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(54) Title: ZNO-AL₂O₃-MGO SPUTTERING TARGET AND METHOD FOR THE PRODUCTION THEREOF

(57) Abstract: It is known to produce a crystalline material consisting of ZnO-Al₂O₃-MgO (ZAMO) by dry mixing powders of raw material components comprising Al₂O₃, ZnO and MgO or precursors of these substances and sintering the powder mixtures so as to obtain the sputtering target. It is an object of the present invention to provide a target with high purity having 3N quality or better and which comprises no or small amounts of single phase consisting of Al₂O₃ and MgO. For that purpose it is proposed that said the step of mixing comprises a mixing of powders of the raw material components in a liquid, whereby the maximum solubility for said raw material powders in said liquid is less than 200 mg/kg and whereby said liquid used in the mixing step comprises water and whereby powder of MgCO₃ is used as a precursor component for MgO. The obtained ZAMO sputtering target is characterized by a micro structure containing single component oxide phase consisting of either Al₂O₃ or of MgO in an amount of less than 1 % per weight, whereby the microstructure contains regions of single and binary phases of zincite (ZnAlO₄) and spinel (MgAlO₄).

**ZnO-Al₂O₃-MgO sputtering target
and method for the production thereof**

DESCRIPTION

Technical Field

5 The present invention relates to a ZnO-Al₂O₃-MgO sputtering target. In the following such material is also referred to as ZAMO.

Furthermore, the invention refers to a method for producing a ZnO-Al₂O₃-MgO sputtering target comprising comprising the steps of mixing powders of raw material components comprising Al₂O₃, ZnO and MgO or precursors of these

10 substances and sintering the powder mixture so as to obtain the sputtering target, whereby the step of mixing comprises a mixing of powders of the raw material components in a liquid.

Prior art

EP 1 887 100 B1 describes a ZAMO material and its production. The material is obtained by dry mixing single oxides on a powder metallurgical way. It is proposed to precalcine mixtures of two single oxides at 1000°C and add the third oxide press-shape finally, and subsequently sintering the target.

The major disadvantages of such method are inhomogeneous oxide distributions and certain amounts of single phases of Al₂O₃ and MgO. The dry mixing in a ball mill is applied sometimes even twice and will cause significant amounts of impurity originating of the milling media.

This makes a pure target with 3N quality or better impossible and has severe consequences for layer properties, especially translucency. Electrical resistance is at least 30 mOhm·cm, e.g. 35 mOhm·cm for the example ZAMO 88/1.5/10.5
25 at%.

The alternative method by producing a ceramic ZnO-Al₂O₃-MgO (ZAMO) with small grain sizes via water based slurry and wet milling causes problems due to solubility of Mg²⁺ ions dissolved from the MgO. These Mg²⁺ ions destabilise the electric double layer surrounding each particle. Therefore a good deflocculating of 5 single oxide particles the slurry cannot be achieved.

This problem can be avoided by a method for producing a sputtering target of Al-Mg-Ga-Zn oxide as it is disclosed in EP 2 514 851 A1. The preparation of the sputtering target comprises wet milling of oxide starting powders in ethanol to a mean particle size < 0.4 microns, drying the powder and hot pressing in a graphite 10 mold under vacuum. The resulting sputtering targets have an electrical resistance of 6 mOhm·cm, respectively 2 mOhm·cm. The portion of Ga₂O₃ is below 0.1 wt.-% but this low content is sufficient to obtain a transparent film from the sputtering target, which after heating in a humid atmosphere retains its low electrical resistivity.

15 US 2009/101493 A1 discloses a sputtering target of Mg-Zn oxide which may additionally contain gallium oxide and/or aluminum oxide. The portion of Mg is 2-30 at.-% and that of Ga₂O₃/Al₂O₃ from 3.2 to 6.5 at.-%. The target material is prepared by a sintering process. The starting powder is very finely ground (< 1µm) and processed together with a binder into granules, which are then pressed 20 into shaped bodies and sintered. The resulting target material consists of a complete solid solution of Al/Ga and Mg in ZnO. In a XRD diagram neither an MgO phase nor a mixed phase MgAl₂O₄ can be found. The electrical resistance of the target material is about 1 mOhm.cm.

