(19) Europäisches Patentamt
European Patent Office
Office européen des brevets

(11) **EP 2 431 446 B1**

(12) **EUROPEAN PATENT SPECIFICATION**

(45) Date of publication and mention of the grant of the patent:
06.11.2013 Bulletin 2013/45

(21) Application number: **09844492.0**

(22) Date of filing: **11.05.2009**

(51) Int Cl.:
C09K 11/78 *(2006.01)*

(86) International application number:
PCT/CN2009/071719

(87) International publication number:
WO 2010/130075 (18.11.2010 Gazette 2010/46)

(54) FULL-COLOR LIGHT-EMITTING MATERIAL AND PREPARATION METHOD THEREOF

VOLLTÖNIGES LICHT EMITTIERENDES MATERIAL UND HERSTELLUNGSVERFAHREN DAFÜR

MATERIAU ELECTROLUMINESCENT PLEINE COULEUR ET SON PROCEDE DE PREPARATION

(84) Designated Contracting States:
AT BE BG CH CY CZ DE DK EE ES FI FR GB GR HR HU IE IS IT LI LT LU LV MC MK MT NL NO PL PT RO SE SI SK TR

(43) Date of publication of application:
21.03.2012 Bulletin 2012/12

(73) Proprietor: **Ocean's King Lighting Science&Technology Co., Ltd.**
Guangdong 518052 (CN)

(72) Inventors:
- **ZHOU, Mingjie**
  Nanshan District Shenzhen
  Guangdong 518052 (CN)
- **MA, Wenbo**
  Nanshan District Shenzhen
  Guangdong 518052 (CN)
- **SHI, Zhaopu**
  Nanshan District Shenzhen
  Guangdong 518052 (CN)

(74) Representative: **Bird, William Edward**
Bird Goën & Co
Wetenschapspark Arenberg
Gaston Geenslaan 9
3001 Heverlee (BE)

(56) References cited:
EP-A1- 1 979 438
CN-A- 1 420 154
CN-A- 1 482 208
JP-A- 2006 257 133
US-A1- 2009 026 477
US-A1- 2009 026 918

EP 2 431 446 B1

Description

FIELD OF THE INVENTION

[0001] The present invention belongs to technical fields of photoelectronic display and lighting, and relates to a fluorescent material, more particularly, to a full-color light-emitting material capable of emitting red-blue-green (R-G-B) full-color light directly and preparation method thereof.

BACKGROUND OF THE INVENTION

[0002] With the development of semiconductor lighting technology (LED), such revolutionary new light source has come into our daily life gradually. When the third generation semiconductor material gallium nitride is used as the semiconductor lighting source, its power consumption is only one-tenth of that of a common incandescent lamp under the same brightness; its lifetime can reach more than 1 million hours as well. As a new-type lighting technology, LED can be applied into varieties of fields such as indication, display, decoration, backlight and general lighting due to its numerous advantages including energy conservation, green environmental protection and flexible application etc., which is to bring about a revolution in the lighting field. Therefore, there is an urgent need for an efficient fluorescent material, which is capable of converting the blue-purple light emitted by light-emitting components including LED into the visible light, thus achieving white-light and multi-color light-emitting devices.

[0003] In the prior art, a main approach for achieving the white light emission of LED is through the cooperation of a blue-light LED chip and a rare earth garnet yellow fluorescent powder (such as YAG:Ce^{3+} or TAG: Ce^{3+}) which is excited by cerium. However, the white light spectrum implemented by such method is short of green and red element, thus leading to two obvious disadvantages namely low color rendering property and high color temperature. In order to solve the disadvantages mentioned above, on one hand, green fluorescent powder or red fluorescent powder is doped into the yellow fluorescent powder excited by blue-light LED chip. On the other hand, an ultraviolent LED chip is used for exciting the red-green-blue tribasic fluorescent powder to enhance the color rendering property and regulate the color temperature. Although the above-mentioned two approaches can solve the problems of color rendering property and color temperature of light source relatively well, it still is required to encapsulate the LED chip after mixing various fluorescent powders with resin. During this process, the problem lies in that different kinds of fluorescent powders may not be mixed uniformly, which causing the non-uniform color of produced white light and influencing its practicability seriously.

[0004] Examples of germanates combined with other phosphors in a light emitting device are disclosed e.g. in US 2009/0026477 and WO 2007/080541.

SUMMARY OF THE INVENTION

[0005] The objective of the present invention is to provide a full-color light-emitting material which is able to emit a red-green-blue (R-G-B) full-color light directly without the need of doping any other substances, has good luminescent property and adapts for being excited by light-emitting components in ultraviolent zone (240~410nm), aiming at the drawbacks that green fluorescent powder or red fluorescent powder should be added in the approach for achieving the white light emission of LED of prior art through the cooperation of a blue-light LED chip and a rare earth garnet yellow fluorescent powder which is excited by cerium, causing the non-uniform color of produced white light and influencing its practicability seriously when various fluorescent powders are not mixed uniformly.

[0006] Another objective of the present invention is to provide a preparation method for full-color light-emitting material which has simple process and stable product quality.

