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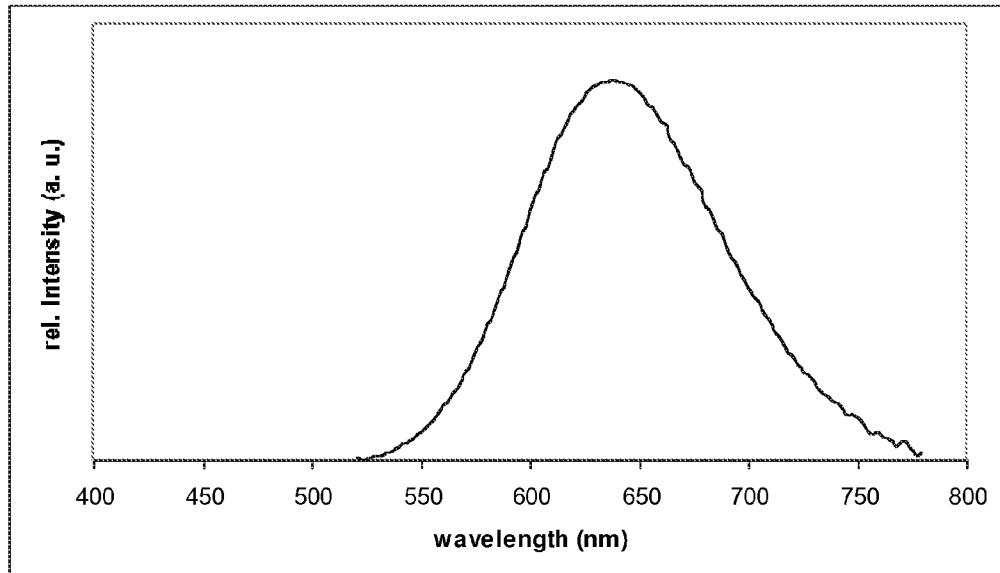
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(54) Title: LIGHT EMITTING DEVICE WITH AN IMPROVED CAALSiN LIGHT CONVERTING MATERIAL



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(57) Abstract: Light emitting device with a light source to emit primary light and a light conversion layer to convert at least a part of the primary light into secondary light comprising a CaAlSiN light converting material with a transmissivity of > 10% to ≤ 80% for a light in the wavelength range from > 580 to ≤ 1000nm.



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DESCRIPTION

Light emitting device with an improved CaAlSiN light converting material

The present invention is directed to light emitting devices, especially to the field
5 of LEDs.

So-called phosphor converted light emitting devices comprising an
electroluminescent light source and a light converting element to absorb light emitted
from the light source at least partially and to re-emit secondary light with longer
10 wavelengths are known from prior art. In recent times, the scope of LEDs comprising a
CaAlSiN light converting phosphor material has been investigated and in this regard it
has been found that such a material can be used in LEDs. An example is e.g. described
in the EP05101834.9 which is incorporated by reference.

15 However, although the phosphor material described in this document shows
good optical characteristics, the demands for LEDs, especially LEDs for use in cars
require in most applications higher effectivity and temperature-stability.

20 It is an object of the present invention to provide a light-emitting device which
comprises a CaAlSiN light converting material with improved effectivity and
temperature stability.

25 This object is solved by a light-emitting device with a light source to emit
primary light and a light conversion layer to convert at least a part of the primary light
into secondary light comprising a CaAlSiN light converting material with a
transmissivity of $\geq 10\%$ to $\leq 80\%$ for a light in the wavelength range from ≥ 580 to \leq
1000nm. Accordingly, a light emitting device, especially a LED is provided,
comprising a CaAlSiN light converting material with a transmissivity of $\geq 10\%$ to $\leq 80\%$

for light in the wavelength area from ≥ 580 to ≤ 1000 nm. The light source may be one or more light sources from the group inorganic LEDs, organic LEDs or laser diodes.

The term “CaAlSiN light converting material” comprises and/or includes
5 especially the following materials:



with $0 \leq x,y,z \leq 1$, $0 \leq a \leq 1$, $0 \leq b < 1$, $0 \leq n \leq 1$ and M being a metal,
according to an embodiment of the present invention selected out of the group
10 comprising Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu or mixtures
thereof

as well as a mixture of these materials with additives which may be added
during ceramic processing. These additives may be incorporated fully or in part into the
15 final material, which then may also be a composite of several chemically different
species (CaAlSiN crystallites embedded into a glassy matrix of slightly different
composition) and particularly include such species known to the art as fluxes. Suitable
fluxes include alkaline earth - or alkaline - metal oxides and fluorides, SiO_2 and the
like.

