(57) The present invention provides a manufacturing method of a light-emitting material of a long service life, which includes adding boron ions and halogen ions in this order to an alcohol solution of an organic silicon compound having a hydroxyl group and a methoxyl group to obtain a solution of a reaction product by hydrolysis/dehydration condensation at normal temperature, then dis-

persing fine particles of a light-emitting material in alcohol under agitation, adding the solution of the reaction product into such alcohol dropwise to form a glass film of the reaction product on the surface of the fine particles of the light-emitting material, then separating the fine particles and spray drying the fine particles at a temperature of 200°C or lower.

Fig. 2



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Description

BACKGROUND OF THE INVENTION

Field of the Invention

[0001] The present invention relates to a manufacturing method of a light-emitting material, capable of maintaining luminance of a light-emitting element such as an EL element, a fluorescent element or a light-accumulating element, thereby achieving long service life thereof.

Description of Related Art

[0002] Electroluminescence (hereinafter represented as EL), discovered 1936 by G. Destriau, France, as a phenomenon of light emission when an AC high voltage was applied to a ZnS phosphor, has been an object of numerous developmental researches for application to a flat panel display and is now reaching a stage of commercial use. In the course of such development, a higher luminance, a higher definition, a lower electric power consumption, a longer service life and a lower cost are set as technical targets to be attained. Difficulties in attaining these targets arise from various causes such as deterioration in the light-emitting material, the electron transport material, the positive hole transport material, the structure of elements, an interface or an electrode. Among these, a deterioration of the light-emitting material is a basically important point, and a complete sealing is required. Therefore, elimination and exclusion of oxygen, water (moisture) and fine dust particles are essential (cf. non-patent literatures 1 - 3).

[0003] For solving these difficulties, various structures of the light-emitting material itself are being tested. For example, patent literature 1 describes an invention attaining a high light-emitting efficiency and a long service life by employing a benzothiophene derivative as an organic field light-emitting element, and, as an example of inventions for attaining a longer service life irrespective of the structure of the high-emitting material itself, patent literature 2 discloses a method of forming a diamond-like carbon (DLC) film so as to cover an entire light-emitting substrate, thereby improving reliability and extending the service life. However, since such DLC film is a thin film, a resinous film has to be further applied in order to protect the DLC film from being peeled for example by a scratch. Also a method of evaporating an inorganic film, for example by sputtering, on the surface of the light-emitting material is being used, but, in this method, the film is merely vapor deposited on the surface and cannot enter small pores, thus being unable to provide a sufficient sealing effect and insufficient to provide a long-life EL display device.

[0004] The present invention has been made in consideration of the aforementioned situation, and has been created to form, on the surface of the light-emitting material, a glass film which is thin but tough and is not peeled by a scratch or the like, and completely seals the surface even including fine pores, and completely excluding oxygen, water (moisture) and fine particles, thereby preventing deterioration of the light-emitting material and maintaining a high luminance, thus achieving a long service life.

[0005] Glass products, having characteristics such as hardness, rigidity, permeation resistance, stain resistance, moisture barrier, and a gas barrier, are utilized in familiar applications such as various containers, illumination devices, display devices, optical glass, bulb glass, color glass, art glass, solder glass, glass for passivation, crystal glass and the like. Also, utilizing the aforementioned properties, glass is used as a barrier film for moisture and gasses in a pinhole-free glass coating, for example on a metal surface.

[0006] However, all the known technologies for forming such glass coating film requires a heat treatment step at a temperature as high as 1500 - 2300°C. Also a recently developed sol-gel method of hydrolyzing an organic silicon compound requires a heating process of 1100°C or higher for glass formation to achieve final complete dehydration, and also a volume contraction at such process is not negligible. It has thus been considered impossible to form a glass film having moisture barrier property and gas barrier property in a low temperature of 200°C or less (hereinafter called "normal temperature range"). It has however become possible to form a glass film in the normal temperature range by utilizing a reaction catalyst, and an invention comprising a manufacturing method of a light-emitting material of a long service life has been made utilizing such technology to form a pinhole-free glass film on the surface of a light-emitting material, thus exploiting the aforementioned physical properties of the glass.

Patent Literature 1: JP-A No. 2001-354668;

Patent Literature 2: JP-A No. 2003-234179;

Non-patent Literature 1: Toshio Inokuchi, "Electroluminescent Display", Sangyo Tosho Co. (1991);

Non-patent Literature 2: Junji Kido, "Yuki EL no subete (all aspects of organic EL)", Nippon Jitsugyo Shuppan Co. (2003);

Non-patent Literature 3: Masayuki Kawamura, "Yokuwaku yuki EL display (organic EL display made easy to understand)", Dempa Shimbun Co. (2003).

SUMMARY OF THE INVENTION

[0007] A problem to be solved by the present invention to attain a method of blocking out moisture and dust particles which prevent long-life luminance of the light-emitting material employed in the EL display is how to form a thin protective glass film, that is rigid and does not decrease the luminance of the EL display, on the surface of the light-emitting material. Therefore, an object of the present invention is to provide a manufacturing method of a light-emitting material of a long service life by a process conducted in a normal temperature range.