[0007] According to an aspect, a full-color light-emitting material is provided, which is a compound of following general formula $(Y_{1-x-y-z}A_xB_yC_z)_2GeO_5$, wherein x is $0<x\leq0.05$, y is $0<y\leq0.15$, z is $0<z\leq0.15$ and x:y:z=1:1~10:1~10; A is one selected from a group of Tm and Ce, B is one selected from a group of Tb, Ho, Er and Dy, and C is one selected from a group of Eu, Pr and Sm.

[0008] Ranges of x, y and z are preferably $0<x\leq0.03$, $0<y\leq0.10$ and $0<z\leq0.10$, respectively.

[0009] The ratio of x:y:z is preferably 1:1~6:1~6.

[0010] According to an aspect, a preparation method for full-color light-emitting material is provided, which comprising taking an oxide, carbonate, oxalate, acetate, nitrate or halide of Y and Ge together with an oxide, carbonate, oxalate, acetate, nitrate or halide of A, B and C as raw materials, grinding the raw materials uniformly, sintering the raw materials at 1300~1500 °C for 6~24 h, cooling down the raw materials to room temperature and then obtaining the full-color light-emitting material; wherein A is one selected from a group of Tm and Ce, B is one selected from a group of Tb, Ho, Er and Dy, and C is one selected from a group of Eu, Pr and Sm.

[0011] In the preparation method for full-color light-emitting material, the method preferably comprises grinding the raw materials uniformly in a mortar, sintering the raw materials at 1350-1450 °C for 8~15 h, cooling down the raw materials to room temperature and then obtaining the full-color light-emitting material.

[0012] In the preparation method for full-color light-emitting material, the method comprises weighting the raw materials in a stoichiometric ratio of each element in a chemical formula $(Y_{1-x-y-z}A_xB_yC_z)_2GeO_5$, that is, weighing the raw materials in accordance with a molar ratio of each element in the chemical formula; wherein ranges of x, y and z are respectively $0<x\leq0.05$, $0<y\leq0.15$

and $0<z\leq0.15$, and ratio of x:y:z is 1:1~10:1~10.

[0013] In the preparation method for full-color light-emitting material, the range of x, y and z is preferably $0<x\leq0.03$, $0<y\leq0.10$ and $0<z\leq0.10$, respectively.

[0014] In the preparation method for full-color light-emitting material, the ratio of x:y:z is preferably 1:1~6:1~6.

[0015] In the raw material, Purity of the oxide, carbonate, oxalate, acetate, nitrate or halide is no less than analytic purity.

[0016] The light-emitting material prepared in the present invention uses germanate doped with rare earth. Accordingly, the light-emitting material is capable of emitting a full-color light when excited in the ultraviolet zone (240~410 nm) and has good luminescent property due to the addition of rare earth. Besides, an ideal white lighting can be achieved by adapting the ratio of the doping rare earth in the germanate.

[0017] The preparation method for full-color light-emitting material of the present invention has simple process, stable product stability, strong practicability and wide range of application.

BRIEF DESCRIPTION OF THE DRAWINGS

[0018] The present invention will be further described with reference to the accompanying drawings and embodiments in the following. In the Figures:

Figure 1 is the emission spectrum of the full-color light-emitting material $(Y_{0.97}Tm_{0.01}Tb_{0.01}Eu_{0.01})_2GeO_5$ prepared in the example 1;

Figure 2 is the emission spectrum of the full-color light-emitting material $(Y_{0.945}Tm_{0.01}Tb_{0.02}Eu_{0.025})_2GeO_5$ prepared in the example 9;

Figure 3 is the emission spectrum of the full-color light-emitting material $(Y_{0.915}Tm_{0.01}Tb_{0.04}Eu_{0.035})_2GeO_5$ prepared in the example 11;

wherein the excitation wavelength of the emission spectrum is 360 nm.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENT

[0019] The full-color light-emitting material of the present invention is the compound of following general formula: $(Y_{1-x-y-z}A_xB_yC_z)_2GeO_5$, wherein the ranges of x, y and z are respectively $0<x\leq0.05$, $0<y\leq0.15$ and $0<z\leq0.15$, and the ratio of x:y:z is 1:1~10:1~10. Among them, A is one selected from the group of Tm and Ce, B is one selected from the group of Tb, Ho, Er and Dy, and C is one selected from the group of Eu, Pr and Sm. The ranges of x, y and z are preferably $0<x\leq0.03$, $0<y\leq0.10$ and $0<z\leq0.10$, respectively. The ratio of x:y:z is preferably 1:1~6:1~6.

[0020] A preparation method for full-color light-emitting material is provided, which comprises taking the oxide, carbonate, oxalate, acetate, nitrate or halide of Y and Ge together with the oxide, carbonate, oxalate, acetate, nitrate or halide of A, B and C as the raw materials, grinding the raw materials uniformly, sintering the raw materials at 1300~1500 °C for 6~24 h, cooling down the raw materials to room temperature and then obtaining the full-color light-emitting material; wherein A is one selected from the group of Tm and Ce, B is one selected from the group of Tb, Ho, Er and Dy, and C is one selected from the group of Eu, Pr and Sm.

[0021] In the preparation method for full-color light-emitting material, the method preferably comprises grinding the raw material uniformly in a mortar, sintering the uniform raw materials at 1350~1450 °C for 8~15 h, cooling down the raw materials to room temperature and then obtaining the full-color light-emitting material.

