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(54) Title: COMPOSITE-PHASE MECHANOLUMINESCENT MATERIALS WITH ADJUSTABLE COLOR AND INTENSITY, AND PREPARATION METHOD THEREOF

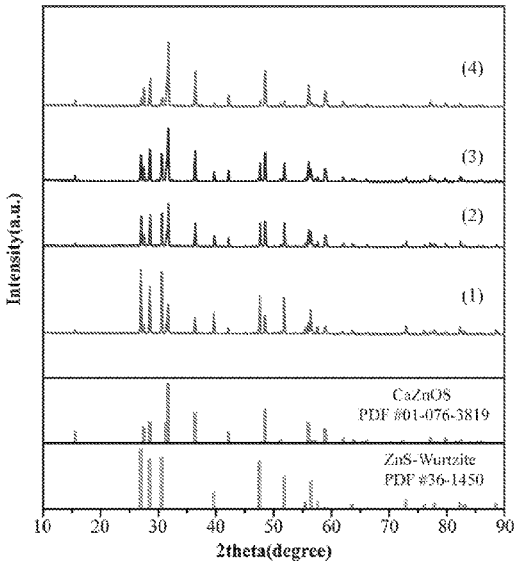

Fig. 1: X-ray diffraction pattern of the composite-phase mechanoluminescent material

(57) Abstract: A composite-phase mechanoluminescent material with adjustable color and intensity, and a preparation method thereof. The material is prepared by using a ZnS-AOZnS composite phase as a matrix, performing ion exchange at an A-site and a Zn-site and co-doping with transition metal ions and rare earth ions. The chemical formula of the mechanoluminescent material is ZnS-AOZnS: xN, yM ($0 \leq x \leq 1$, $0 \leq y \leq 1$), where N is a substitution element at the Zn-site and is one or more of Cu, Mn and Cr, M is a substitution element at the A-site and is one or more of Pr, Tb, Sm, Ho, Nd, Bi, Pb, Ag, Ti, Gd, Eu and Yb, A is one or more of Ca, Sr and Ba, and x and y represent molar percentages. The composite phase indicates that the matrix material prepared by compounding two phases by a high-temperature solid-state reaction method.

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COMPOSITE-PHASE MECHANOLUMINESCENT MATERIALS WITH ADJUSTABLE COLOR AND INTENSITY, AND PREPARATION METHOD THEREOF

TECHNICAL FIELD

The present invention belongs to the technical field of mechanoluminescent functional materials, and relates to a composite-phase mechanoluminescent material with adjustable color and intensity, and a preparation method thereof.

BACKGROUND OF THE PRESENT INVENTION

Mechanoluminescence (ML) is dynamic light emission resulting from any mechanical action (such as twisting, pressing, stretching, breaking, etc.) on a solid material. Unlike the excitation modes of common photoluminescence and electroluminescence, the mechanoluminescence is direct light emission resulting from a mechanical stress, requiring no energization or illumination. Moreover, within a certain elasticity range, the intensity is linearly related to the mechanical stress. The prepared devices have characteristics of wireless transmission, non-destruction, repeatability, real-time visualization and the like. As one kind of novel inorganic illuminants, intelligent mechanoluminescent materials become a hot research field at present, and have great application prospect in fields such as artificial intelligence, paperless office, intelligent anti-counterfeiting, stress sensing, touch screen technology and electronic skin.

Under a weak external force, intelligent mechanoluminescent materials exhibit MLcharacteristics visible to the naked eyes. Within a certain stress range, the light emission of an object resulting from an external force is called elastic ML, and this ML is non-destructive and self-restorative. Due to the characteristic that the non-destructive luminescence and the ML intensity of the elastic mechanoluminescent materials are directly proportional to the applied mechanical stress, the elastic mechanoluminescent materials can be applied in the preparation of intelligent stress distribution sensors to realize real-time stress distribution visualization for an object