[0008] The aforementioned object can be attained, according to the present invention, by a manufacturing method of a light-emitting material which includes adding boron ions and halogen ions in this order to an alcohol solution of an organic silicon compound having a hydroxyl group and a methoxyl group to obtain a solution of a reaction product by hydrolysis/dehydration condensation at normal temperature, then dispersing fine particles of a light-emitting material in alcohol under agitation, adding the solution of the reaction product into such alcohol dropwise to form a glass film of the reaction product on the surface of the fine particles of the light-emitting material, then separating the fine particles and spray drying the fine particles at a temperature of 200°C or lower.

[0009] The invention also provides a manufacturing method of a light-emitting material which includes adding boron ions and halogen ions in this order to an alcohol solution of an organic silicon compound having a hydroxyl group and a methoxyl group to obtain a solution of a reaction product by hydrolysis/dehydration condensation at normal temperature, then misting and spraying the solution of the reaction product on fine particles of a light-emitting material dispersed and floating in a mixer to form a glass film of the reaction product on the surface of the fine particles of the light-emitting material, and spray drying the fine particles at a temperature of 200°C or lower.

[0010] In the invention, it is effective to add, in the case of hydrolysis/dehydration condensation of the organic silicon compound, alkyl titanate or titanium chloride as an accelerator. Also, a compound having an isocyanate group (-NCO) may be added so as to be 1- 10 wt. % of the organic silicon compound to promote gelation and to increase viscosity, thereby accelerating glass formation by hydrolysis/dehydration condensation. Incidentally, a light-accumulating material is included in the light-emitting material in the invention.

[0011] The invention further provides a manufacturing method of a light-emitting material which comprises dissolving, in alcohol, silicon tetrachloride in an amount which is 0.05 - 3.0 wt.% that of the fine particles of the light-emitting material, deflocculating and dispersing the fine particles of the light-emitting material in the solution, causing a hydroxyl group on the surface of the fine particles of the light-emitting material to react with silicon tetrachloride while mixing to form a glass film of silicon oxide on the surface of the fine particles, then separating the fine particles and spray drying the fine particles at a temperature of 200°C or lower. In this method, the thickness of the film can be controlled by changing the amount of silicon tetrachloride relative to the fine particles of the light-emitting material within the aforementioned range.

[0012] This manufacturing method for the light-emitting material forms a thin glass film of a thickness of 0.05 μm or less, which is strong, not peelable by a scratch or the like and not sacrificing luminance in an EL display, forming the film while maintaining a normal temperature range on the light-emitting material, so as to completely seal the surface of the light-emitting material, thereby completely blocking out oxygen, water (moisture) and fine particles, and provides the particular advantage of preventing the deterioration of the light-emitting material, thereby providing a light-emitting material of a long service life capable of maintaining a high luminance.

BRIEF DESCRIPTION OF THE DRAWINGS

[0013]

Fig. 1 is a view showing a comparison of change in luminance of light-emitting materials;

Fig. 2 is an electron photomicrograph of fine particles of a light-emitting material produced by a method of the present invention; and

Fig. 3 is a view showing a comparison of change in afterglow luminance of light-accumulating materials.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

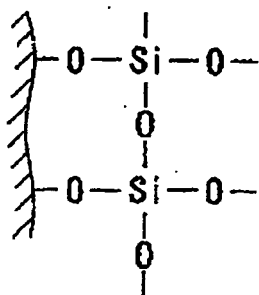
[0014] The method of the present invention for producing a light-emitting material includes following three steps:

- (1) a step of preparing a solution of a reaction product for film formation;
- (2) a step of forming a glass film on the surface of a light-emitting material; and
- (3) a spray drying step.

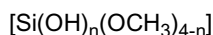
[0015] Here, these steps will be explained in this order.

(1) Step of preparing a solution of a reaction product for film formation (hereinafter called "principal reactant solution")
In the invention, a glass film to be formed on a surface of a light-emitting material is a thin film of a thickness of 0.05 μm or less, basically having a network of a silicon oxide chemically fixed to the surface of the light-emitting material as shown in the following chemical structure 1.

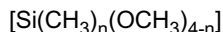
[chemical structure 1]



An organic silicon compound employed as a raw material is an alcohol-soluble silane compound having a hydroxyl group and a methoxyl group. A particularly preferred organic silicon compound has a hydroxyl group (-OH), a methoxyl group (-OCH₃) and a methyl group (-CH₃) group, such as a hydroxymethoxysilane or a methoxymethylsilane, which are respectively represented by general formulas:

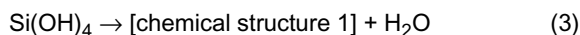
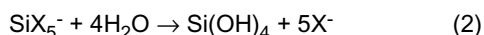
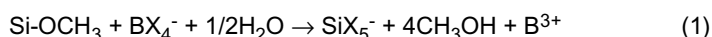


and



wherein n indicates an integer from 1 to 3. Among such organic silicon compounds there are commercial products that are easily available and usable. Also, such organic silicon compound is commercially available as a low condensate obtained by a partial hydrolysis/dehydration condensation, which can also be employed as a raw material. In the method of the invention, the main reactant solution to be employed for forming the glass film is obtained by executing a hydrolysis/dehydration condensation of the organic silicon compound at the normal temperature, in an alcohol solution and in the presence of boron ions and halogen ions. At first a compound providing a boron ion B³⁺ and a compound providing a halogen ion X⁻ are added in this order to form a complex ion BX₄⁻ in the solution. The compound providing a boron ion B³⁺ can advantageously be boric acid H₃BO₃ or triethoxyborane B(OEt)₃. A concentration of B³⁺ ions in the reaction solution is preferably within a range of 1.0 to 3.0 wt.% with respect to the organic silicon compound. Also halogen ion X⁻ can be a fluorine ion F⁻, a chlorine ion Cl⁻ or a mixture thereof. The compound to be employed is preferably a salt capable of generating such an ion in the reaction liquid. The F⁻ ion source is preferably ammonium hydrogen fluoride (NH₄F·HF) or sodium fluoride (NaF) or the like, and the Cl⁻ ion source is preferably ammonium chloride (NH₄Cl) or the like. The concentration of halogen ions in the reaction solution, in the form of NH₄F·HF or NH₄Cl, is preferably within a range of 1.0 to 3.0 wt.% with respect to the organic silicon compound.

Apparently, the complex ion BX₄⁻ generated in the solution easily exchanges with a methoxyl group portion Si(OCH₃) of the organic silicon compound to become a complex ion SiX₅⁻ as indicated by a reaction formula (1) and to accelerate a hydrolysis/dehydration condensation reaction as indicated by formulas (2) and (3), thereby forming a glass film having a silicon oxide network as shown in the chemical structural 1, in the normal temperature range.



In the method of the invention, for preparing the main reactant solution to be employed for forming the glass film, the aforementioned organic silicon compound may be directly employed in the reaction, but it is preferably diluted

with a suitable solvent in order to facilitate control of the reaction. As the diluting solvent, an aliphatic lower alcohol, such as methanol, ethanol, propanol, or isopropanol, either singly or in a mixture can be advantageously chosen. The amount of the diluting solvent is variable depending on a particle size of the organic silicon compound, but it is preferable to dissolve the organic silicon compound within a range of 50 - 400 g in 1 liter of the solvent.

In the preparation of the main reactant solution, an alkyl titanate or titanium tetrachloride is effective as an accelerator to be added for the hydrolysis/dehydration condensation reaction. The alkyl titanate can be, for example, tetrabutyl titanate. Its amount is advantageously 1 - 10 wt. % with respect to the organic silicon compound. It is also possible to add a compound having an isocyanate group (-NCO) for promoting gelation and causing a viscosity increase, thereby accelerating glass formation by the hydrolysis/dehydration condensation. Its amount is preferably 1 - 10 wt. % with respect to the organic silicon compound. The compound having the isocyanate group can be, for example, tolylene diisocyanate (TDI) or diphenylmethane diisocyanate. The promotion of gelation by an isocyanate group is apparently due to the formation of an urethane bond (-NHCOO-) by reaction of a hydroxyl group (OH) of the organic silicon compound.

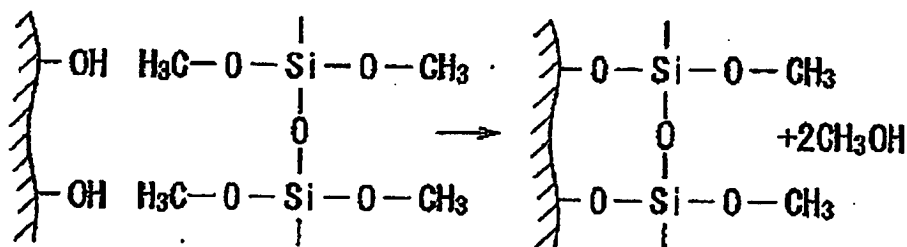
(2) Step of forming a film on a surface of a light-emitting material

In the method of the invention, the main reactant solution obtained as explained above is coated on the surface of the light-emitting material, to form a thin glass film, which is spray dried at a normal temperature range to obtain a light-emitting material. Formation of the glass film by coating the surface of the light-emitting material with the reaction product can be achieved by two methods. In one method, a fine powder of the light-emitting material is dispersed under agitation in alcohol while stirring, and the main reactant solution mentioned above is dropwise added or sprayed into the alcohol. In this method, the fine powder of the light-emitting material dispersed in alcohol preferably has a concentration within a range of 50 to 800 g per 1 liter of the alcohol. Also the main reactant solution is added to the fine particles of the light-emitting material silicon compound in the main reactant solution within a range of 0.05 to 3.0 wt. % with respect to the fine particles, for example so that there is 0.05 to 3.0 g of the organic silicon compound to 100 g of the light-emitting material. Outside such range, it becomes difficult to regulate the thickness of the glass film to be 0.05 μm or less. After the dropwise addition of the main reactant solution is terminated, the mixing is continued for a while and the entire system is transferred to a decantation container, and the layer containing the particles of the light-emitting material is separated.