20

The term “transmissivity” in the sense of the present invention means especially
that $\geq 10\%$, according to an embodiment of the present invention $\geq 20\%$, according to
an embodiment of the present invention $\geq 30\%$, according to an embodiment of the
present invention $\geq 40\%$ and $\leq 80\%$ of the incident light of a wavelength, which cannot
25 be absorbed by the material, is transmitted through the sample for normal incidence in
air. This wavelength is according to an embodiment of the present invention, in the
range of ≥ 580 nm and ≤ 1000 nm.

When using such a CaAlSiN light converting material, the features of the light-
30 emitting device may in most applications be greatly improved (as will for some

applications be described later on).

According to an embodiment of the present invention, the CaAlSiN light converting material emits secondary light in the red visible light wavelength area with a maximum intensity at a wavelength of ≥ 590 nm to ≤ 700 nm. This allows building up a light emitting device with improved characteristics. According to an embodiment of the present invention, the CaAlSiN light converting material has an emission band in the red visible light wavelength area with a maximum of ≥ 600 nm to ≤ 680 nm, according to an embodiment of the present invention, of ≥ 620 nm to ≤ 670 nm.

10

According to an embodiment of the present invention, the CaAlSiN light converting material emits secondary light with a wavelength distribution with a half-width of ≥ 50 nm to ≤ 150 nm. This results in a sharp emission, which allows to further improve the light emitting device. According to an embodiment of the present invention, the CaAlSiN light converting material emits secondary light with a wavelength distribution with a half-width of ≥ 60 nm to ≤ 110 nm.

According to an embodiment of the present invention, the CaAlSiN light converting material has $\geq 97\%$ to $\leq 100\%$ of the theoretical single crystal density. By doing so, the CaAlSiN light converting material shows greatly improved mechanical and optical characteristics compared to materials with a lesser density. According to an embodiment of the present invention, the CaAlSiN light converting material has $\geq 98\%$ to $\leq 100\%$ of the theoretical single crystal density, according to an embodiment of the present invention $\geq 99\%$ to $\leq 100\%$ of the theoretical single crystal.

25

According to an embodiment of the present invention, the CaAlSiN light converting material is a polycrystalline material.

The term "polycrystalline material" in the sense of the present invention means especially a material with a volume density larger than 90 percent of the main

constituent, consisting of more than 80 percent of single crystal domains, with each domain being larger than 0.5 μm in diameter and having different crystallographic orientations. The single crystal domains may be connected by amorphous or glassy material or by additional crystalline constituents.

5

According to an embodiment of the present invention, the CaAlSiN converting material material is a ceramic material.

10 The term “ceramic material” in the sense of the present invention means especially a crystalline or polycrystalline compact material or composite material with a controlled amount of pores or which is free of pores.

15 According to an embodiment of the present invention the thickness of the ceramic material D is $30\mu\text{m} \leq D \leq 5000\ \mu\text{m}$, according to an embodiment of the present invention $60\mu\text{m} \leq D \leq 2000\ \mu\text{m}$ and according to an embodiment of the present invention $80\mu\text{m} \leq D \leq 1000\ \mu\text{m}$.

20 According to an embodiment of the present invention, the shift of the maximum and/or the half-width of the secondary light emission in the red visible light wavelength area of the CaAlSiN light converting material is $\leq 20\ \text{nm}$ over the whole temperature range from $\geq 50^\circ\text{C}$ to $\leq 150^\circ\text{C}$. By doing so, the light emitting device will show a constant behaviour during performance e.g. when used in a car.

25 According to an embodiment of the present invention, the shift of the maximum and/or the half-width in the emission band in the red visible light wavelength area of the CaAlSiN light converting material is $\geq 0\ \text{nm}$ to $\leq 20\ \text{nm}$ over the whole temperature range from $\geq 0^\circ\text{C}$ to $\leq 200^\circ\text{C}$, and according to an embodiment of the present invention, from $\geq -40^\circ\text{C}$ to $\leq 250^\circ\text{C}$.