to be detected. The elastic ML materials also have considerable advantages in artificial intelligence, bridge stress detection, earthquake prediction, electronic skin and intelligent anti-counterfeiting systems. So far, there have been dozens of elastic ML material systems. However, the already reported ML materials show low non-destructive ML intensity, low luminous efficiency (below 1% in most cases), and low repeatability, and thus cannot meet the production requirements. Only few intensiveML materials have been developed at present, including $SrAl_2O_4:Eu^{2+}/Dy^{3+}$, ZnS:Mn/Cu/Ag, and CaZnOS:Mn/Ln(Ln=Pr, Ho, Er, Nd, Tb, Dy, Eu, Sm, Yb, and Tm), etc, while above mentioned materials are single phase . In order to obtainML materials with high brightness and multiple colors, the present invention provides a diphasichero-structure ML material with simple operation, low cost, and adjustable color and intensity.

SUMMARY OF THE PRESENT INVENTION

In order to solve the limited varieties of mechanoluminescent materials with high brightness and adjustable color, an objective of the present invention is to provide a composite-phase mechanoluminescent material with adjustable color and intensity. The material is high in elastic mechanoluminescenceintensity, simple in preparation process, low in cost and stable in chemical properties, and can directly respond to mechanical stress signals in various forms (e.g., compression, stretching, bending, collision, rubbing, twisting, etc.). Light emitted by the material can be observed with the naked eyes in a dark environment.

The chemical formula of the composite-phase mechanoluminescent material with adjustable color and intensity in the present invention is ZnS-AOZnS:xN,yM ($0\leq x\leq 1$, $0\leq y\leq 1$), where N is a substitution element at the Zn-site and is one or more of Cu, Mn and Cr, M is a substitution element at the A-site and is one or more of Pr, Tb, Sm, Ho, Nd, Bi, Pb, Ag, Ti, Gd, Eu and Yb, A is one or more of Ca, Sr and Ba, and x and y represent molar percentages.

A method for preparing the composite-phase mechanoluminescent material with adjustable color and intensity in the present invention includes the following steps:

(1) preparing raw materials: using oxides, hydroxides or carbonates of Ca, Ba, Sr, Cu, Mg, Mn and Cr, sulfides of Zn and S, and oxides, carbonates or soluble nitrates, sulfates or chlorides of Pr and co-doped rare earth elements and transition metal elements as raw materials; adding cosolvents LiF, NaF, KF, LiCl, NaCl, KCl, LiBr, NaBr and KBr; weighing the raw materials according to stoichiometric ratios, then adding a proper amount of absolute ethyl alcohol or deionized water in the mixed raw materials, grinding and uniformly mixing in an agate mortar, and drying in an oven at 40℃ to 300℃ to obtain mixed powder;

(2) putting the uniformly grinded powder into an alumina crucible, heating to 800℃ to 1400℃at a rate of 1℃/min to 100℃/min in an inert atmosphere, calcining for 0.5h to 24h, and naturally cooling; and

(3) finely grinding the cooled powder obtained in the step (2), and sieving with a 300-mesh sieve to obtain composite-phase mechanoluminescent material ZnS-AOZnS:xN,yM powder.

In the present invention, the prepared mechanoluminescent powder, which is already grinded and sieved, is formed as a composite together with an optical transparent organic high-molecular elastic material, preferably PET, TPU or PDMS; the composite is machined into sheets or coated on the surface of a component to be measured; and, the mechanical stress applied onto the composite or component can be transformed into optical signals which are then transmitted, so that the stress-light direct energy conversion with adjustable color and intensity is realized. Moreover, within a certain elasticity range, the intensity is directly proportional to the applied stress, and the change in light brightness can be observed with the naked eyes.

The composite-phase mechanoluminescent material of the present invention is mixed with PDMS and then machined into sheets. Under a certain stress, the mechanoluminescence color of samples changes and the mechanoluminescence peak is red-shifted.

With the use of the composite-phase mechanoluminescent material of the present invention, the intensity of the samples is enhanced, and the intensity can be observed in a bright environment.

In the present invention, the composite-phase mechanoluminescent material is synthesized in a single step, the preparation process is simple, the cost is low, and the presence of impurity phases during the preparation is reduced. Accordingly, the composite-phase mechanoluminescent material with excellent light emission characteristics is synthesized.