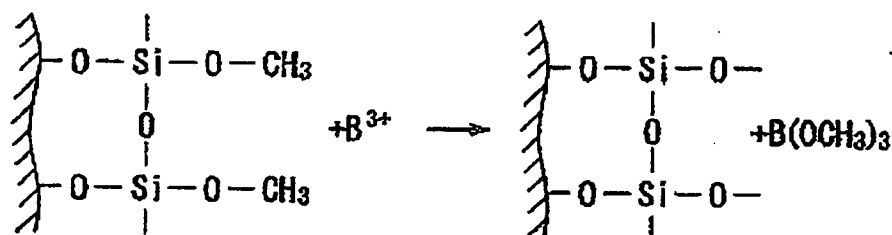
Another method of forming the glass film by coating the surface of the light-emitting material with a film of the reaction product is executed without a dispersion medium. A fine powder of a light-emitting material is well agitated for example by a Henschel mixer to form a floating dispersion in the air, and the main reactant solution in an amount of 0.1 - 2.0 wt. % to the fine powder is sprayed to the fine powder. A sufficient agitation is executed after the spraying, then the mixer is stopped and the fine powder is taken out from the mixer.

In such glass film forming step, the organic silicon compound present in the main reactant solution apparently reacts with the hydroxyl group present on the surface of the fine particles of the light-emitting material to generate methanol, whereby the silicon oxide network of the chemical structure 1 is chemically fixed to the surface of the fine particles of the light-emitting material as indicated by following chemical structures 2 and 3, thereby forming a glass film. The aforementioned hydroxyl group on the surface is spontaneously generated generally on a surface of an inorganic light-emitting material, but may be absent depending on the substance constituting the light-emitting material, and, in such case, such hydroxyl group has to be artificially introduced for example by an ultraviolet irradiation.

[chemical structure 2]



[chemical structure 3]



In the foregoing, there has been explained a producing method for the light-emitting material utilizing a main reactant solution as a film forming agent, but the producing method of the invention can also be executed utilizing silicon tetrachloride. In this case, silicon tetrachloride, in an amount of 0.05 - 3.0 wt.% with respect to the fine particles of the light-emitting material, is dissolved in alcohol with a concentration of 50 - 400 g of silicon tetrachloride in 1 liter of the solvent, then fine particles of the light-emitting material in a calculated amount are charged into the solution, and deflocculated and dispersed therein, and while mixing a reaction of a hydroxyl group on the surface of the fine particles of the light-emitting material with silicon tetrachloride is caused, thereby forming a glass film of silicon oxide on the surface of the light-emitting material, and then the layer containing the light-emitting material is separated by a decantation, as in the above-described solvent handling method.

(3) Spray drying step

In case of the aforementioned method utilizing the dispersion medium, the obtained light-emitting material is in a state of mixture with an alcohol, and is therefore spray dried at a temperature of 200°C or lower for example by a spray dryer. In case of the method not utilizing the dispersion medium, the spray drying is not essential, but it is important to disperse crystalline particles of the light-emitting material so that they may not condense.

(Example 1)

[0016] Formation of glass film having moisture and gas barrier property on fine particles of light-emitting material

[0017] A main reactant (containing a catalyst) was prepared in the following manner.

1. 20 g of an organic silicon compound (trade name: "Powder Processing Silicone", manufactured by Toshiba Silicones Co., with a chemical structure 4 and with physical properties shown in Table 1) were dissolved in 200 ml of ethyl alcohol;
2. 0.4 g of boric acid (H_3BO_3) were added to and mixed with thus obtained solution;
3. 0.4 g of ammonium hydrogen fluoride ($\text{NH}_4\text{F} \cdot \text{HF}$) were added and mixed thereto;
4. 1 g of tetrabutyl titanate [$(\text{C}_4\text{H}_9)_4\text{TiO}_4$] was further added and mixed.

[chemical structure 4]

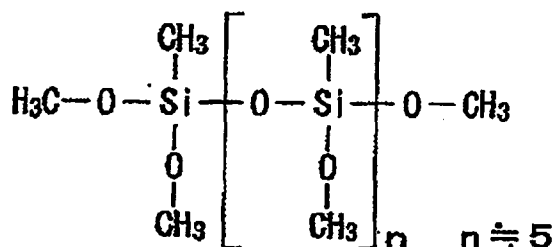


Table 1

Appearance	colorless transparent liquid
Dynamic viscosity (25°C)	3.7 cst

(continued)

Appearance	colorless transparent liquid
Specific gravity (25°C)	1.06
Refractive index (n_D)	1.3829
Flash point	52°C

[0018] A process utilizing a dispersion medium was conducted in the following procedure.