US 2012/0153237 A1 describes a varistor composition based on ZnO and 25 additional components in the form of Ca- and Mg-containing compounds, which shall be suitable to reduce the dielectric constant of the varistor material. It is mentioned, that the Mg-containing additive is MgO or MgCO₃. Both compounds show high solubility in ZnO crystal matrix so that a solid solution with ZnO can be obtained.

Technical objective

However perfect deflocculating is a prerequisite for well dispersed oxide mixtures as well in milling processes which is demanded in the case of ZAMO. This is to guarantee homogenous oxide distribution.

5 Therefore, smooth sputter behaviour by avoiding single inclusions of single nonconductive oxides is aspired, especially if even more strict demands for producing homogenous optical layers from such sputtering target shall be met. The enrichments of single oxides in a sputter target are disadvantageous since arcing, preferential sputtering and inhomogeneous sputter layer can be the
10 consequence.

Using fine milled powders as a starting material requires intensive milling procedures involving time and money and resulting inevitably in abrasion which may bring impurities into the powder.

Therefore, it is an object of the present invention to provide a target with high
15 purity having 3N quality or better and which is suitable to produce homogenous optical layers fulfilling high quality demands.

Furthermore, it is an object of the invention to provide a method which is suitable for the inexpensive manufacturing of sputtering targets.

General description of the invention

20 By starting from the aforementioned method this object is achieved according to the invention with respect to the method, ,in that the maximum solubility for said raw material powders in said liquid is less than 200 mg/kg and whereby said liquid comprises water, and powder of $MgCO_3$ is used as a precursor component for MgO .

25 According to the method a wet mixing of the component powders is proposed. In a wet mixing process less impurity is released from the mixing devices so that a high purity of the final product can be maintained. In order to avoid negative

effects on deflocculating caused by electrical charged ions of any of the components dissolved in the liquid, a liquid is used in which the raw material powders have low solubility. Low solubility in this respect means a solubility of the component which is as low as possible; that means here that the dissolved solid 5 content of the raw material components is less than 200 mg in kg liter of said liquid.

After drying the well mixed oxides of the slurry are supporting the formation of solid solutions between single oxides during sintering due to shorter diffusion paths. Further enrichment of single elements or oxides in the bulk is avoided by a 10 well distributed starting mixture.

A target is obtained comprising minimal amounts of single phase oxides. Such single phase oxides are negatively affecting the homogeneity of the target and consequently of the sputtering homogeneity. The microstructure is essentially composed of ZnO and ternary and binary oxides. It contains no single component 15 oxide phase consisting of either Al_2O_3 or of MgO or it contains such single component oxide phases in an amount of less than 1 % per weight.

The liquid used in the mixing step comprises water and powder of MgCO_3 is used as a precursor component for MgO . MgCO_3 has a low solubility in water compared to MgO . The specific maximum solubility of MgCO_3 in 1kg distillated water is less 20 than 200 mg.

MgCO_3 is a precursor material of MgO . In a calcination step prior to sintering MgCO_3 is oxidized forming MgO .

In a preferred modification of the method the mixing in the liquid comprises a wet milling process, whereby the wet mixing results in fine powders of the raw material 25 components having particle medium particle sizes (d_{90} value) of less than 10 μm , preferably less than 5 μm , but having a d_{50} -value of more than 1 μm .

Due to the small particle size, said powder has a high surface so that a high sinter activity of the powder mixture is achieved requiring temperatures for sintering which are lower than in the case of less sinter active powder mixtures. The lower

sintering temperature avoids excess grain growth during sintering and helps minimize the amount of oxide phase regions consisting of MgO or Al₂O₃. Nonetheless, the medium particle size of said powder is quite large (d₅₀-value > 1 µm) when compared to the prior art mentioned above. Therefore, cost-intensive 5 and impurity-sensitive milling procedures can be minimized.

In a preferred modification of the method a raw material powder is provided within the following composition ranges (in wt.-%):

Al₂O₃: 0,5-5, preferably 2-4
MgO: 0,5-15, preferably 3-9 (balance = ZnO).

10 With respect to the ZnO-Al₂O₃-MgO sputtering target, the aforementioned object is achieved according to the invention in that it comprises a microstructure composed of ZnO and ternary and binary oxides but containing single component oxide phase consisting of either Al₂O₃ or of MgO in an amount of less than 1 % per weight whereby the microstructure contains regions of single and binary 15 phases of zincite, gahnite (ZnAlO₄) and spinel (MgAlO₄).