30 According to an embodiment of the present invention, the shift of the maximum

and/or the half-width in the the secondary light emission in the red visible light wavelength area of the CaAlSiN light converting material is ≥ 5 nm to ≤ 18 nm over the whole temperature range from $\geq 50^{\circ}\text{C}$ to $\leq 150^{\circ}\text{C}$, According to an embodiment of the present invention, $\geq 0^{\circ}\text{C}$ to $\leq 200^{\circ}\text{C}$, and According to an embodiment of the present

5 invention, from $\geq -40^{\circ}\text{C}$ to $\leq 250^{\circ}\text{C}$. According to an embodiment of the present invention, the shift of the maximum and/or the half-width in the secondary light emission in the red visible light wavelength area of the CaAlSiN light converting material is ≥ 10 nm to ≤ 15 nm over the whole temperature range from $\geq 50^{\circ}\text{C}$ to $\leq 150^{\circ}\text{C}$, According to an embodiment of the present invention, $\geq 0^{\circ}\text{C}$ to $\leq 200^{\circ}\text{C}$, and

10 According to an embodiment of the present invention, from $\geq -40^{\circ}\text{C}$ to $\leq 250^{\circ}\text{C}$.

According to an embodiment of the present invention, the CaAlSiN light converting material is selected out of a material comprising $(\text{Ca}_{1-x-y-z}\text{Sr}_x\text{Ba}_y\text{Mg}_z)_{1-n}\text{Al}_{1-a+b}\text{B}_a\text{Si}_{1-b}\text{N}_{3-b}\text{O}_b:\text{RE}_n$ with $0 \leq x, y, z \leq 1$, $0 \leq a \leq 1$, $0 \leq b < 1$, $0 \leq n \leq 1$ and RE selected

15 out of the group comprising Eu, Ce, or mixtures thereof.

According to an embodiment of the present invention, the CaAlSiN light converting material is selected out of a material comprising $(\text{Ca}_{1-x-y-z}\text{Sr}_x\text{Ba}_y\text{Mg}_z)_{1-n}\text{Al}_{1-a+b}\text{B}_a\text{Si}_{1-b}\text{N}_{3-b}\text{O}_b:\text{M}_n$ with $0 \leq x, y, z \leq 1$, $0 \leq a \leq 1$, $0 \leq b < 1$, $0.002 \leq n \leq 0.2$ and M

20 being a metal, according to an embodiment of the present invention selected out of the group comprising Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu or mixtures thereof.

According to an embodiment of the present invention, the CaAlSiN light

25 converting material is selected out of a material comprising $(\text{Ca}_{1-x-y-z}\text{Sr}_x\text{Ba}_y\text{Mg}_z)_{1-n}\text{Al}_{1-a+b}\text{B}_a\text{Si}_{1-b}\text{N}_{3-b}\text{O}_b:\text{RE}_n$ with $0 \leq x, y, z \leq 1$, $0 \leq a \leq 1$, $0 \leq b < 1$, $0.002 \leq n \leq 0.2$ and RE selected out of the group comprising Eu, Ce, or mixtures thereof.

According to an embodiment of the present invention, the CaAlSiN light

30 converting material is selected out of a material comprising $((\text{Ca}_{1-x-y-z}\text{Sr}_x\text{Ba}_y\text{Mg}_z)_{1-$

$0.5*k*nRE^{k+}n)_{1-a}(Al_{1-m}B_m)_bSi_{2-b}N_{3-o}O_o$ with $0 \leq x, y, z \leq 1$, $0 \leq m \leq 1$, $0.002 \leq n \leq 0.2$, $b < o+1$, $a = 0.5(1-o+b)$, $0 \leq o \leq 1$, $k = 2$ or 3 and RE selected out of the group comprising Eu, Ce, or mixtures thereof.

5 According to an embodiment of the present invention, the CaAlSiN light converting material is selected out of a material comprising $((Ca_{1-x-y-z}Sr_xBa_yMg_z)_{1-0.5*k*nRE^{k+}n})_{1-a}(Al_{1-m}B_m)_bSi_{2-b}N_{3-o}O_o$ with $0 \leq x, y, z \leq 1$, $0 \leq m \leq 1$, $0.002 \leq n \leq 0.2$, $b < o+1$, $a = 0.5(1-o+b)$, RE selected out of the group comprising Eu, Ce, or mixtures thereof, with $0 \leq o \leq 0.15$; according to another preferred embodiment $0 \leq o \leq 0.01$,
10 according to a third preferred embodiment $0 \leq o \leq 0.02$.