[0022] In the preparation method for full-color light-emitting material, the method comprises weighting the raw materials in the stoichiometric ratio of each element in the chemical formula $(Y_{1-x-y-z}A_xB_yC_z)_2GeO_5$, that is, weighing the raw material in accordance with the molar ratio of each element in the formula; wherein the ranges of x, y and z are respectively $0<x\leq0.05$, $0<y\leq0.15$ and $0<z\leq0.15$, and the ratio of x:y:z is 1:1~10:1~10. Preferably, the ranges of x, y and z are respectively $0<x\leq0.03$, $0<y\leq0.10$ and $0<z\leq0.10$, and the ratio of x:y:z is 1:1~6:1~6.

[0023] In the raw material, the purity of the oxide, carbonate, oxalate, acetate, nitrate or halide is preferably no less than analytic purity.

[0024] The present invention will be further explained in detail according to some examples in the following.

Example 1: $(Y_{0.97}Tm_{0.01}Tb_{0.01}Eu_{0.01})_2GeO_5$ prepared by high temperature solid-state method

[0025] At room temperature, 0.97 mmol Y_2O_3, 0.01 mmol Tm_2O_3, 0.005 mmol Tb_4O_7, 0.01 mmol Eu_2O_3 and 1 mmol GeO_2 are placed in an agate mortar and grinded to be uniform. Then the obtained powder is transferred to a corundum crucible and then placed in a high temperature box-type furnace, in which the powder is sintered at 1350 □ for 15 h. Subsequently, the yielded product is cooled down to room temperature and further grinded in a mortar. Then a full-color light-emitting material $(Y_{0.97}Tm_{0.01}Tb_{0.01}Eu_{0.01})_2GeO_5$ is obtained. As shown in figure 1, it is the emission spectrum of the full-color light-emitting material $(Y_{0.97}Tm_{0.01}Tb_{0.01}Eu_{0.01})_2GeO_5$ prepared in the example. As shown in the figure 1, when excited at 360 nm, the full-color light-emitting material prepared in the example emits a blue light at 455, 460 and 487 nm, a yellow-green light at 544, 548, 580 and 587 nm, as well as an orange red light at 594, 611, 618 and 622 nm, thus realizing the full-color composite luminescence. The preparation method above has simple

steps and stable product quality.

Example 2: $(Y_{0.97}Tm_{0.01}Tb_{0.01}Eu_{0.01})_2GeO_5$ prepared by high temperature solid-state method

[0026] At room temperature, 1.94 mmol $Y(NO_3)_3$, 0.02 mmol $Tm(NO_3)_3$, 0.02 mmol $Tb(NO_3)_3$, 0.02 mmol $Eu(NO_3)_3$ and 1 mmol GeO_2 are placed in an agate mortar and grinded to be uniform. Then the obtained powder is transferred to a corundum crucible and placed in a high temperature box-type furnace, in which the powder is sintered at 1300 0 for 24 h. Subsequently, the yielded product is cooled down to room temperature and further grinded in a mortar. Then a full-color light-emitting material $(Y_{0.97}Tm_{0.01}Tb_{0.01}Eu_{0.01})_2GeO_5$ is obtained.

Example 3: $(Y_{0.945}Tm_{0.01}Dy_{0.02}Eu_{0.025})_2GeO_5$ prepared by high temperature solid-state method

[0027] At room temperature, 0.945 mmol Y_2O_3, 0.01 mmol Tm_2O_3, 0.02 mmol Dy_2O_3, 0.025 mmol Eu_2O_3 and 1 mmol GeO_2 are placed in an agate mortar and grinded to be uniform. Then the obtained powder is transferred to a corundum crucible and placed in a high temperature box-type furnace, in which the powder is sintered at 1450 □ for 8 h. Subsequently, the yielded product is cooled down to room temperature and further grinded in a mortar. Then a full-color light-emitting material $(Y_{0.945}Tm_{0.01}Dy_{0.02}Eu_{0.025})_2GeO_5$ is obtained.

Example 4: $(Y_{0.945}Tm_{0.01}Ho_{0.015}Eu_{0.03})_2GeO_5$ prepared by high temperature solid-state method

[0028] At room temperature, 0.945 mmol Y_2O_3, 0.01 mmol Tm_2O_3, 0.015 mmol Ho_2O_3, 0.03 mmol Eu_2O_3 and 1 mmol GeO_2 are placed in an agate mortar and grinded to be uniform. Then the obtained powder is transferred to a corundum crucible and placed in a high temperature box-type furnace, in which the powder is sintered at 1500 □ for 6 h. Subsequently, the yielded product is cooled down to room temperature and further grinded in a mortar. Then a full-color light-emitting material $(Y_{0.945}Tm_{0.01}Ho_{0.015}Eu_{0.03})_2GeO_5$ is obtained.

Example 5: $(Y_{0.94}Tm_{0.01}Er_{0.025}Eu_{0.025})_2GeO_5$ prepared by high temperature solid-state method

[0029] At room temperature, 0.94 mmol Y_2O_3, 0.01 mmol Tm_2O_3, 0.025 mmol Er_2O_3, 0.025 mmol Eu_2O_3 and 1 mmol GeO_2 are placed in an agate mortar and grinded to be uniform. Then the obtained powder is transferred to a corundum crucible and placed in a high temperature box-type furnace, in which the powder is sintered at 1400 □ for 11 h. Subsequently, the yielded product is cooled down to room temperature and further grinded in a mortar. Then a full-color light-emitting material $(Y_{0.94}Tm_{0.01}Er_{0.025}Eu_{0.025})_2GeO_5$ is obtained.