In the ideal case, the microstructure of the target material completely consists of ZnO and of binary or ternary oxides each forming a solid solution of several elements. In practice, a certain amount of MgO or Al₂O₃ may be contained even if it is below detection limit of x-ray diffraction. An elemental analysis results in a 20 homogeneous distribution of Zn, Al, Mg and oxygen.

The target of the invention has a fine grained structure with homogeneous distribution of the elements belonging to ZnO-Al₂O₃-MgO in the bulk material.

However, it contains several crystalline phases. Commonly, e.g. in the prior art mentioned above, a solid solution without such crystalline binary or ternary 25 phases is wanted. In contrast to this, the microstructure of the sputtering target contains regions of single and binary phases of zincite, gahnite (ZnAlO₄) and spinel (MgAlO₄), whereby, preferably, the regions of gahnite phases have lateral dimensions smaller than 5 µm and the regions of spinel phases have lateral dimensions smaller than 1 µm. The lateral dimensions are medium values.

It was found that said zincite, gahnite ($ZnAlO_4$) and spinel ($MgAlO_4$) regions are a result of using relative coarse starting grains. Coarse grain requires relatively less time and energy for milling and it contains less impurity caused by wear and abrasion. Surprisingly, it was found that the multi-crystalline structure works very well. It was shown that instead of seeking an expensive solid-solution crystalline phase a much cheaper structure using coarser powders or shorter milling is also successful.

Said target is obtainable by the method of the invention as described above.

In a preferred modification of the sputtering target it is characterized by a composition ZnO 92 / Al_2O_3 3 / MgO 5 (in wt.%).

Besides negligible amounts below detection limit, this material contains no single component oxide phases consisting of either Al_2O_3 or of MgO but the microstructure almost consists of ZnO and of binary or ternary oxides each forming a solid solution of several elements.

In another preferred modification of the sputtering target the microstructure has a medium crystallite size below 5000 Angstrom preferably below 2000 Angstrom..

The crystallite size is measured by x-ray diffraction.. A small medium crystallite size in the sputtering target is result especially of a wet mixing process of the raw material. A fine grained microstructure gives better results in sputtering homogeneity.

In a preferred modification of the sputtering target the electrical resistivity of the target is less than 50 mOhm·cm, preferably less than 30 mOhm·cm.

In a preferred modification of the sputtering target it is made in form of tiles having a tile size larger than 100 cm^2 , preferably larger than 200 cm^2 .

This is a relative large tile size which can be achieved by using the claimed method. Therefore, the number of bond gaps in the target can be reduced.

Preferred embodiments

The invention will now be explained in more detail with reference to embodiments and a drawing, in which shows:

5 **Fig. 1** a diagram showing the particle size distribution of a slurry after finishing of the wet milling process,

Fig. 2 a diagram showing micro-arcing of a sputter target cathode (PK 500) made of ZAMO material produced by either by a dry mixing method according to the prior art and by a wet mixing process according to the invention,

10 **Fig. 3** a photo of a cross-section polish showing of a ZAMO material according to the invention after sintering (showing agglomerates and phases),

Fig. 4 a photo of a cross-section polish of a ZAMO material according to the invention after sintering and subsequent thermal etching (showing grain sizes), and

15 **Fig. 5** an XRD diagram of a ZAMO material according to the invention showing the occurrence of crystalline phases consisting of zincite, gahnite and spinel.

Example 1

A mixture of ZnO (86 wt.%), Al₂O₃ (3 wt.%) and MgCO₃ (11 wt.%) in water was prepared. A Na⁺ containing dispersant was added in order to stabilize the slurry 20 and to increase solid loading of the slurry, which was 55 wt.%.

The slurry was treated with a high energy ball mill using milling balls of ZrO₂, and diameter 1 mm. During milling energy of 0.025 kWh/kg slurry was applied. After milling a particle size measurement of the slurry showed a d₁₀ value less than 1 µm and a d₉₀ value less than 4 µm (see **Fig. 1**). The left ordinate axis in the 25 diagram of Fig. 1 represents the amount Q [in %]. The right ordinate gives the total sum S [in %] and the x-axis represents the particle size d in [µm].