According to an embodiment of the present invention, the CaAlSiN light converting material is selected out of a material comprising $((Ca_{1-x-y-z}Sr_xBa_yMg_z)_{1-0.5*k*nRE^{k+}n})_{1-a}(Al_{1-m}B_m)_bSi_{2-b}N_{3-o}O_o$ with $0 \leq x, y, z \leq 1$, $0 \leq m \leq 1$, $0.002 \leq n \leq 0.2$, $b < o+1$, $a = 0.5(1-o+b)$, RE selected out of the group comprising Eu, Ce, or mixtures thereof, with $0 \leq o \leq 0.15$; according to another preferred embodiment $0.01 \leq o \leq 0.15$, according to a third preferred embodiment $0.02 \leq o \leq 0.1$.

According to an embodiment of the present invention, the glass phase ratio of
20 the CaAlSiN light converting material is ≥ 2 vol% to ≤ 5 vol%, according to an embodiment of the present invention, ≥ 3 vol% to ≤ 4 vol%. It has been shown in practice that materials with such a glass phase ratio show the improved characteristics, which are advantageous and desired for the present invention. The term “vol%” denotes the volume fraction given in percentage.

25

The term “glass phase” in the sense of the present invention means especially non-crystalline grain boundary phases, which may be detected by scanning electron microscopy or transmission electron microscopy.

30

According to a preferred embodiment of the present invention, the surface roughness RMS (disruption of the planarity of a surface; measured as the geometric

mean of the difference between highest and deepest surface features) of the surface(s) of the CaAlSiN light converting material is $\geq 0.001 \mu\text{m}$ and $\leq 100 \mu\text{m}$. According to an embodiment of the present invention, the surface roughness of the surface(s) of the CaAlSiN light converting material is $\geq 0.01 \mu\text{m}$ and $\leq 10 \mu\text{m}$, according to an 5 embodiment of the present invention $\geq 0.1 \mu\text{m}$ and $\leq 5 \mu\text{m}$, according to an embodiment of the present invention $\geq 0.15 \mu\text{m}$ and $\leq 3 \mu\text{m}$. and according to an embodiment of the present invention $\geq 0.2 \mu\text{m}$ and $\leq 2 \mu\text{m}$.

According to a preferred embodiment of the present invention, the specific 10 surface area of the CaAlSiN light converting material structure is $\geq 10^{-7} \text{ m}^2/\text{g}$ and $\leq 1 \text{ m}^2/\text{g}$.

The present invention furthermore relates to a method of producing a CaAlSiN light converting material for a light-emitting device according to the present invention 15 comprising a sintering step.

The term “sintering step” in the sense of the present invention means especially densification of a precursor powder under the influence of heat, which may be combined with the application of uniaxial or isostatic pressure, without reaching the 20 liquid state of the main constituents of the sintered material.

According to an embodiment of the present invention, the sintering step is performed at ambient pressure, preferably in reducing or inert atmosphere.

According to an embodiment of the present invention, method furthermore 25 comprises the step of pressing the CaAlSiN precursor material to $\geq 50\%$ to $\leq 70\%$, according to an embodiment of the present invention, $\geq 55\%$ to $\leq 60\%$ of its theoretical single crystal density before sintering. It has been shown in practice that this improves the sintering steps for most CaAlSiN light converting materials as described with the 30 present invention.