BRIEF DESCRIPTION OF THE DRAWINGS

Fig. 1 is an X-ray diffraction pattern of a composite-phase mechanoluminescent material according to Embodiment 1 of the present invention.

Fig. 2 is a photoluminescence (PL) spectrum of the composite-phase mechanoluminescent material product according to Embodiment 1 of the present invention.

Fig. 3 is a mechanoluminescence (ML) spectrum of the composite-phase mechanoluminescent material product according to Embodiment 1 of the present invention.

Fig. 4 is a mechanoluminescence (ML) spectrum of a composite-phase mechanoluminescent material according to Embodiment 2 of the present invention.

Fig. 5 is a mechanoluminescence (ML) spectrumintegratedintensity over 500~750 nm of the composite-phase mechanoluminescent material product under mechanical excitation at 35N as a function of CaZnOS/ZnS molar ratio in the precursoraccording to Embodiment 1 of the present invention.

DETAILED DESCRIPTION OF THE PRESENT INVENTION

The present invention will be further described below by specific embodiments. However, it should be understood that these embodiments are merely used for describing the present invention rather than limiting the scope of the present invention.

Specifically, the preparation methods in the embodiments will be described below.

Embodiment 1

By taking ZnS-CaZnOS:Mn as an example, the specific ratio of samples and the mass of raw materials were shown in Table 1.

Sample No.	Ratio	Mass of raw materials (g)	Phase structure
(1)	ZnS/CaZnOS=4/1	ZnS=9.74 $CaCO_3$=2 $MnCO_3$=0.12	Composite phase: ZnS-CaZnOS
(2)	ZnS/CaZnOS=3/2	ZnS=9.74 $CaCO_3$=4 $MnCO_3$=0.12	Composite phase: ZnS-CaZnOS
(3)	ZnS/CaZnOS=1/1	ZnS=9.74 $CaCO_3$=5 $MnCO_3$=0.12	Composite phase: ZnS-CaZnOS
(4)	ZnS/CaZnOS=1/4	ZnS=9.74 $CaCO_3$=8 $MnCO_3$=0.12	Composite phase: ZnS-CaZnOS

The mixed raw materials at the above ratio were put into an agate mortar, then added with absolute ethyl alcohol or deionized water, and grinded and uniformly mixed in the agate mortar. The mixture was dried in an oven at 100℃ to obtain mixed powder. The uniformly grinded powder was put into an alumina crucible, then heated to 800℃ to 1400℃at a rate of 1℃/min to 100℃/min in an inert atmosphere, calcined for 0.5h to 24h, and naturally cooled. The cooled powder was finely grinded, and then sieved with a 300-mesh sieve to obtain composite-phase mechanoluminescent material ZnS-CaZnOS:Mn powder. The obtained mechanoluminescent powder was formed as a composite with an optical transparent organic high-molecular elastic material (PET, TPU or PDMS), and a device was prepared from the composite to test the mechanoluminescence characteristic.

Embodiment 2

By taking ZnS-CaZnOS:0.1%Mn,2%Pr as an example, specific operation steps were as follows.

a) Raw materials are prepared: $CaCO_3$, ZnS, $MnCO_3$, $Pr(NO_3)_3$-$6H_2O$ and LiF are used as raw materials, wherein oxides or carbonates of $CaCO_3$, sulfides of Zn and S, and oxides, carbonates or soluble nitrates, sulfates or chlorides of Mn and Pr were used as raw materials. A cosolvent LiF was added. The raw materials were weighed according to stoichiometric ratios. A proper amount of absolute ethyl alcohol or deionized water was then added in the mixed raw materials, and the mixture was grinded and uniformly mixed in an agate mortar and then dried in an oven at 100℃ to obtain mixed powder.

b) The uniformly grinded powder was put into an alumina crucible, then heated to 1000℃ to 1200℃at a rate of 1℃/min to 100℃/min in an inert atmosphere, calcined for 6h to 8h, and naturally cooled.

c) The cooled powder obtained in the step b) was grinded for 10 min to 30 min, and then sieved with a 300-mesh sieve to obtain composite-phase mechanoluminescent material ZnS-CaZnOS:Mn powder. The obtained mechanoluminescent powder was formed as a composite with an optical transparent organic high-molecular elastic material (PET, TPU or PDMS), and a device was prepared from the composite to test the mechanoluminescence characteristic.