After milling particle size measurement of the slurry showed a d10 value less than 1 µm and a d90 value less than 4 µm. Subsequently, a binder (i.e. PVA) was added and the material was spray dried. Agglomerates and granules of ZAMO were obtained. Said granules have a particle size between 20µm (d10 value) and 5 60µm (d90 value).

The granules were press shaped at 50 MPa and a tile of 380*150 mm² was achieved. Material was debindered and calcined. Calcination took place in the range up to 700°C by carefully heating with a rate of 5 K/h to avoid high internal gas pressures from CO₂ destroying the shaped green body. Then sintering in 10 ambient atmosphere up to 1400°C was performed.

A density of 5.2 g/cm³ was achieved. Elemental distribution of the elements Zn, Al, Mg, and oxygen was homogeneous and almost no singular phases of insulated Al₂O₃ und MgO in a ZnO based matrix were found. The composition contained 1% spinel, 4% gahnite and 95% zincite.

15 The photo of **Fig. 3** of this material shows agglomerates and phases of gahnite “G” and spinel “S”. The photo of **Fig. 4** of this material obtained after etching shows the grain sizes of the microstructure.

In the XRD analysis diagram of **Fig. 5** the measured defracted intensity I is plotted 20 against diffraction angle 2theta. It shows that the microstructure – within the detection limit - completely consists of ZnO and of binary or ternary oxides, namely gahnite and spinel. Besides ZnO, the diagram shows no line referring to a single component oxide phase consisting of MgO or Al₂O₃.

Comparative example 1

A mixture of a first powder of ZnO (90 wt.%) + MgO (10 wt.%) and a second 25 powder of Al₂O₃ (50 wt.%) + MgO (50 wt.%) (with particle sizes as example 1) were dry mixed and heat treated up to 1400°C.

X ray analysis of this mixed powder showed no crystalline single component phases of MgO nor Al₂O₃ but only spinel and zincite phase (with solid solution of MgO). The formed mixed powders were mixed with fresh ZnO to give a final composition of ZnO (90 wt.%), Al₂O₃ (3 wt.%) and MgO (6 wt.%) and a slurry of 5 this powder mixtures in water was prepared. A Na⁺ containing dispersant was added in order to stabilize the slurry and to increase solid loading of the slurry which was 55 wt.%.

Wet milling was applied as described for example 1. Then a binder (i.e. PVA) was added and the material was spray dried. Agglomerates or granules of ZAMO 10 formed having a size between 20µm (d10 value) and 60µm (d90 value). The granules were press shaped at 50 MPa and a tile of 380 mm x 150 mm.

Debinding occurred by heating with 10 K/h up to 600 °C and sintering in ambient atmosphere by heating up to 1400°C.

Comparative Example 2

15 A dry mixture of ZnO (92 wt.%), Al₂O₃ (3 wt.%) and MgO (5 wt.%) was homogenized in a tubular mixer for 5 h. During the mixing process in parallel milling was applied with a ZrO₂ milling balls, diameter 10 mm which were added to the mixing containment.

It was found that this milling process added 3000 wtppm ZrO₂ impurities in the 20 powder mixture.

The mixture was sieved and press shaped at 50 MPa to a size of 100 x100 mm². Larger tile sizes were impossible to shape since they fractured during handling/pressing.

The process was finished by sintering in ambient atmosphere up to 1450°C.

25 A density of 4.8 g/cm³ was achieved. Distribution of the elements Zn, AL, Mg, and oxygen was found to be inhomogeneous and singular phases of insulated Al₂O₃

und MgO in a ZnO based matrix were still found. Further the sintered bulk contained porosity in the macroscopic scale.

Evaluation of quality of sputtering targets and sputtering behaviour

The shaped tiles of example 1 and that of comparative example 1 were machined 5 and grinded and bonded to a backing copper plate. For examples 1 obtained by the wet mixing route according to the invention larger tiles sized 300 x 100 mm² with less bonding gaps per target could be prepared and used.

While the target material obtained by dry mixing route according to comparative example 1 ended up with a tile size 75 x 75 mm², so that consequently the target 10 was composed of multiple segments with a significantly higher amount of bonding gaps.