According to an embodiment of the present invention, the method of producing CaAlSiN light converting material for a light-emitting device according to the present invention comprises the following steps:

5

- (a) Mixing the precursor materials for the CaAlSiN light converting material
- (b) optional firing of the precursor materials, preferably at a temperature of ≥ 1300 °C to ≤ 1800 °C to remove volatile materials (such as CO₂ in case carbonates are used)
- 10 (c) optional grinding and washing
- (d) a first pressing step, preferably a uniaxial pressing step at ≥ 10 kN using a suitable powder compacting tool with a mould in the desired shape (e.g. rod- or pellet-shape) and/ or a cold isostatic pressing step preferably at ≥ 3000 bar to ≤ 3500 bar
- 15 (e) a sintering step at ≥ 1500 °C to ≤ 2200 °C at ambient pressure
- (f) a hot pressing step, preferably a hot isostatic pressing step preferably at ≥ 100 bar to ≤ 2500 bar and preferably at a temperature of ≥ 1500 °C to ≤ 2000 °C and/or a hot uniaxial pressing step preferably at ≥ 100 bar to ≤ 2500 bar and preferably at a temperature of ≥ 1500 °C to ≤ 2000 °C
- 20 (g) optionally a post annealing step at >1000 °C to <1700 °C in inert atmosphere or air

According to an embodiment of the present invention, the light-emitting device comprises a light emitting diode (LED), according to an embodiment of the present invention a LED based on AlInGaN material as the electroluminescent material.

According to an embodiment of the present invention, the light-emitting device comprises a first light converting material, according to an embodiment of the present invention a ceramic first light converting material, and a second light converting material.

According to an embodiment of the present invention, this second light converting comprises a YAG:Ce material. These materials have proven themselves in practice to be suitable for many applications within the present invention.

5

According to an embodiment of the present invention, the light emitting device has a color temperature of $\geq 3000\text{K}$ to $\leq 5000\text{K}$, according to an embodiment of the present invention 3500K to $\leq 4000\text{K}$.

10

The present invention also includes a method of preparing a light-emitting device according to the present invention comprising the steps of:

15

- (a) Providing at least a LED comprising a CaAlSiN first light converting material and at least a second luminescent color converter as described above
- (b) measuring the color temperature of the light emitting device
- (c) adjusting the color temperature of the light emitting device

whereby the steps (b) and (c) may be repeated ad libitum.

20

This method allows for most applications to define the color temperature of the light emitting device as desired and for most applications also to adjust the color temperature of the light-emitting device even after production.

25

According to an embodiment of the present invention, step c) is performed by

abrasion of either the CaAlSiN material and/or the second luminescence color converter material.

30

According to an embodiment of the present invention, step c) is performed by changing the ratio of the amount of CaAlSiN light converting material and the second light converting material.

According to an embodiment of the present invention, this is performed by

using plates or stripes of the CaAlSiN light converting material and the second luminescent color converter material, which are arranged laterally on the light emitting area of the LED chip. The color adjustment is performed by changing the ratio of plates or stripes of the CaAlSiN light converting material towards the plates or stripes of the 5 second light converting material.

According to an embodiment of the present invention, color adjustment is performed by using several prefabricated LED chips which comprise certain different ratios of the CaAlSiN light converting material and second luminescent color converter 10 material or comprise only one of the two materials, so that the adjustment may be performed by changing the brightness of the different LED chips in the light emitting device, or the amount of LED chips of a certain type. The first option will result in a color tunable device.

15 Additionally the color temperature may be set by changing the doping level in either the CaAlSiN light converting material and the second luminescent color converter material or changing the host lattice of either the CaAlSiN light converting material and the second light converting material (e.g. by adding Ga, Gd or Lu in case of a YAG material and/or adding Sr, Ba, Mg or O in case of the CaAlSiN light 20 converting material). These measures may be used alone or in combination with the methods stated above.

25 A light emitting device according to the present invention as well as a CaAlSiN light converting material as produced with the present method may be of use in a broad variety of systems and/or applications, amongst them one or more of the following:

- office lighting systems
- household application systems
- shop lighting systems,
- home lighting systems,
- accent lighting systems,

- spot lighting systems,
- theater lighting systems,
- fiber-optics application systems,
- projection systems,
- 5 - self-lit display systems,
- pixelated display systems,
- segmented display systems,
- warning sign systems,
- medical lighting application systems,
- 10 - indicator sign systems, and
- decorative lighting systems
- portable systems
- automotive applications
- green house lighting systems

15

The aforementioned components, as well as the claimed components and the components to be used in accordance with the invention in the described embodiments, are not subject to any special exceptions with respect to their size, shape, material selection and technical concept such that the selection criteria known in the pertinent 20 field can be applied without limitations.