Embodiment 3

By taking ZnS-BaZnOS:Mn as an example, the specific ratio of samples and the mass of raw materials were similar to those in Table 1, with the element Ca substituted by the element Ba, and the other steps were similar to those in Embodiment 1.

Embodiment 4

By taking ZnS-BaZnOS:0.1%Mn,2%Pr as an example, specific operation steps were as follows.

d) Raw materials are prepared: $BaCO_3$, ZnS, $MnCO_3$, $Pr(NO_3)_3$-$6H_2O$ and LiF were used as raw materials, wherein oxides or carbonates of $BaCO_3$, sulfides of Zn and S, and oxides, carbonates or soluble nitrates, sulfates or chlorides of Mn and Pr

were used as raw materials. A cosolvent LiF was added. The raw materials were weighed according to stoichiometric ratios. A proper amount of absolute ethyl alcohol or deionized water was then added in the mixed raw materials, and the mixture was grinded and uniformly mixed in an agate mortar and then dried in an oven at 100℃ to obtain mixed powder.

e) The uniformly grinded powder was put into an alumina crucible, then heated to 1000℃ to 1200℃at a rate of 1℃/min to 100℃/min in an inert atmosphere, calcined for 6h to 8h, and naturally cooled.

f) The cooled powder obtained in the step b) was grinded for 10 min to 30 min, and then sieved with a 300-mesh sieve to obtain composite-phase mechanoluminescent material ZnS-BaZnOS:Mn powder. The obtained mechanoluminescent powder was formed as a composite with an optical transparent organic high-molecular elastic material (PET, TPU or PDMS), and a device was prepared from the composite to test the mechanoluminescence characteristic.

Embodiment 5

By taking ZnS-SrZnOS:Mn as an example, the specific ratio of samples and the mass of raw materials were similar to those in Table 1, with the element Ca substituted with the element Sr, and the other steps were similar to those in Embodiment 1.

Embodiment 6

By taking ZnS-SrZnOS:0.1%Mn,2%Pr as an example, specific operation steps were as follows.

g) Raw materials are prepared: $SrCO_3$, ZnS, $MnCO_3$, $Pr(NO_3)_3$-$6H_2O$ and LiF were used as raw materials, wherein oxides or carbonates of $SrCO_3$, sulfides of Zn and S, and oxides, carbonates or soluble nitrates, sulfates or chlorides of Mn and Pr were used as raw materials. A cosolvent LiF was added. The raw materials were weighed according to stoichiometric ratios. A proper amount of absolute ethyl alcohol or deionized water was then added in the mixed raw materials, and the mixture was grinded and uniformly mixed in an agate mortar and then dried in an oven at 100℃ to obtain mixed powder.

h) The uniformly grinded powder was put into an alumina crucible, then heated to 1000℃ to 1200℃at a rate of 1℃/min to 100℃/min in an inert atmosphere, calcined for 6h to 8h, and naturally cooled.

i) The cooled powder obtained in the step b) was grinded for 10 min to 30 min, and then sieved with a 300-mesh sieve to obtain composite-phase mechanoluminescent material ZnS-SrZnOS:Mn powder. The obtained mechanoluminescent powder was formed as a composite with an optical transparent organic high-molecular elastic material (PET, TPU or PDMS), and a device was prepared from the composite to test the mechanoluminescence characteristic.

CLAIMS

1. A composite-phase mechanoluminescent material with adjustable color and intensity, which is prepared by using a ZnS-AOZnS composite phase as a matrix, performing ion exchange at an A-site and a Zn-site and co-doping with transition metal ions and rare earth ions; and the chemical formula of the mechanoluminescent material is ZnS-AOZnS:xN,yM ($0 \leq x \leq 1$, $0 \leq y \leq 1$), where N is a substitution element at the Zn-site and is one or more of Cu, Mn and Cr, M is a substitution element at the A-site and is one or more of Pr, Tb, Sm, Ho, Nd, Bi, Pb, Ag, Ti, Gd, Eu and Yb, A is one or more of Ca, Sr and Ba, and x and y represent molar percentages.