[0019] At first 200 g of fine particles of a light-emitting material (phosphor-based light-emitting material, manufactured by Ecran Ltd.) to be coated with a film were added to 1 liter of ethyl alcohol, then completely dispersed in a Henshel mixer, and 22 g of the aforementioned main reactant solution (organic silicon compound being about 1 wt.% of the light-emitting material) were dropwise added to the obtained dispersion and well agitated. Then the mixer was once stopped, and the dispersion liquid containing the particles was transferred to a decantation vessel and subjected to decantation by being kept still. As the fine particles of the light-emitting material on which a barrier film was formed floated upward, this particle layer was separated from a solution layer, and was spray dried with a spray dryer to obtain a light-emitting material of the invention.

[0020] The barrier property of the film was tested in a water repellency test. 200 ml of water are charged in a 300-ml beaker, and powder is poured therein, whereupon all untreated fine particles precipitate. Fine particles of a high level of glass film formation, that is, with the main reactant constituting 1.0 - 3.0 wt.% of the particle, floated upward without causing turbidity in the aqueous layer, while those with the main reactant constituting 0.05 - 1.0 wt.% showed a difference in that there was turbidity of the aqueous layer when the particles floated upward.

[0021] A process not utilizing a dispersion medium was conducted in the following procedure.

[0022] 200 g of fine particles of a light-emitting material (same as above) to be coated with a film were well agitated in the air by a Henshel mixer to obtain a floating fluid state, and 22 g of the aforementioned main reactant solution (proportion of organic silicon compound to the light-emitting material being same as in the method utilizing the dispersion medium) were sprayed to the fine powder. A sufficient agitation was executed after the spraying, then the mixer was stopped, and the powder was taken out from the mixer to obtain a light-emitting material of the invention.

[0023] In the fine particles of the light-emitting material, obtained by the method utilizing the dispersion medium in Example 1, an X-ray diffraction of the glass film on the particle surface showed only a single large peak (center at 504 nm) corresponding to an amorphous phase, and thus indicated that a glass film was formed. The glass film had a thickness of 0.03 μm in an EPMA measurement.

[0024] Fig. 1 shows a change in time of the luminance of the light-emitting material. A curve 1 indicates the change with time of the luminance of a light-emitting material prepared in Example 1, measured with a spectral teleluminance meter. Also a curve 2 indicates the change with time of the luminance of the same light-emitting material which has not been processed, measured in the same manner. Both showed a decrease in the luminance by about 20% after 50 hrs., but the unprocessed material showed a further decrease in the luminance down to about 40% after 20,000 hrs. On the other hand, the light-emitting material of Example 1 maintained almost the same luminance even after 20,000 hrs. as indicated by the curve 1.

[0025] Fig. 2 is an electron photomicrograph of the fine particles of the light-emitting material prepared in Example 1.

(Example 2)

[0026] Fine particles of a phosphor-based phosphorescent material (green, 60 μm , manufactured by Ecran Ltd.) were processed in the identical manner as in Example 1 to obtain particles coated with a glass film of the invention on the surface, and such particles and particles of the same material which were not thus processed were compared with regard to change with time of the luminance of afterglow when irradiated with 4 black lamps of 20 W for 30 minutes. Results are shown in Fig. 3. The measurement was conducted with an equipment LS-100 manufactured by Minolta Co., at a measuring distance of 100 cm and at a vertical measuring angle. In Fig. 3, the light-accumulating material subjected to the glass film forming process (◆) had an afterglow luminance of 5 mcd (millicandela)/ m^2 even after 16 hours, while the unprocessed same material (▲) showed a rapid decrease in the afterglow luminance after 8 hours.

[0027] As explained above, the manufacturing method of the invention for the light-emitting material can form a thin glass film on the surface of fine particles in a liquid phase or in a gaseous phase, and the organic silicon compound or silicon tetrachloride employed in the method can form a thin protective film 0.05 - 3 wt.% that of the light-emitting material, which is strong, free from pinholes and does not decrease the luminance in an EL display, and can provide a moisture barrier and a gas barrier. Thus the present invention is applicable not only to a light-emitting material for a flat EL display but also in areas requiring a moisture barrier and a gas barrier such as pharmaceuticals, precision parts, semiconductor

devices, high-voltage insulation devices and the like.

Claims

1. A manufacturing method of a light-emitting material comprising steps of:

adding boron ions and halogen ions in this order to an alcohol solution of an organic silicon compound having a hydroxyl group and a methoxyl group and conducting hydrolysis/dehydration condensation to adjust the solution of the reaction product;
dispersing fine particles of a light-emitting material in alcohol while mixing;
adding dropwise the solution of the reaction product into the alcohol to form a glass film of the reaction product on a surface of the fine particles of the light-emitting material;
and then separating the fine particles and spray drying the fine particles at a temperature of 200°C or lower.

2. A manufacturing method of a light-emitting material comprising steps of:

adding boron ions and halogen ions in this order to an alcohol solution of an organic silicon compound having a hydroxyl group and a methoxyl group and conducting hydrolysis/dehydration condensation to obtain a solution of a reaction product;
spraying the solution of the reaction product to fine particles of a light-emitting material dispersed and floating in a mixer to form a glass film of the reaction product on a surface of the fine particles of the light-emitting material; and
then spray drying the fine particles at a temperature of 200°C or lower.