Example 6: $(Y_{0.95}Tm_{0.01}Tb_{0.02}Sm_{0.02})_2GeO_5$ prepared by high temperature solid-state method

[0030] At room temperature, 0.95 mmol Y_2O_3 0.01 mmol Tm_2O_3, 0.01 mmol Tb_4O_7, 0.02 mmol Sm_2O_3 and 1 mmol GeO_2 are placed in an agate mortar and grinded to be uniform. The remaining steps are the same as those in example 1. Then a full-color light-emitting material $(Y_{0\cdot95}Tm_{0.01}Tb_{0.02}Sm_{0.02})_2GeO_5$ is obtained.

Example 7: $(Y_{0\cdot915}Tm_{0.015}Tb_{0.04}Pr_{0.03})_2GeO_5$ prepared by high temperature solid-state method

[0031] At room temperature, 0.915 mmol Y_2O_3, 0.015 mmol Tm_2O_3, 0.02 mmol Tb_4O_7, 0.01 mmol Pr_6O_{11} and 1 mmol GeO_2 are placed in an agate mortar and grinded to be uniform. The remaining steps are the same as those in example 1. Then a full-color light-emitting material $(Y_{0.915}Tm_{0.015}Tb_{0.04}Pr_{0.03})_2GeO_5$ is obtained.

Example 8: $(Y_{0.93}Ce_{0.01}Tb_{0.03}Eu_{0.03})_2GeO_5$ prepared by high temperature solid-state method

[0032] At room temperature, 0.93 mmol Y_2O_3, 0.02 mmol CeO_2 0.015 mmol Tb_4O_7, 0.03 mmol Eu_2O_3 and 1 mmol GeO_2 are placed in an agate mortar and grinded to be uniform. The remaining steps are the same as those in example 1. Then a full-color light-emitting material $(Y_{0.93}Ce_{0.01}Tb_{0.03}Eu_{0.03})_2GeO_5$ is obtained.

Example 9: $(Y_{0.945}Tm_{0.01}Tb_{0.02}Eu_{0.025})_2GeO_5$ prepared by high temperature solid-state method

[0033] At room temperature, 0.945 mmol Y_2O_3, 0.01 mmol Tm_2O_3, 0.01 mmol Tb_4O_7, 0.025 mmol Eu_2O_3 and 1 mmol GeO_2 are placed in an agate mortar and grinded to be uniform. The remaining steps are the same as those in example 1. Then a full-color light-emitting material $(Y_{0.945}Tm_{0.01}Tb_{0.02}Eu_{0.05})_2GeO_5$ is obtained. As shown in figure 2, it is the emission spectrum of the full-color light-emitting material $(Y_{0.945}Tm_{0.01}Tb_{0.02}Eu_{0.025})_2GeO_5$ prepared in the example. As shown in figure 2, when excited at 360 nm, the full-color light-emitting material prepared in the example emits a blue light at 455, 460 and 487 nm, a yellow-green light at 544, 548, 580 and 587 nm, as well as an orange red light at 594, 611, 618 and 622 nm. The color coordinate of the combined light in the example is (0.3364, 0.3282), which is close to that of an ideal white light, i.e. (0.33, 0.33). Thus, a white light emission is achieved.

Example 10: $(Y_{0.92}Tm_{0.01}Tb_{0.04}Eu_{0.03})_2GeO_5$ prepared by high temperature solid-state method

[0034] At room temperature, 0.92 mmol Y_2O_3, 0.01 mmol Tm_2O_3,0.02 mmol Tb_4O_7, 0.03 mmol Eu_2O_3 and 1 mmol GeO_2 are placed in an agate mortar and grinded

to be uniform. The remaining steps are the same as those in example 1. Then a full-color light-emitting material $(Y_{0.92}Tm_{0.01}Tb_{0.04}Eu_{0.03})_2GeO_5$ is obtained.

Example 11: $(Y_{0.915}Tm_{0.01}Tb_{0.04}Eu_{0.03})_2GeO_5$ prepared by high temperature solid-state method

[0035] At room temperature, 0.915 mmol Y_2O_3, 0.01 mmol Tm_2O_3, 0.02 mmol Tb_4O_7, 0.035 mmol Eu_2O_3 and 1 mmol GeO_2 are placed in an agate mortar and grinded to be uniform. The remaining steps are the same as those in example 1. Then a full-color light-emitting material $(Y_{0.915}Tm_{0.01}Tb_{0.04}Eu_{0.035})_2GeO_5$ is obtained. As shown in figure 3, it is the emission spectrum of the full-color light-emitting material $(Y_{0.915}Tm_{0.01}Tb_{0.04}Eu_{0.035})_2GeO_5$ prepared in the example 11. As shown in figure 3 when excited at 360 nm, the full-color light-emitting material prepared in the example emits a blue light at 455, 460 and 487 nm, a yellow-green light at 544, 548, 580 and 587 nm, as well as an orange red light at 594, 611, 618 and 622 nm. The color coordinate of the combined light in the example is (0.3387, 0.3355), which is close to that of an ideal white light, i.e. (0.33, 0.33). Thus, a full-color composite luminescence is achieved.