2. A composite-phase mechanoluminescent material with adjustable color and intensity, wherein the material matrix contains two phases.

3. A composite-phase mechanoluminescent material with adjustable color and intensity, wherein an elastic mechanoluminescence peak of the material is shifted from 594 nm to 609 nm, red-shifted by 15 nm; the material is high in elasticmechanoluminescence intensity; the elasticmechanoluminescence intensity observed with naked eyes in a dark environment is more than two times of the elasticmechanoluminescence intensity of ZnS:Mn; and, within a certain elasticity range, the elasticmechanoluminescence intensity is linearly related to the mechanical stress, and increases with the increase of stress.

4. A method for preparing the composite-phase mechanoluminescent material with adjustable color and intensity according to claims 1, 2 and 3, wherein a high-temperature solid-state reaction method is used, and the preparation process comprises the following steps:

a) preparing raw materials: using oxides, hydroxides or carbonates of Ca, Ba, Sr, Cu, Mg, Mn and Cr, sulfides of Zn and S, and oxides, carbonates or soluble nitrates, sulfates or chlorides of Pr and co-doped rare earth elements and transition metal elements as raw materials; adding cosolvents LiF, NaF, KF, LiCl, NaCl, KCl, LiBr,

NaBr and KBr; weighing the raw materials according to stoichiometric ratios, then adding a proper amount of absolute ethyl alcohol or deionized water in the mixed raw materials, grinding and uniformly mixing in an agate mortar, and drying in an oven at 40℃ to 300℃ to obtain mixed powder;

b) putting the uniformly grinded powder into an alumina crucible, heating to 800℃ to 1400℃at a rate of 1℃/min to 100℃/min in an inert atmosphere, calcining for 0.5h to 24h, and naturally cooling; and

c) finely grinding the cooled powder obtained in the step b), and sieving with a 300-mesh sieve to obtain composite-phase mechanoluminescent material ZnS-AOZnS:xN,yM powder.

5. The composite-phase mechanoluminescent material with adjustable color and intensity according to claims 1 to 4, wherein the prepared mechanoluminescent powder, which is already grinded and sieved, is formed as a composite together with an optical transparent organic high-molecular elastic material, preferably PET, TPU or PDMS; the composite is machined into sheets or coated on the surface of a component to be measured; and, the mechanical stress applied onto the composite or component can be transformed into optical signals which are then displayed, so that the stress-light direct energy conversion with adjustable color and intensity is realized.

6. The composite-phase mechanoluminescent material with adjustable color and intensity according to claims 1 to 5, which can be widely applied in various fields, such as real-time detection of stress distribution in buildings, mechanical components and the like, stress sensors, intelligent anti-counterfeiting, dimming for multicolor illumination in displays, earthquakeprediction, and novel artificial intelligence devices.