3. A manufacturing method of a light-emitting material according to claim 1 or 2, wherein alkyl titanate or titanium chloride serving as an accelerator for the hydrolysis/dehydration condensation is added.

4. A manufacturing method of a light-emitting material according to claim 1 or 2, wherein a compound having an isocyanate group is added so as to be 1 - 10 wt. % of the organic silicon compound to promote gelation for increasing viscosity, thereby accelerating the hydrolysis/dehydration condensation.

5. A manufacturing method of a light-emitting material comprising steps of:

dissolving, in alcohol, silicon tetrachloride so as to be 0.05 - 3.0 wt. % of the fine particles of a light-emitting material;
deflocculating and dispersing the fine particles of the light-emitting material in the solution;
causing a hydroxyl group on the surface of the fine particles of the light-emitting material to react with silicon tetrachloride while mixing to form a glass film of silicon oxide on the surface of the fine particles;
and then separating the fine particles and spray drying the fine particles at a temperature of 200°C or lower.

Fig. 1

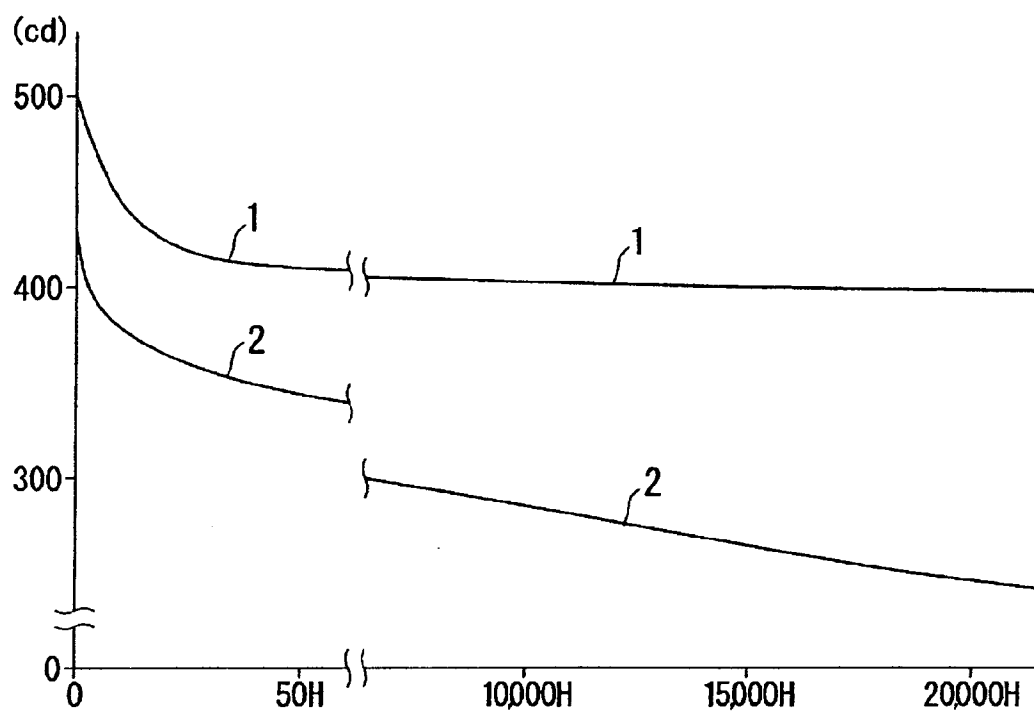


Fig.2

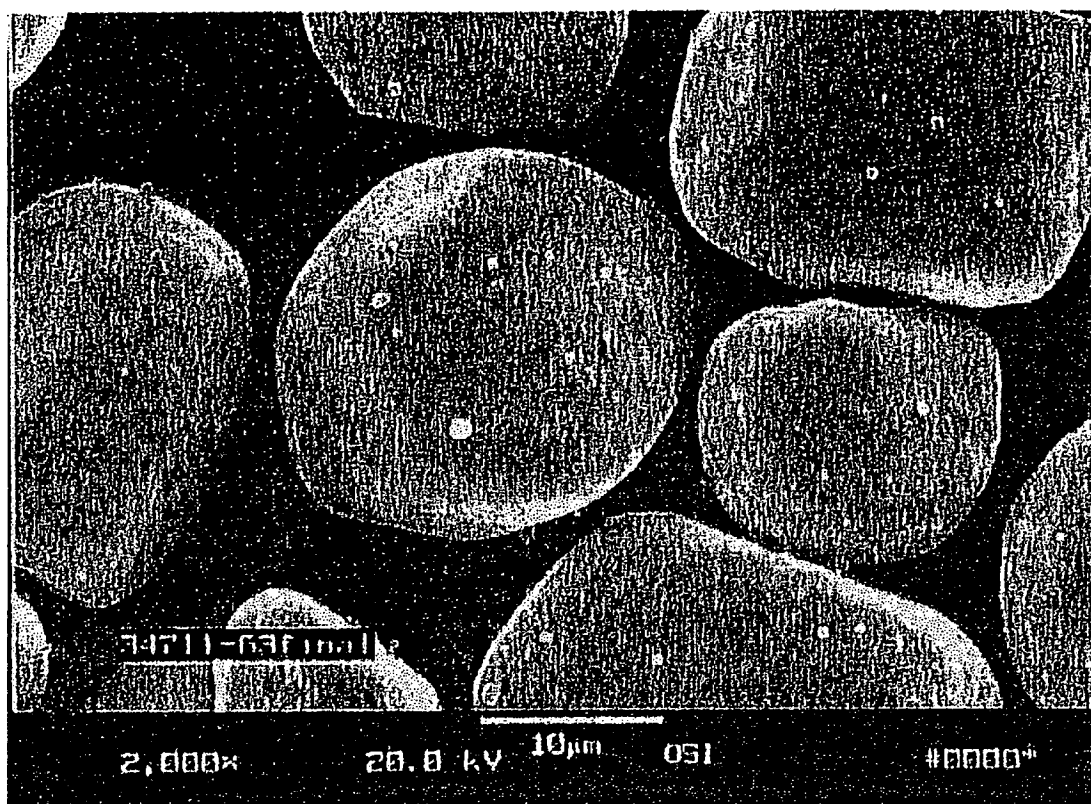
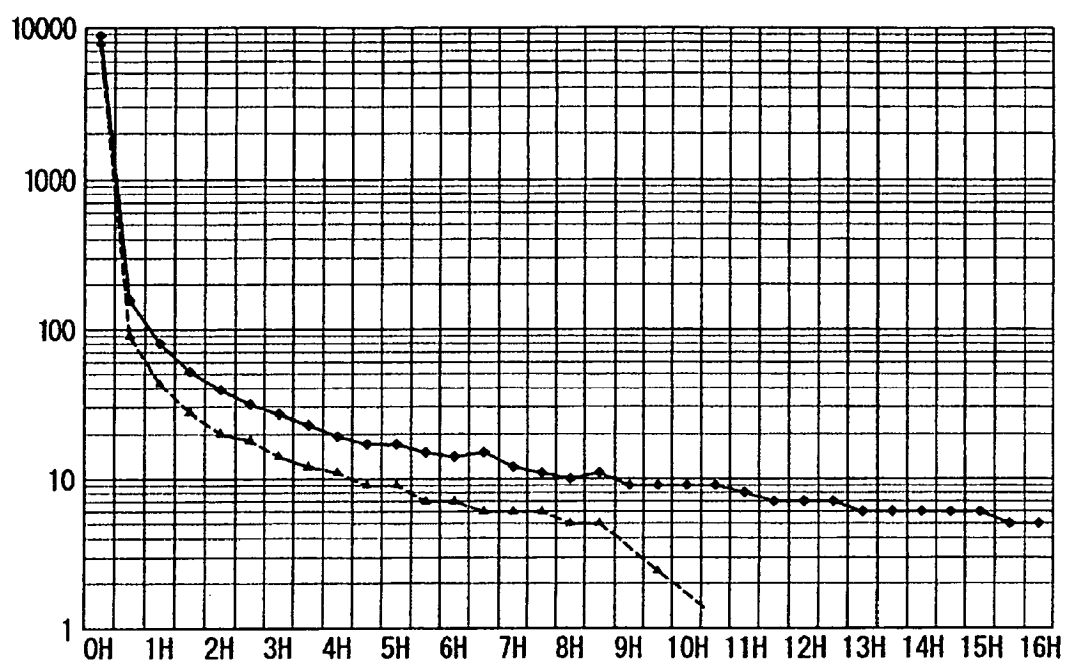


Fig. 3





European Patent
Office