Example 12: $(Y_{0.88}Tm_{0.01})Tb_{0.05}Eu_{0.06})_2GeO_5$ prepared by high temperature solid-state method

[0036] At room temperature, 0.88 mmol Y_2O_3, 0.01 mmol Tm_2O_3, 0.025 mmol Tb_4O_7, 0.06 mmol Eu_2O_3 and 1 mmol GeO_2 are placed in an agate mortar and grinded to be uniform. The remaining steps are the same as those in example 1. Then a full-color light-emitting material $(Y_{0.88}Tm_{0.01}Tb_{0.01}Eu_{0.06})_2GeO_5$ is obtained.

Example 13: $(Y_{0.79}Ce_{0.01}Tb_{0.1}Eu_{0.1})_2GeO_5$ prepared by high temperature solid-state method

[0037] At room temperature, 1.58 mmol $Y(CH_3COO)_3$, 0.02 mmol $Ce(CH_3COO)_3$, 0.2 mmol $Tb(CH_3COO)_3$, 0.2 mmol $Eu(CH_3COO)_3$ and 1 mmol $Ge(NO_3)_4$ are placed in an agate mortar and grinded to be uniform. The remaining steps are the same as those in example 1. Then a full-color light-emitting material $(Y_{0.79}Ce_{0.01}Tb_{0.1})_2Eu_{0.1}GeO_5$ is obtained.

Example 14: $(Y_{0.815}Tm_{0.015}Er_{0.02}Pr_{0.15})_2GeO_5$ prepared by high temperature solid-state method

[0038] At room temperature, 0.815 mmol $Y_2(CO_3)_3$, 0.015 mmol $Tm_2(CO_3)_3$, 0.02 mmol $Er_2(CO_3)_3$, 0.3 mmol $Pr(CH_3COO)_3$ and 1 mmol $Ge(C_2O_4)_2$ are placed in an agate mortar and grinded to be uniform. The remaining steps are the same as those in example 1. Then a full-color light-emitting material $(Y_{0.815}Tm_{0.015}Er_{0.02}Pr_{0.15})_2GeO_5$ is obtained.

Example 15: $(Y_{0.82}Tm_{0.05}Ho_{0.01}Eu_{0.12})_2GeO_5$ prepared by high temperature solid-state method

[0039] At room temperature, 0.82 mmol $Y_2(C_2O_4)_3$, 0.05 mmol $Tm_2(CO_3)_3$, 0.01 mmol $Ho_2(C_2O_4)_3$, 0.24 mmol $Eu(CH_3COO)_3$ and 1 mmol $Ge(CH_3COO)_4$ are placed in an agate mortar and grinded to be uniform. The remaining steps are the same as those in example 1. Then a full-color light-emitting material

[0040] $(Y_{0.82}Tm_{0.05}Ho_{0.01}Eu_{0.12})_2GeO_5$ is obtained.

Example 16: $(Y_{0.805}Tm_{0.015}Dy_{0.15}Sm_{0.03})_2GeO_5$ prepared by high temperature solid-state method

[0041] At room temperature, 1.61 mmol YCl_3, 0.03 mmol $TmCl_3$, 0.3 mmol $DyCl_3$, 0.06 mmol $Sm(CH_3COO)_3$ and 1 mmol $GeCl_4$ are placed in an agate mortar and grinded to be uniform. The remaining steps are the same as those in example 1. Then a full-color light-emitting material $(Y_{0.805}Tm_{0.015}Dy_{0.15}Sm_{0.03})_2GeO_5$ is obtained.

Claims

1. A full-color light-emitting material, wherein said full-color light-emitting material is a compound of following general formula $(Y_{1-x-y-z}A_xB_yC_z)_2GeO_5$, wherein x is $0<x\leq0.05$, y is $0<y\leq0.15$, z is $0<z\leq0.15$ and x:y:z=1:1~10:1~10; A is one selected from a group of Tm and Ce, B is one selected from a group of Tb, Ho, Er and Dy, and C is one selected from a group of Eu, Pr and Sm.

2. The full-color light-emitting material according to claim 1, wherein ranges of x, y and z are $0<x\leq0.03$, $0<y\leq0.10$ and $0<z\leq0.10$, respectively.

3. The full-color light-emitting material according to claim 1, wherein the ratio of x:y:z is 1:1~6:1~6.

4. A preparation method for the full-color light-emitting materials in claim 1~3, wherein comprising following steps: taking an oxide, carbonate, oxalate, acetate, nitrate or halide of Y and Ge together with an oxide, carbonate, oxalate, acetate, nitrate or halide of A, B and C as raw materials, grinding said raw materials uniformly, sintering said raw materials at 1300~1500 °C for 6-24 h, cooling down said material to room temperature and then obtaining the full-color light-emitting material; wherein A is one selected from a group of Tm and Ce, B is one selected from a group of Tb, Ho, Er and Dy, and C is one selected from a group of Eu, Pr and Sm.

5. The preparation method for full-color light-emitting material according to claim 4, wherein further comprising grinding said raw materials uniformly, sintering said raw materials at 1350-1450 °C for 8~15 h,

cooling down said raw materials to room temperature and then obtaining the full-color light-emitting material.