Additional details, characteristics and advantages of the object of the invention are disclosed in the sub-claims, the figure and the following description of the respective figures and examples, which --in an exemplary fashion-- show an 25 embodiment of a CaAlSiN light converting material for use in a light emitting device according to the invention.

30 Fig. 1 shows an emission spectra of an LED comprising a CaAlSiN light converting material according to Example I of the present invention

EXAMPLE I:

Fig. 1 refers to $(\text{Ca}_{0.95}\text{Sr}_{0.05})_{0.98}\text{AlSiN}_3:\text{Eu}_{0.02}$ (Example I) which was produced as follows:

5

$(\text{Ca}_{0.95}\text{Sr}_{0.05})_{0.98}\text{AlSiN}_3:\text{Eu}_{0.02}$ was synthesized from 6.894 g Ca_3N_2 (Alfa Aesar, Karlsruhe, Germany), 710 mg SrN_2 (Cerac, Milwaukee, WI, USA), 6.148 g AlN (Nanoamor, Los Alamos, NM, USA), amorphous 7.364 g SiN (Alfa Aesar) and 627 mg EuF_3 (Aldrich, Taufkirchen, Germany). The powders were mixed in a porcelain mortar, 10 filled into Molybdenum crucibles and fired for 4 h at 1450°C in forming gas atmosphere. The resulting red powder was washed with acetic acid, water and 2-propanol to remove by-products and finer particles.

15 The obtained powder was compressed into pellets, cold isostatically pressed at 3200 bar and sintered at 1700°C in forming gas atmosphere for 4h. The resulting pellets displayed a closed porosity and were subsequently hot isostatically pressed at 2000 bar and 1700°C to obtain dense ceramics with >98% of the theoretical density.

20 The thus produced $(\text{Ca}_{0.95}\text{Sr}_{0.05})_{0.98}\text{AlSiN}_3:\text{Eu}_{0.02}$ was incorporated into a light emitting device in form of a luminescence converting layer (638 nm peak wavelength, 200 μm $(\text{Ca}_{0.93}\text{Sr}_{0.05}\text{Eu}_{0.02})\text{AlSiN}_3$ plate) using AlInGaN as an electroluminescent material having a wavelength of 450 nm.

25 Fig. 1 shows an emission spectra of this LED. The Color point is: x = 0.615, y = 0.384

30 The particular combinations of elements and features in the above detailed embodiments are exemplary only; the interchanging and substitution of these teachings with other teachings in this and the patents/applications incorporated by reference are also expressly contemplated. As those skilled in the art will recognize, variations, modifications, and other implementations of what is described herein can occur to those

of ordinary skill in the art without departing from the spirit and the scope of the invention as claimed. Accordingly, the foregoing description is by way of example only and is not intended as limiting. The invention's scope is defined in the following claims and the equivalents thereto. Furthermore, reference signs used in the description and

5 claims do not limit the scope of the invention as claimed.

CLAIMS

1. Light emitting device with a light source to emit primary light and a light conversion layer to convert at least a part of the primary light into secondary light comprising a CaAlSiN light converting material with a transmissivity of $\geq 10\%$ to $\leq 80\%$ for a light in the wavelength range from ≥ 580 to ≤ 1000 nm.

5

2. The light emitting device of claim 1, whereby the CaAlSiN light converting material emits secondary light in the red visible light wavelength range with a maximum intensity at a wavelength of ≥ 590 nm to ≤ 700 nm.

10 3. The light emitting device of claim 1 or 2, whereby the CaAlSiN light converting material emits secondary light with an wavelength distribution with a half-width of ≥ 50 nm to ≤ 150 nm.

15 4. The light emitting device of any of the claims 1 to 3 whereby the CaAlSiN light converting material has a density of $\geq 97\%$ to $\leq 100\%$ of the theoretical single crystal density.

20 5. The light emitting device of any of the claims 1 to 4 whereby the shift of the maximum and/or the half-width in the secondary light emission in the red visible light wavelength range of the CaAlSiN light converting material is ≤ 20 nm over the whole temperature range from $\geq 50^{\circ}\text{C}$ to $\leq 150^{\circ}\text{C}$.