EUROPEAN SEARCH REPORT

Application Number
EP 05 00 9354

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.7)
Y	US 6 591 634 B1 (MORIZANE TOSHINORI) 15 July 2003 (2003-07-15) Method prod. thin flim: 1-hydrolyz. of metal alkoxide, with boron ions and halogen ions, dehydration and condensation 2- react. with mat.t to be coated 3- vitrif. at temp. <200 Si(OEt) ₄ , no OH, no OMe* claims 1-3 * * example 3 * * column 1, lines 13-19,63-67 * * column 2, lines 1-25,34-63 * * column 3, lines 1-10,39-53 * * column 4, lines 15-30,39-50 * * column 5, lines 15-35 *	1-5	C09K11/02
Y	US 6 177 030 B1 (SHOJI TAKEHIKO ET AL) 23 January 2001 (2001-01-23) Method for prep. phosphor with protective coating: 1- Form. phosphor 2- Coat. with metal oxide and silance coupling agent (dry or in sol.) 3-Separate and dry * claims 1,5,6 * * example 1 * * column 1, lines 6-14 * * column 5, lines 17-30 * * column 6, lines 5-11 * * column 10, lines 26-40 * * columns 10-11, lines 1-68 * * column 11, lines 40-58 * * column 12, lines 13-21 *	1-5	TECHNICAL FIELDS SEARCHED (Int.Cl.7) C09K C09C
The present search report has been drawn up for all claims			
Place of search The Hague		Date of completion of the search 18 July 2005	Examiner Vanier, C
CATEGORY OF CITED DOCUMENTS X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document	