The targets were examined in a sputtering trail. First the target was ramped up in argon atmosphere. Then slight oxygen of 5 sccm was used for a pre-sputtering procedure. Within 30 min the power was increased up to 5,5 → 7,0 → 10 W/cm².

15 At each power level glass plates (80x30 mm²) were coated. The arcing was examined at the respective different power levels and the result is shown in the diagram of **Fig. 2**. On the y-axis in the diagram of Fig. 2 the occurrence of micro-arcs per second is plotted against the power P in [W/cm²]

The targets prepared according to the invention (type I) as well as the target 20 prepared by dry mixing of the raw material (type II) show similar sputter rates in the range of 50 nm*m/min.

However, only for the target prepared by dry mixing (type II) hardarcs were observed. At 10W/cm² nearly one hardarc per second was observed.

After sputtering of 36 hours at maximum power level the surface of targets of type 25 I and type II looked significantly different:

Type I: a smooth surface was observed.

Type II: a rough and ragged surface, with pins and dust deposition was achieved.

Patent claims

1. A method for producing a ZnO-Al₂O₃-MgO sputtering target, comprising the steps of mixing powders of raw material components comprising Al₂O₃, ZnO and MgO or precursors of these substances and sintering the powder mixtures so as to obtain the sputtering target, said step of mixing comprises a mixing of powders of the raw material components in a liquid, characterized in that the maximum solubility for said raw material powders in said liquid is less than 200 mg/kg and whereby said liquid comprises water, and powder of MgCO₃ is used as a precursor component for MgO.
- 10 2. The method of claim 1, characterized in that the mixing in the liquid comprises a wet milling process.
- 15 3. The method of claim 2, characterized in that the wet milling process results in fine powders of the raw material components having medium particle sizes (d₉₀ value) of less than 10 µm, preferably less than 5 µm, but having a d₅₀-value of more than 1 µm.
4. The method of any one of the preceding claims, characterized in that a raw material powder is provided within the following composition ranges (in wt.%):
Al₂O₃: 0,5-5, preferably 2-4
MgO: 0,5-15, preferably 3-9 (balance = ZnO).
- 20 5. A ZnO-Al₂O₃-MgO sputtering target characterized by a microstructure containing single component oxide phase consisting of either Al₂O₃ or of MgO in an amount of less than 1 % per weight whereby the microstructure contains regions of single and binary phases of zincite, gahnite (ZnAlO₄) and spinel (MgAlO₄).
- 25 6. The ZnO-Al₂O₃-MgO sputtering target of claim 5, characterized in a composition ZnO 92 / Al₂O₃ 3/ MgO 5 (in wt.%).

7. The ZnO-Al₂O₃-MgO sputtering target of claim 5, characterized in that the regions of gahnite phases have lateral dimensions smaller than 5 µm and the regions of spinel phases have lateral dimensions smaller than 1 µm.
8. The ZnO-Al₂O₃-MgO sputtering target of any one of the preceding claims 5 to 7, characterized in that the microstructure has medium crystallite size below 5000 Angstrom, preferably below 2000 Angstrom.
9. The ZnO-Al₂O₃-MgO sputtering target of any one of the preceding claims 5 to 8, characterized in that the electrical resistivity of the target is less than 50 mOhm·cm, preferably less than 30 mOhm·cm.
10. The ZnO-Al₂O₃-MgO sputtering target of any one of the preceding claims 5 to 9, characterized in that it is made in form of tiles having a tile size larger than 100 cm², preferably larger than 200 cm².