6. The light emitting device of any of the claims 1 to 5 whereby the CaAlSiN light converting material is selected out of a material comprising $(Ca_{1-x-y-z}Sr_xBa_yMg_z)_{1-n}Al_{1-a+b}B_aSi_{1-b}N_{3-b}O_b:M_n$ with $0 \leq x,y,z \leq 1$, $0 \leq a \leq 1$, $0 \leq b < 1$, $0.002 \leq n \leq 0.2$ and M being a metal, according to an embodiment of the present invention selected out of the group comprising Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu or mixtures thereof.

7. The light emitting device of any of the claims 1 to 6 whereby the glass phase ratio of the CaAlSiN light converting material is ≥ 2 vol% to ≤ 5 vol%.

10

8. A method of producing a CaAlSiN light converting material for a light emitting device according to any of the claims 1 to 7 comprising a sintering step.

15

9. A method of producing a CaAlSiN light converting material as claimed in claim 8, further comprising a step of pressing the CaAlSiN precursor material to $\geq 50\%$ to $\leq 70\%$ of its theoretical density before sintering.

20

10. A system comprising a light emitting device according to any of the claims 1 to 7 and/or a CaAlSiN light converting material as produced according to the method of any of the claims 8 to 9, the system being used in one or more of the following applications:

25

- Office lighting systems
- household application systems
- shop lighting systems,
- home lighting systems,
- accent lighting systems,
- spot lighting systems,
- theater lighting systems,

- fiber-optics application systems,
- projection systems,
- self-lit display systems,
- pixelated display systems,
- 5 - segmented display systems,
- warning sign systems,
- medical lighting application systems,
- indicator sign systems, and
- decorative lighting systems
- 10 - portable systems
- automotive applications
- green house lighting systems

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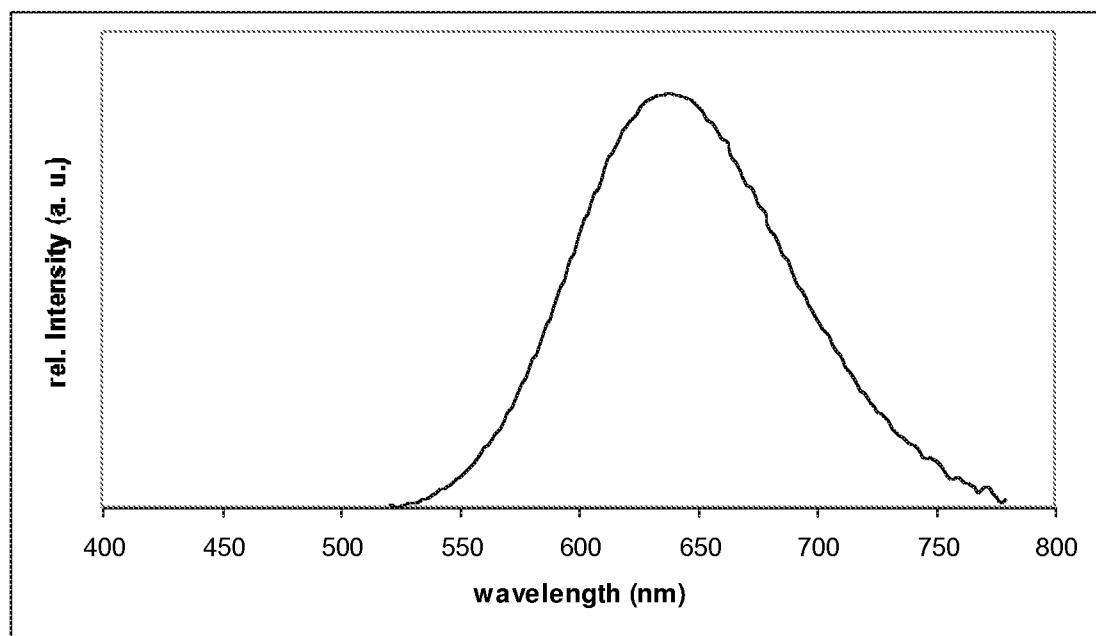


Fig. 1

INTERNATIONAL SEARCH REPORT

International application No

PCT/IB2006/053969

A. CLASSIFICATION OF SUBJECT MATTER
INV. C09K11/77 H01L33/00

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)
C09K H01L

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 practical, search terms used)

EPO-Internal, WPI Data, CHEM ABS Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

| Category* | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. |
|-----------|---|-----------------------|
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Further documents are listed in the continuation of Box C.

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