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EPO FORM 1503 03 82 (P04C01)



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EUROPEAN SEARCH REPORT

Application Number
EP 05 00 9354

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.7)
A	EP 0 455 401 A (MINNESOTA MINING AND MANUFACTURING COMPANY) 6 November 1991 (1991-11-06) Method prep. encapsul. phos. Moisture and oxidation protec. coating. Low temp. vapor hydrolysis. Use SiCl₄ in combination with TiCl₄* figure 2 * * claims 6-8 * * page 3, lines 33-59 * * page 5, lines 15-22 * * page 6, lines 10-14,39-49 * -----	5	
A	US 5 087 523 A (SIGAI ET AL) 11 February 1992 (1992-02-11) Chemical vapor deposition TMOS, TEOS* figure 5 * * example 1 * * column 3, lines 7-19 * * column 4, lines 15-61 * -----	1-5	
The present search report has been drawn up for all claims			TECHNICAL FIELDS SEARCHED (Int.Cl.7)
Place of search		Date of completion of the search	Examiner
The Hague		18 July 2005	Vanier, C
CATEGORY OF CITED DOCUMENTS X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document			

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**ANNEX TO THE EUROPEAN SEARCH REPORT
ON EUROPEAN PATENT APPLICATION NO.**

EP 05 00 9354

This annex lists the patent family members relating to the patent documents cited in the above-mentioned European search report. The members are as contained in the European Patent Office EDP file on
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18-07-2005

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US 6591634 B1	15-07-2003	US 6408648 B1	25-06-2002
US 6177030 B1	23-01-2001	JP 11344598 A	14-12-1999
		JP 2000144128 A	26-05-2000
		JP 2000160155 A	13-06-2000
EP 0455401 A	06-11-1991	US 5156885 A	20-10-1992
		CA 2035294 A1	26-10-1991
		DE 69120864 D1	22-08-1996
		DE 69120864 T2	06-02-1997
		EP 0455401 A2	06-11-1991
		JP 2756044 B2	25-05-1998
		JP 4230996 A	19-08-1992
		US 5439705 A	08-08-1995
		US 5908698 A	01-06-1999
		US 5418062 A	23-05-1995
US 5087523 A	11-02-1992	US 5051277 A	24-09-1991
		AU 642967 B2	04-11-1993
		AU 7145291 A	05-08-1991
		BR 9104189 A	24-12-1991
		CA 2034423 A1	23-07-1991
		CA 2360398 A1	23-07-1991
		DE 69112508 D1	05-10-1995
		DE 69112508 T2	08-02-1996
		EP 0464179 A1	08-01-1992
		FI 98925 B	30-05-1997
		JP 3001630 B2	24-01-2000
		JP 4504441 T	06-08-1992
		MX 171701 B	10-11-1993
		WO 9110715 A1	25-07-1991
		US 5309069 A	03-05-1994

REFERENCES CITED IN THE DESCRIPTION

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Patent documents cited in the description

- JP 2001354668 A [0006]
- JP 2003234179 A [0006]

Non-patent literature cited in the description

- **TOSHIO INOKUCHI.** Electroluminescent Display. Sangyo Tosho Co, 1991 [0006]
- **JUNJI KIDO.** Yuki EL no subete (all aspects of organic EL. Nippon Jitsugyo Shuppan Co, 2003 [0006]
- **MASAYUKI KAWAMURA.** Yokuwaku yuki EL display. Dempa Shimbun Co, 2003 [0006]

专利名称(译)	发光材料的制造方法		
公开(公告)号	EP1717289A1	公开(公告)日	2006-11-02
申请号	EP2005009354	申请日	2005-04-28
[标]申请(专利权)人(译)	森实茂		
申请(专利权)人(译)	MORIZANE茂		
当前申请(专利权)人(译)	MORIZANE茂		
[标]发明人	MORIZANE SHIGERU		
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IPC分类号	C09K11/02		
CPC分类号	C09K11/025 C09K11/08		
其他公开文献	EP1717289B1		
外部链接	Espacenet		

摘要(译)

本发明提供一种使用寿命长的发光材料的制造方法，其包括将硼离子和卤素离子依次加入到具有羟基和甲氧基的有机硅化合物的醇溶液中，得到通过在常温下水解/脱水缩合反应产物的溶液，然后在搅拌下将发光材料的细颗粒分散在醇中，将反应产物的溶液滴加到这种醇中以形成反应产物的玻璃膜。在发光材料的细颗粒表面上，然后分离细颗粒并在200°C或更低的温度下喷雾干燥细颗粒。

Fig.2