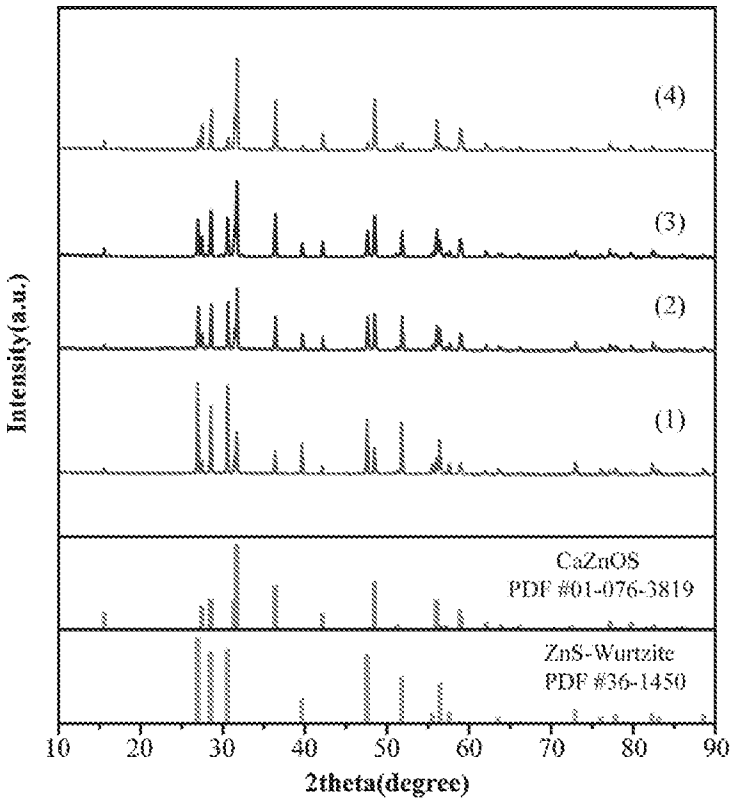

Fig. 1: X-ray diffraction pattern of the composite-phase mechanoluminescent material

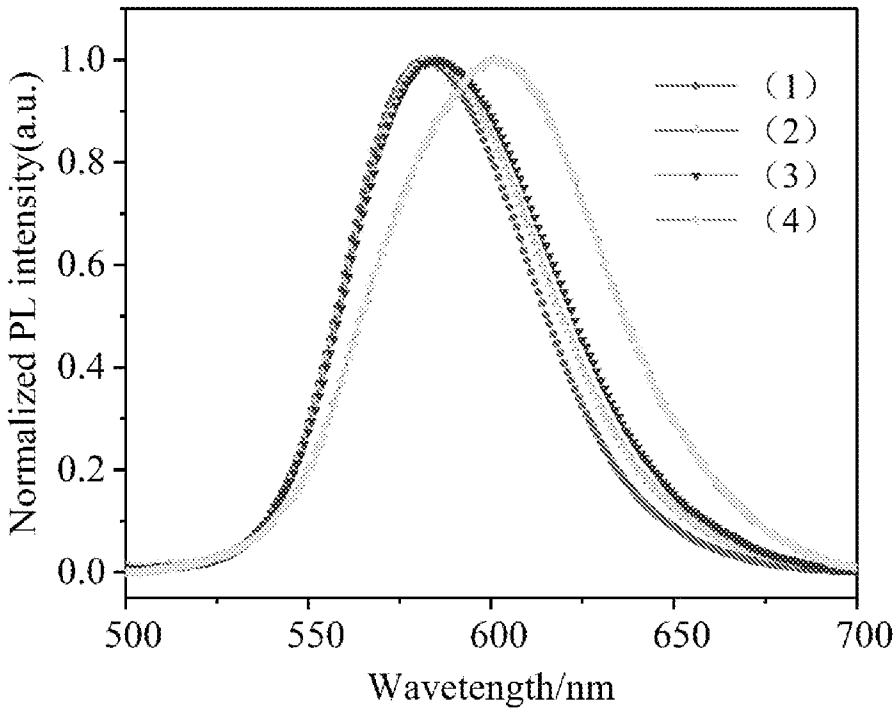

Fig. 2: Photoluminescence spectrum of the composite-phase mechanoluminescent material

