

Method for producing a composite body of a material with a high silicic acid content

Description

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Technical Background

The present invention relates to a method for producing a composite body with a first layer of a material of a high silicic acid content with an additional component in a first concentration, connected to a second layer of a material of a high silicic acid content with an additional component in a second concentration that differs from
10 the first concentration, and the first and the second concentration are greater than or equal to zero.

Prior Art

A material with a high silicic acid content is here understood to be doped or undoped quartz glass with a SiO_2 content of at least 85%. This material will also
15 briefly be called "quartz glass" in the following. Quartz glass is distinguished by a low coefficient of thermal expansion, by optical transparency over a wide wavelength range and by high chemical and thermal resistance.

To achieve special properties, quartz glass is doped with other substances, such as titanium, aluminum, boron or germanium. These substances are added in very
20 small amounts and act as foreign atoms in the glass structure without forming a second phase in addition to the matrix.

A quartz glass which contains an additional component is here designated as "black glass", wherein a second phase of carbon, silicon, silicon carbide, silicon nitride, titanium nitride or titanium carbide is intercalated in a matrix of quartz glass
25 and consolidated into a composite material. The fine-particle regions of the intercalated phase (e.g. Si phase or carbon phase) act as optical defects and have the effect that the composite material has a black or gray visual appearance at room temperature. On the other hand, the defects also have impacts on the heat absorption or heat emission of the composite material on the whole. Thermal
30 radiation between $2\text{ }\mu\text{m}$ and $8\text{ }\mu\text{m}$, i.e. in the wavelength range of the infrared radiation, is strongly absorbed. The high absorption and emission capacity for

thermal radiation reduces the reflection thereof on the surfaces of the composite material. Local, non-reproducible heating up by reflected thermal radiation is thereby prevented and a uniform temperature distribution is achieved in the environment of the composite material. The high degree of emission of the composite material makes the component particularly suitable for use in thermal treatments where a reproducible and homogeneous temperature distribution is of importance.

Frequently, there is a need to connect quartz glass elements to one another, for example for the manufacture of quartz glass components having a complex shape. Commonly, this connection is provided by welding the components to one another. For example EP 1 042 241 B1 describes a method for the butt-like welding of quartz glass tubes. Welding comprises melting the surfaces to be connected to one another and pressing the softened surfaces against one another, which may easily lead to undesired plastic deformation in the area of the welding zone.

Though it is possible to remove deformations of this kind by extensive after-treatment, some dimensional deviations usually remain.

Further difficulties arise during the welding of black glass: During heating the temperature is possibly limited to values below the melting point of the intercalated phase (Si phase: 1410°C). In the case of intercalated carbon the limiting factor is that carbon detaches from the free surface due to strong heating or it oxidizes or burns off due to the high process temperatures; this may lead to the formation of bubbles. Moreover, due to the above-mentioned high emission degree of the black glass most of the heat that has been introduced by a welding torch is emitted again, so that the material is shining in a brightly glowing manner and rapidly cools down again as compared with transparent or opaque quartz glass. A bonding or shaping of black glass is therefore not possible by way of hot working. It is known from EP 2 048 121 A1 that a component of black glass is provided on all sides of the surface with a layer of transparent quartz glass. Thanks to the layer it is possible to process the component of black glass in hot processes, specifically to weld it to another quartz glass component. The transparent outer layer is generated by subjecting a porous SiO₂ soot body to a gas phase reaction with organic silicon compounds, followed by vitrification under vacuum. The carbon component of the organic Si compound cannot diffuse out of the core region of the

soot body during vitrification, resulting in a component of black glass with a carbon phase in the core region (carbon content 30 wt. ppm to 50,000 wt. ppm) and with a carbon-free (less than 30 wt. ppm carbon), transparent outer layer. The outer layer has a layer thickness in the range of one millimeter to 10 millimeters, the layer thickness depending on the negative pressure set during vitrification of the component. The setting of the layer thickness and the adjustment of the carbon amount in the core region of the component turn out to be complex, so that the method according to EP 2 048 121 A1 must be considered to be complicated on the whole. Moreover, this will only yield a component of black glass that is provided on all sides with a transparent outer layer; the selective formation of only one connecting surface is not possible.