6. The preparation method for full-color light-emitting material according to claim 4, wherein further comprising weighting said raw materials in a stoichiometric ratio of each element in the chemical formula $(Y_{1-x-y-z}A_xB_yC_z)_2GeO_5$; wherein ranges of x, y and z are respectively $0<x\leq0.05$, $0<y\leq0.15$ and $0<z\leq0.15$,
and ratio of x:y:z is 1:1~10:1~10.

7. The preparation method for full-color light-emitting material according to claim 6, wherein said ranges of x, y and z are $0<x\leq0.03$, $0<y\leq0.10$ and $0<z\leq0.10$, respectively.

8. The preparation method for full-color light-emitting material according to claim 6, wherein said ratio of x:y:z is 1:1~6:1~6.

9. The preparation method for full-color light-emitting material according to claim 6, wherein purity of the oxide, carbonate, oxalate, acetate, nitrate or halide in said raw materials is no less than analytic purity.

Patentansprüche

1. Vollfarben-Leuchtmaterial, wobei das Vollfarben-Leuchtmaterial eine Verbindung der folgenden allgemeinen Formel $(Y_{1-x-y-z}A_xB_yC_z)_2GeO_5$ ist, wobei x gleich $0 < x \leq 0,05$, y gleich $0 < y \leq 0,15$, z gleich $0 < z \leq 0,15$ und x:y:z = 1:1-10:1-10 ist; A ausgewählt ist aus einer Gruppe aus Tm und Ce; B ausgewählt ist aus einer Gruppe aus Tb, Ho, Er und Dy und C ausgewählt ist aus einer Gruppe aus Eu, Pr und Sm.

2. Vollfarben-Leuchtmaterial nach Anspruch 1, wobei Bereiche von x, y und z gleich $0 < x \leq 0,03$, $0 < y \leq 0,10$ bzw. $0 < z \leq 0,10$ sind.

3. Vollfarben-Leuchtmaterial nach Anspruch 1, wobei das Verhältnis von x:y:z 1:1-6:1-6 ist.

4. Herstellungsverfahren für die Vollfarben-Leuchtmaterialien in Anspruch 1 bis 3, das die folgenden Schritte umfasst: Verwenden eines Oxids, Karbonats, Oxalats, Acetats, Nitrats oder Halogenids von Y und Ge gemeinsam mit einem Oxid, Karbonat, Oxalat, Acetat, Nitrat oder Halogenid von A, B und C als Rohmaterialien, gleichförmiges Mahlen der Rohmaterialien, Sintern der Rohmaterialien bei 1300-1500°C über 6-24 h, Abkühlen des Materials auf Raumtemperatur und dann Erhalten des Vollfarben-Leuchtmaterials; wobei A ausgewählt ist aus einer Gruppe aus Tm und Ce; B ausgewählt ist aus einer Gruppe aus Tb, Ho, Er und Dy und C ausgewählt ist aus einer Gruppe aus Eu, Pr und Sm.

5. Herstellungsverfahren für ein Vollfarben-Leuchtmaterial nach Anspruch 4, des Weiteren umfassend das gleichförmige Mahlen der Rohmaterialien, Sintern der Rohmaterialien bei 1350-1450°C über 8-15 h, das Abkühlen des Materials auf Raumtemperatur und dann Erhalten des Vollfarben-Leuchtmaterials.

6. Herstellungsverfahren für ein Vollfarben-Leuchtmaterial nach Anspruch 4, des Weiteren umfassend das Wiegen der Rohmaterialien in einem stöchiometrischen Verhältnis jedes Elements in der chemischen Formel $(Y_{1-x-y-z}A_xB_yC_z)_2GeO_5$, wobei die Bereiche von x, y und z gleich $0 < x \leq 0,05$, $0 < y \leq 0,15$ bzw. $0 < z \leq 0,15$ sind und das Verhältnis von x:y:z gleich 1:1-10:1-10 ist.

7. Herstellungsverfahren für ein Vollfarben-Leuchtmaterial nach Anspruch 6, wobei die Bereiche von x, y und z gleich $0 < x \leq 0,03$, $0 < y \leq 0,10$ bzw. $0 < z \leq 0,10$ sind.

8. Herstellungsverfahren für ein Vollfarben-Leuchtmaterial nach Anspruch 6, wobei das Verhältnis von x:y:z gleich 1:1-6:1-6 ist.

9. Herstellungsverfahren für ein Vollfarben-Leuchtmaterial nach Anspruch 6, wobei die Reinheit des Oxids, Karbonats, Oxalats, Acetats, Nitrats oder Halogenids in den Rohmaterialien nicht geringer als analytische Reinheit ist.

Revendications

1. Matériau électroluminescent coloré, dans lequel ledit matériau électroluminescent coloré est un composé de la formule générale suivante $(Y_{1-x-y-z}A_xB_yC_z)_2GeO_5$, où x est $0 < x \leq 0,05$, y est $0 < y \leq 0,15$, z est $0 < z \leq 0,15$ et x:y:z = 1:1~10:1~10 ; A est un élément choisi dans le groupe composé de Tm et Ce, B est un élément choisi dans le groupe composé de Tb, Ho, Er et Dy, et C est un élément choisi dans le groupe composé de Eu, Pr et Sm.