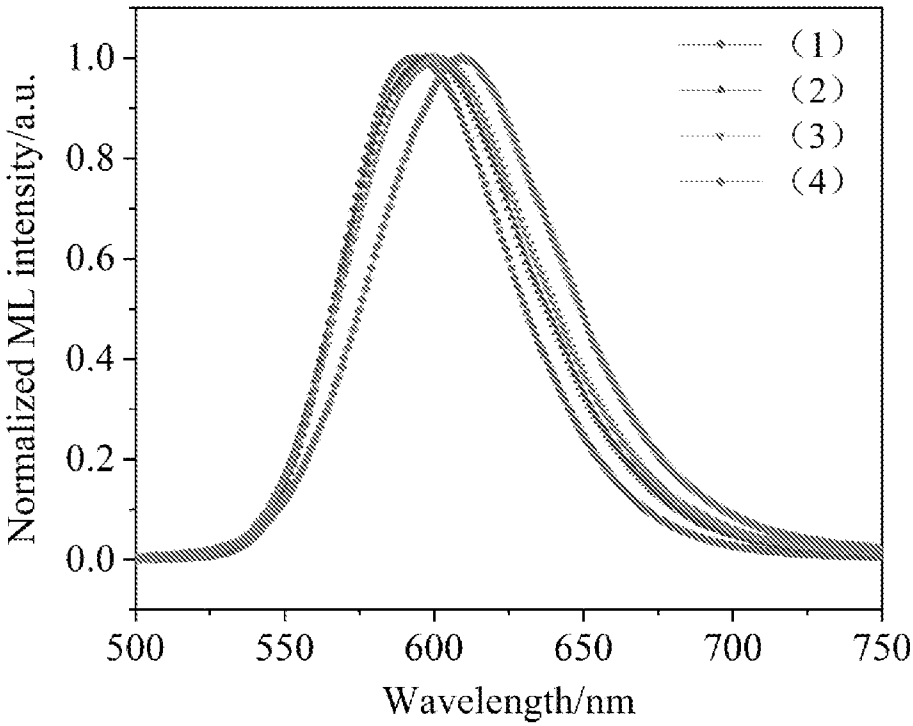

Fig. 3: Mechanoluminescence spectrum of the composite-phase mechanoluminescent material

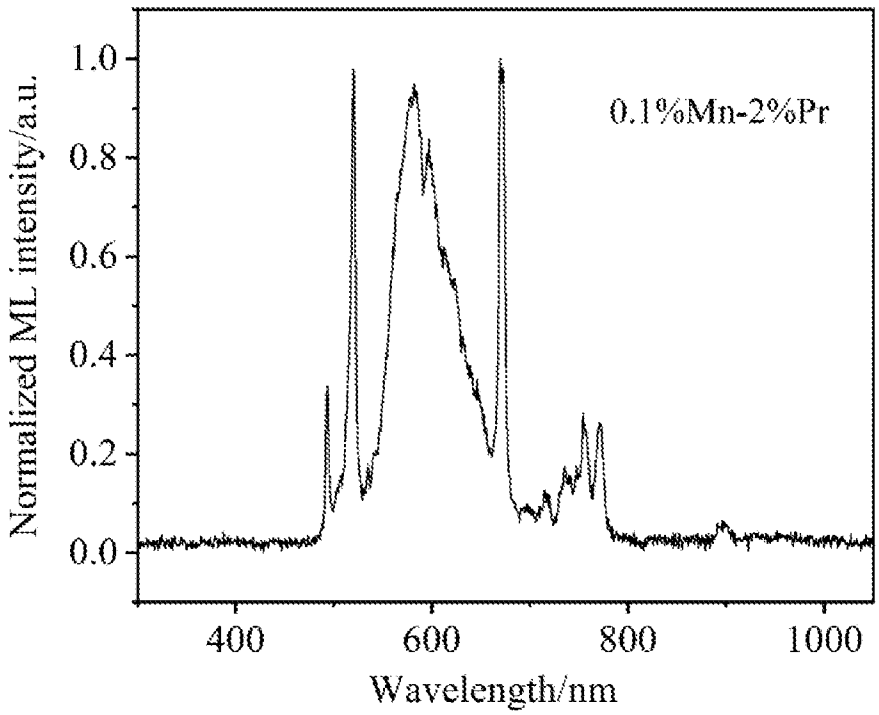

Fig. 4: Mechanoluminescence spectrum of the composite-phase mechanoluminescent materialZnS-CaZnOS:0.1%Mn,2%Pr