A component of black glass with a transparent outer layer is also disclosed in US 2014/0072811 A1. A plate of black glass is here brought into contact with a SiO₂-containing slurry mass after sintering at 1500°C, so that the plate is provided on all sides with a thin slurry layer. This is followed by a temperature treatment at 1000°C and 1600°C, respectively, resulting in a densely sintered plate of black glass with a transparent, bubble-free outer layer having a thickness of 0.1 mm. Since an already sintered plate is used, the adhesion of the slurry mass to the surface is relatively poor, so that one obtains only a thin slurry layer and, as a result thereof, also only a 0.1 mm thin, transparent outer layer. Such a thin outer layer is not sufficient for subsequent bonding by welding because, as has been explained above, the heat of a welding torch immediately penetrates the outer layer and acts on the black glass positioned thereunder, it is there emitted again and the component thus cools down all in all too rapidly to allow bonding with another material having a high silicic acid content.

Furthermore, DE 10 2004 054 392 A1 discloses a method for connecting components consisting of a material with a high silicic acid content, in which method a pourable or pasty slurry mass which contains amorphous SiO₂ particles is applied to individual connecting surfaces of densely sintered quartz glass components. Immediately thereafter, the connecting surfaces are fixed with respect to or onto each other. The bonding mass that is more or less enclosed between the two connecting surfaces is then dried. A correspondingly slow drying process yields a dried layer without cracks that is suited for connecting relatively

small plates. Said method is no longer suited for large-area connections with a correspondingly high pressing pressure already due to the dead weight of the quartz glass parts to be connected, or it would require long and uneconomic drying times to remove the dispersing agent of the bonding mass from the bonding site
5 without any defects

Technical objective

It is thus the object of the present invention to indicate a method that permits an inexpensive manufacture of a mechanically and thermally stable composite body
10 of a material with a high silicic acid content. Specifically, it is the aim to provide large-area bonding connections by way of welding.

General description of the invention

This object is achieved by the method according to the invention comprising the
15 following method steps:

- (a)** preparing a first slurry layer having a free surface by using a first slurry mass containing a first dispersing agent and, dispersed therein, first SiO_2 particles and the additional component in a first concentration,
- (b)** providing a second slurry mass containing a second dispersing agent and,
20 dispersed therein, second SiO_2 particles and the additional component in a second concentration that differs from the first concentration,
- (c)** forming a composite-body intermediate product by applying the second slurry mass to the free surface of the first slurry layer, and
- (d)** heating the composite-body intermediate product while forming the composite
25 body.

In the method according to the invention the manufacture of the composite body from materials of a high silicic acid content is based on the measures that after providing a first slurry layer of a SiO_2 -containing slurry mass a second SiO_2 -containing slurry mass is applied to the first one and the two slurry layers are
30 jointly sintered or vitrified.

The first slurry layer must be understood within the meaning of the method according to the invention as an intermediate product for a base body layer, or for a functional layer of the composite body.

5 The preparation of a first slurry layer comprising a free surface is carried out by using a first SiO_2 slurry mass that when poured out inherently forms a smooth free surface. The dispersing agent of the slurry mass is still substantially present in this slurry layer, so that the first slurry layer does here not correspond to a porous green body mass, as is normally created during drying of at least a part of the dispersing agent of a slurry mass. The surface of the slurry layer forms a planar
10 boundary or connecting surface to which the second slurry mass can subsequently be applied without the two slurry masses being intermixed. The composite-body intermediate product is thus created by two mutually contacting layers of SiO_2 -containing slurry masses that only differ slightly in their moisture content (proportion of dispersing agent) or solids content. The dispersing agent acts here
15 on the boundary surface as a kind of "binding phase" between the first slurry layer and the second SiO_2 slurry mass. This procedure has the advantage that the two slurry masses show a similar shrinkage behavior during subsequent heating.