2. Matériau électroluminescent coloré selon la revendication 1, dans lequel les plages de x, y et z sont respectivement $0 < x \leq 0,03$, $0 < y \leq 0,10$ et $0 < z \leq 0,10$.

3. Matériau électroluminescent coloré selon la revendication 1, dans lequel le rapport x:y:z est 1:1~6:1~6.

4. Procédé de préparation de matériaux électroluminescents colorés selon la revendication 1 à 3, comprenant les étapes suivantes : le choix d'un oxyde,

d'un carbonate, d'un oxalate, d'un acétate, d'un nitrate ou d'un halogénure de Y et de Ge avec un oxyde, un carbonate, un oxalate, un acétate, un nitrate ou un halogénure de A, B et C comme matières premières, le broyage uniforme des dites matières premières, le frittage des dites matières premières à 1300-1500 °C pendant 6-24 h, le refroidissement de ladite matière à une température ambiante et ensuite l'obtention du matériau électroluminescent coloré ; dans lequel A est un élément sélectionné dans le groupe constitué de Tm et Ce, B est un élément sélectionné dans le groupe constitué de Tb, Ho, Er et Dy, et C est un élément sélectionné dans le groupe constitué de Eu, Pr et Sm.

5. Procédé de préparation d'un matériau électroluminescent coloré selon la revendication 4, comprenant en outre le broyage uniforme des dites matières premières, le frittage des dites matières premières à 1350-1450 °C pendant 8-15 h, le refroidissement des dites matières premières à une température ambiante et ensuite l'obtention du matériau électroluminescent coloré.

6. Procédé de préparation pour le matériau électroluminescent coloré selon la revendication 4, comprenant en outre la pondération desdites matières premières dans un rapport stoechiométrique de chaque élément de la formule chimique $(Y_{1-x-y-z}A_xB_yC_z)_2GeO_5$, où x, y et z sont respectivement $0 < x \leq 0,05$, y est $0 < y \leq 0,15$, z est $0 < z \leq 0,15$ et le rapport x:y:z = 1:1~10:1~10.

7. Procédé de préparation pour un matériau électroluminescent en couleur selon la revendication 6, dans lequel lesdites plages de x, y et z sont respectivement $0 < x \leq 0,03$, $0 < y \leq 0,10$ et $0 < z \leq 0,10$.

8. Procédé de préparation pour un matériau électroluminescent coloré selon la revendication 6, dans lequel ledit rapport x:y:z est 1:1~6:1~6.

9. Procédé de préparation pour un matériau électroluminescent coloré selon la revendication 6, dans lequel la pureté de l'oxyde, du carbonate, de l'oxalate, de l'acétate, du nitrate ou de l'halogénure dans lesdites matières premières n'est pas inférieure à la pureté analytique.