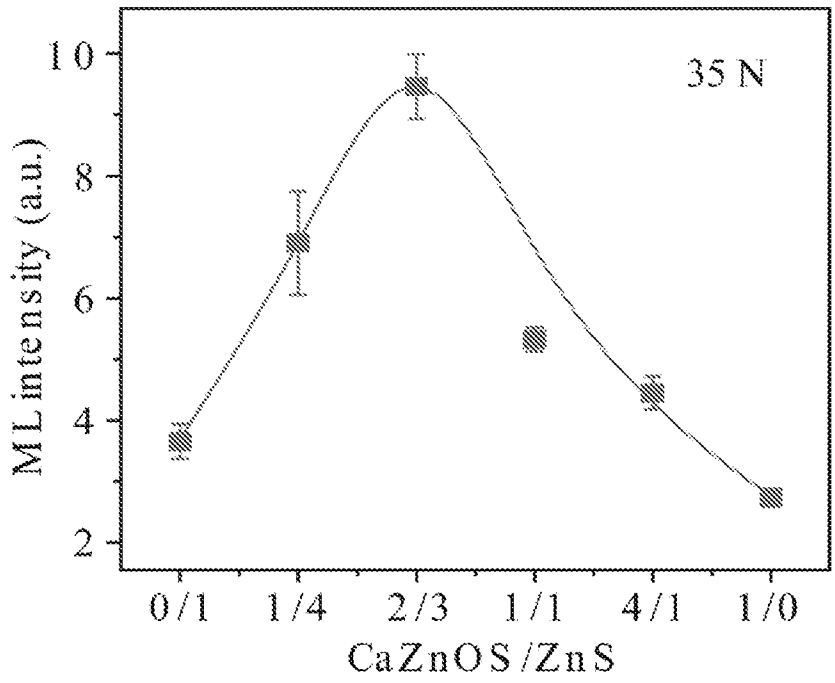

Fig. 5:Integrated intensity over 500~750 nm of the composite-phase mechanoluminescent material under mechanical excitation at 35N as a function of CaZnOS/ZnS molar ratio in the precursor

INTERNATIONAL SEARCH REPORT

International application No.
PCT/CN2019/114684

A. CLASSIFICATION OF SUBJECT MATTER

C09K 11/57(2006.01)i

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)

C09K11/-

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

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

CNPAT,CNKI,EPODOC,WPI,Google scholar,STN,+luminesc+, ZnS,+OZnS,+ZnOS, Ca ,Sr,Ba, calcium, Strontium,barium, Pr, Tb, Sm, Ho, Nd, Bi, Pb, Ag, Ti, Gd, Eu ,Yb,praseodymium,terbium,samarium, holmium, neodymium, bismuth, plumbum, silver, titanium, gadolinium,Europium,ytterbium

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	TSUJIMOTO Yoshihiro et al. "Function of Tetrahedral ZnS3O Building Blocks in the Formation of SrZn2S2O: A Phase Matchable Polar Oxysulfide with a Large Second Harmonic Generation Response" *Chem. Mater.*, Vol. 30, 22 August 2018 (2018-08-22), 6486–6493, EXPERIMENTAL SECTION	1-6
PX	CN 109880617 A (UNIV SHENZHEN) 14 June 2019 (2019-06-14) paragraph[0007]	1-6
A	CN 102925142 A (UNIV HUNAN NORMAL) 13 February 2013 (2013-02-13)	1-6
A	CN 107565944 A (UNIV SHENZHEN) 09 January 2018 (2018-01-09)	1-6
A	CN 104388898 A (UNIV HUBEI) 04 March 2015 (2015-03-04)	1-6
A	CN 105734491 A (UNIV HUBEI) 06 July 2016 (2016-07-06)	1-6

☐ Further documents are listed in the continuation of Box C. ☑ See patent family annex.

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"A" document defining the general state of the art which is not considered to be of particular relevance
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"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
13 January 2020	**01 February 2020**
Name and mailing address of the ISA/CN **National Intellectual Property Administration, PRC** **6, Xitucheng Rd., Jimen Bridge, Haidian District, Beijing 100088** **China** Facsimile No. **(86-10)62019451**	Authorized officer **CHEN,Chunchun** Telephone No. **86-(10)-53962329**

INTERNATIONAL SEARCH REPORT
Information on patent family members

International application No.
PCT/CN2019/114684

Patent document cited in search report	Publication date (day/month/year)	Patent family member(s)	Publication date (day/month/year)
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CN 104388898 A	04 March 2015	CN 104388898 B	01 February 2017
CN 105734491 A	06 July 2016	None	