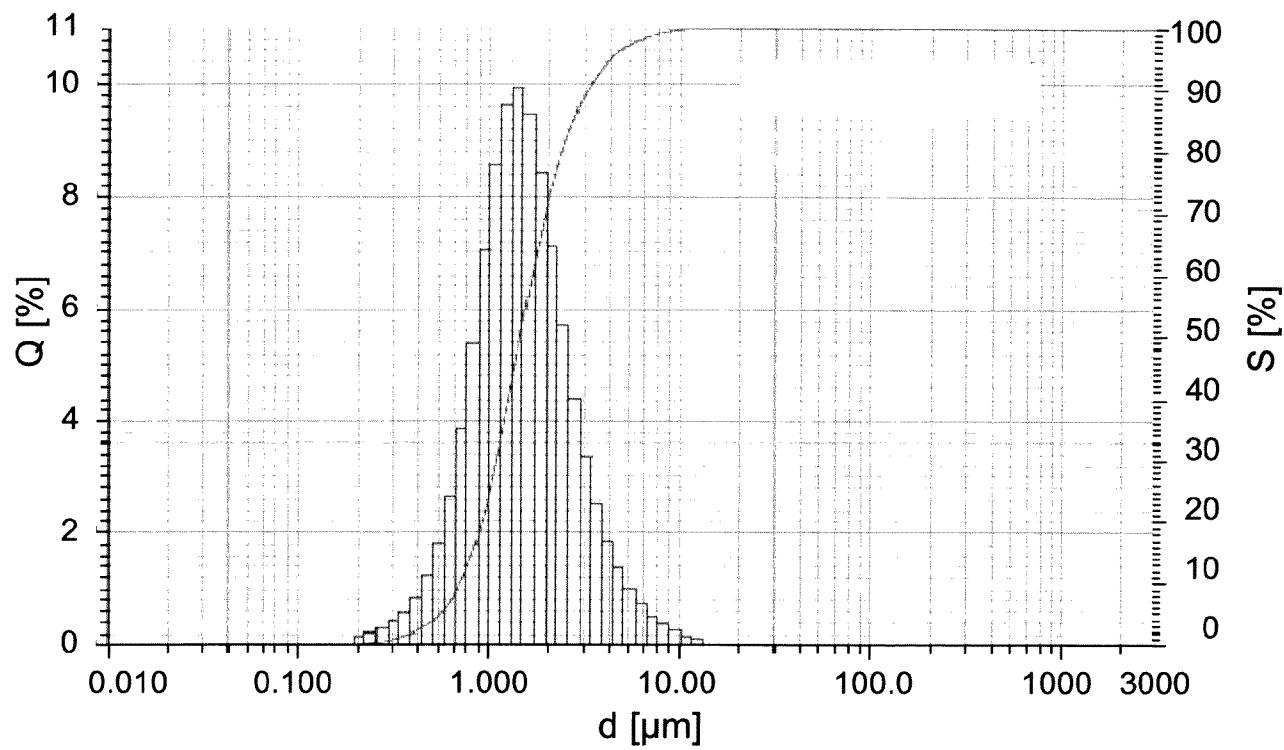


Fig. 1

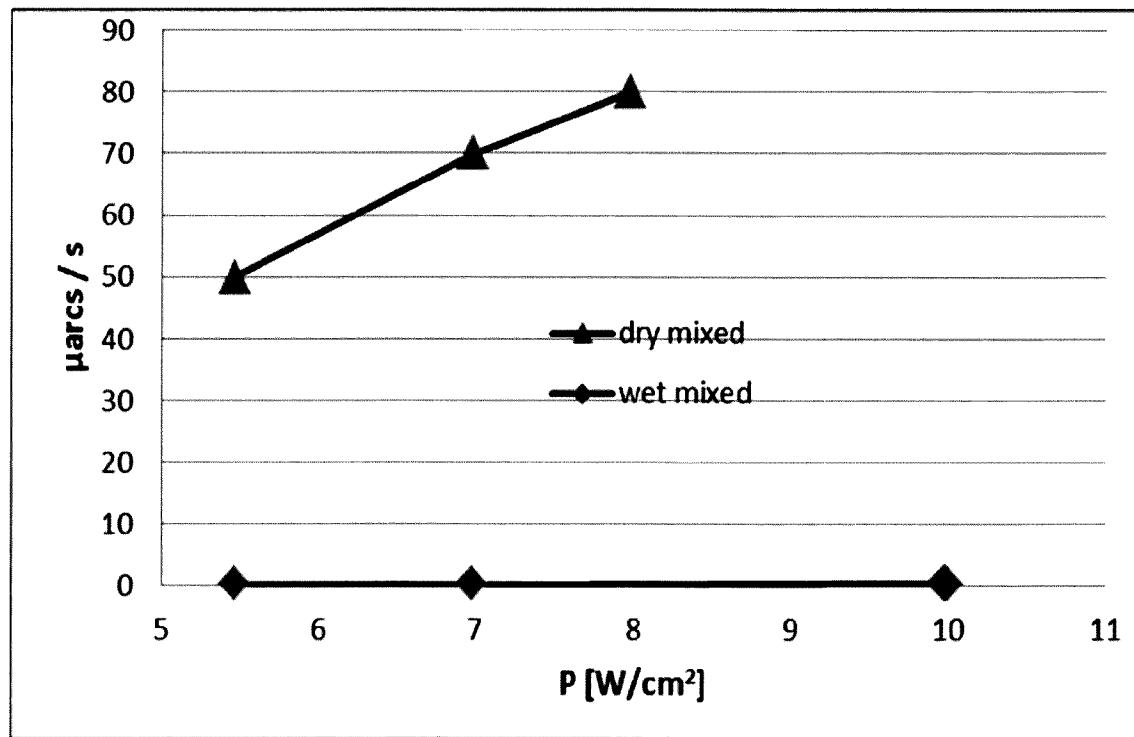


Fig. 2

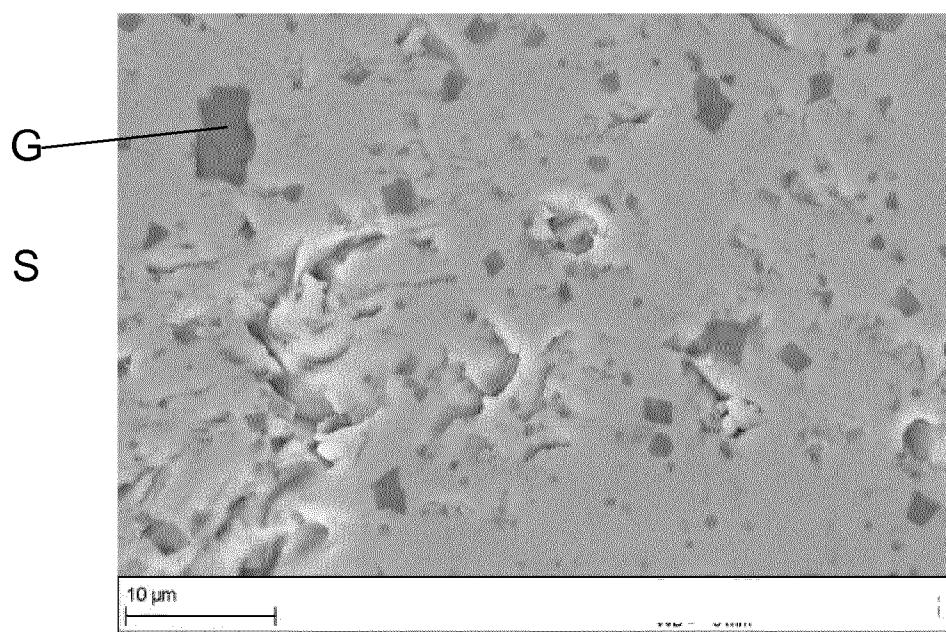


Fig. 3

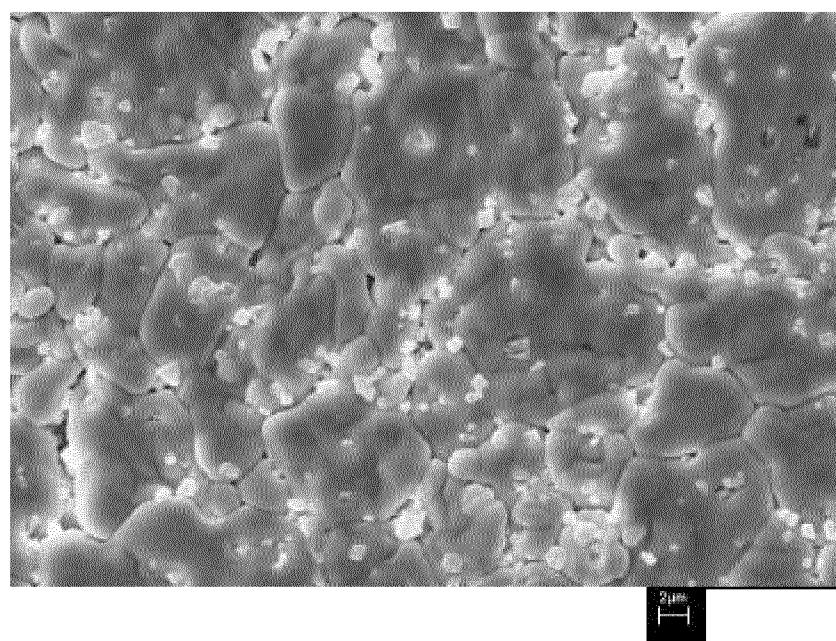


Fig. 4

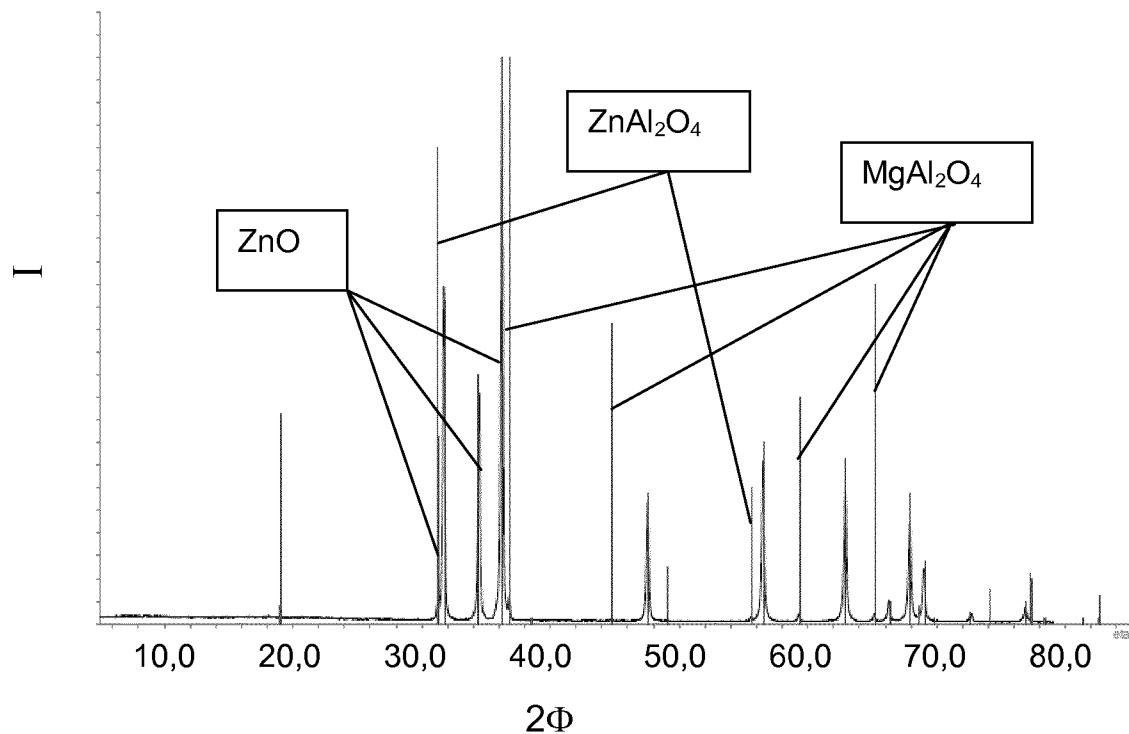


Fig. 5

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2014/053128

A. CLASSIFICATION OF SUBJECT MATTER
INV. C23C14/34 C04B35/453 C04B35/626
ADD.

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

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
C23C C04B

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

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

EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	<p>EP 2 514 851 A1 (MITSUBISHI MATERIALS CORP [JP]) 24 October 2012 (2012-10-24) cited in the application page 8, paragraph 36 - page 9, paragraph 40; table 1</p> <p style="text-align: center;">-----</p>	1,5



Further documents are listed in the continuation of Box C.



See patent family annex.

* Special categories of cited documents :

- "A" document defining the general state of the art which is not considered to be of particular relevance
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Date of the actual completion of the international search	Date of mailing of the international search report
11 April 2014	09/05/2014
Name and mailing address of the ISA/ European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016	Authorized officer Schuhmacher, Jörg

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.

PCT/EP2014/053128

Patent document cited in search report	Publication date	Patent family member(s)		Publication date
EP 2514851	A1 24-10-2012	CN 102666910 A	EP 2514851 A1	12-09-2012