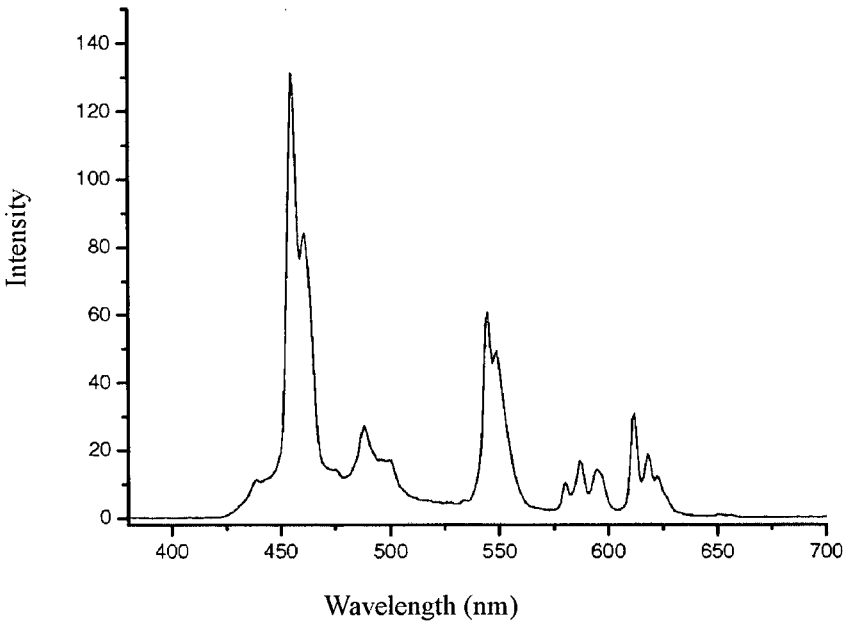

Figure 1

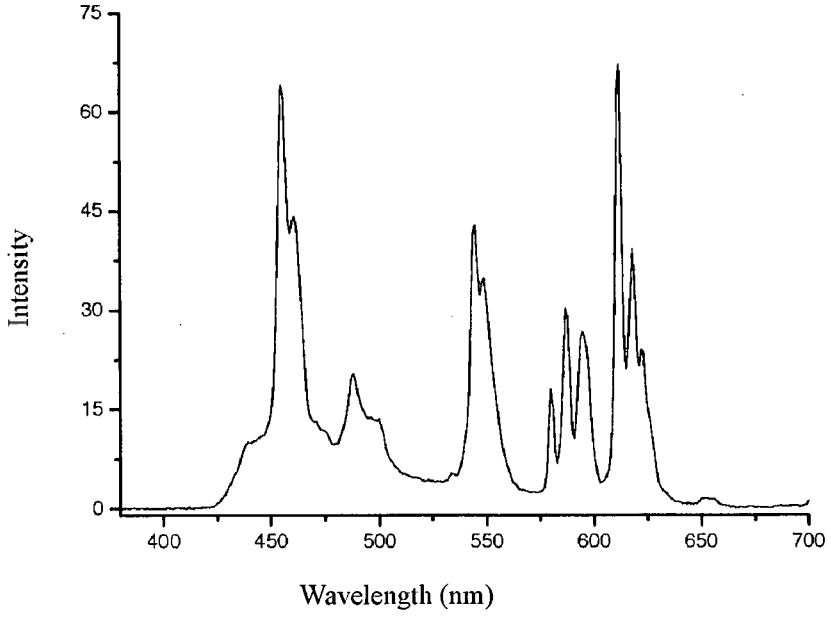

Figure 2

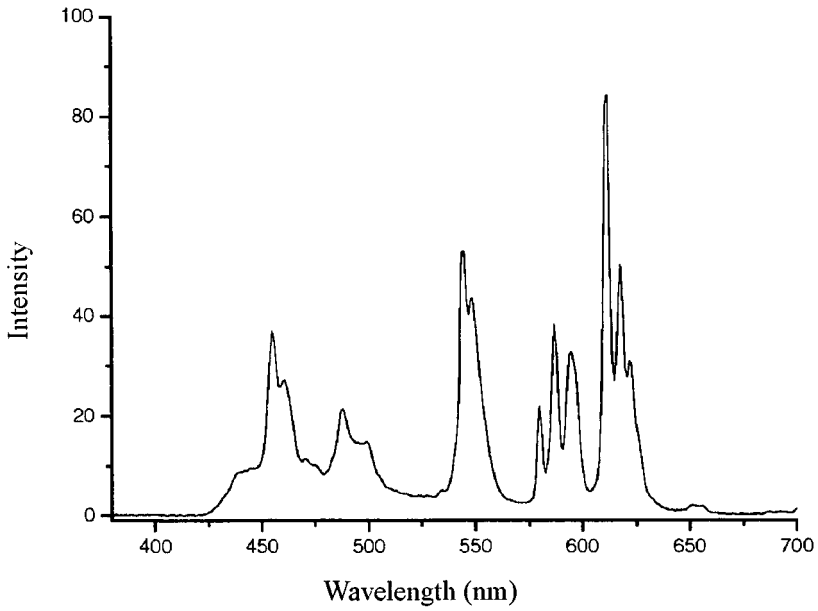
100
80
60
40
20
0
Intensity
400
450
500
550
600
650
700
Wavelength (nm)

Figure 3

REFERENCES CITED IN THE DESCRIPTION

This list of references cited by the applicant is for the reader's convenience only. It does not form part of the European patent document. Even though great care has been taken in compiling the references, errors or omissions cannot be excluded and the EPO disclaims all liability in this regard.

Patent documents cited in the description

- US 20090026477 A **[0004]**
- WO 2007080541 A **[0004]**

专利名称(译)	全彩色发光材料及其制备方法		
公开(公告)号	EP2431446B1	公开(公告)日	2013-11-06
申请号	EP2009844492	申请日	2009-05-11
[标]申请(专利权)人(译)	海洋王照明科技股份有限公司		
申请(专利权)人(译)	海洋王照明科技股份有限公司.		
当前申请(专利权)人(译)	海洋王照明科技股份有限公司.		
[标]发明人	ZHOU MINGJIE MA WENBO SHI ZHAOPU		
发明人	ZHOU, MINGJIE MA, WENBO SHI, ZHAOPU		
IPC分类号	C09K11/78		
CPC分类号	C09K11/7793 C09K11/7775		
其他公开文献	EP2431446A1 EP2431446A4		
外部链接	Espacenet		

摘要(译)

提供一种全色发光材料及其制备方法。发光材料遵循通式化合物（Y1-xy-zAxByCz）2GeO5，其中0 <x≤0.05,0<y≤0.15,0<z≤0.15，x：y：z = 1：1〜10： 1〜10，A是Tm和Ce之一，B是Tb，Ho，Er和Dy之一，C是Eu，Pr和Sm之一。制备方法是：将原料均匀研磨，然后在1300~1500℃下烧结6~24h，将物料冷却至室温，得到产物。本发明提供一种全彩色发光材料，可直接发射红绿蓝全色光，适用于紫外区激发的发光器件，不需要其他掺杂材料。并且提供了一种制备方法，该方法工艺简单，产品质量稳定，适用于全彩色发光材料。

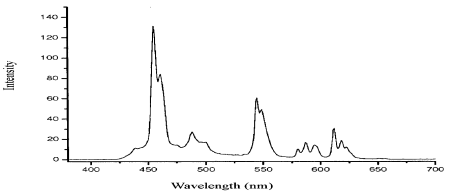

Figure 1

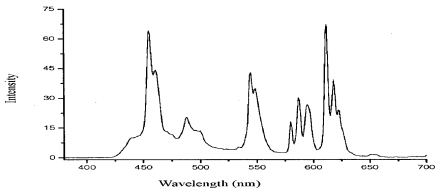

Figure 2