Heating comprises expelling the dispersing agent out of the two slurry masses and a sintering or vitrifying phase, whereby a multilayered composite body of a
20 material of a high silicic acid content is obtained as a solid component. The composite body comprises a first layer of a material of a high silicic acid content with an additional component in a first concentration that is connected to a second layer of a material of a high silicic acid content with an additional component in a second concentration that differs from the first concentration. The second layer is
25 able due to its share in the additional component to form a connecting surface with a further component of a material of a high silicic acid content. The composite body is created in a single hot process, whereby the method according to the invention allows an inexpensive production.

By contrast, additional hot processes, such as the subsequent welding of two,
30 already sintered components or the vitrification of a slurry mass on a densely sintered or pre-sintered basic body, are required in the methods according to the prior art.

A dispersion or suspension of SiO_2 particles in a liquid, e.g. water or alcohol or a mixture of said substances, is called SiO_2 -containing slurry mass. The proportion of SiO_2 particles may be the same in both slurry masses or may differ by up to 5% by wt.

- 5 Each of the first and the second SiO_2 -containing slurry masses contains additional components that may be the same or may be different in their type. The concentration of the additional component in the second slurry mass differs from the concentration in the first slurry mass, wherein the first and the second concentration is greater than or equal to zero. This means that the concentration
10 of the additional component in one of the two slurry masses may also be zero.

Doping in the sense of the introduction of foreign atoms into a SiO_2 matrix is here not regarded as an additional component. Rather, an addition of substances in amounts suited to macroscopically form a second phase, besides the SiO_2 matrix, which leads to targeted changes in physical and chemical properties of the
15 component produced thereby, is understood as the additional component.

The solids content of the two SiO_2 -containing slurry masses is substantially determined by the proportion of SiO_2 particles. In addition, the additional component accounts for a further proportion of the solid. The SiO_2 proportion in the solid is at least 85% by wt. to obtain a mechanically and thermally stable
20 composite body of a material of a high silicic acid content.

In a preferred procedure, the first and the second SiO_2 -containing slurry masses differ in their solids content from each other by not more than 5% by wt. In the case of great differences in the solids content of the two SiO_2 slurry masses, great differences in the shrinkage of the two masses may arise during the joint heating
25 and sintering, and these may lead to cracks and other defects in the composite body.

It has turned out to be useful that each of the SiO_2 -containing slurry masses has a solids content of at least 65% by wt., preferably of at least 75% by wt. In slurry masses with solids contents of this type the shrinkage caused by drying and
30 sintering is relatively small, which reduces the risks arising during manufacture and contributes to an inexpensive production. Moreover, this improves the form stability and dimensional accuracy of the composite body.

Advantageously, the preparation of the first slurry layer according to method step (a) and the formation of the composite-body intermediate product according to method step (c) are carried out by using an absorbent mold into which the first SiO₂ slurry mass and the second SiO₂ slurry mass are introduced one after the other. The dispersing agent contained in the slurry masses is absorbed by the absorptive mold at least in part and accelerates the process for forming the composite body. The mold is here configured such that after introduction of the first slurry mass said mass forms a slurry layer with a large-area free surface in the mold and the second slurry mass is applied thereto. The use of an absorbent mold, as is e.g. common as a plaster mold in the slip casting method, facilitates the production of a composite body close to the final contour. The absorbent mold in the method according to the invention, however, may also comprise sub-areas of a non-absorbent material, whereby the body formation into a specific spatial direction can be influenced in a targeted manner.

To minimize a mixing risk during over-coating of the first slurry layer with the second slurry mass, it has turned out to be useful when the preparation of the first slurry layer according to method step (a) comprises a settling phase of at least 30 minutes, after which a supernatant of first slurry mass is removed from the surface of the first slurry layer. In the preparation of the first slurry layer, settling or consolidation of the first slurry mass by slow removal of the dispersing agent takes place in a time-dependent manner, resulting in a first slurry layer over which there is still a supernatant of the first slurry mass. This supernatant is e.g. removed by suction, and there remains the free surface of the first slurry layer that has been subjected to stabilization by way of the settling or consolidation phase. This avoids penetration of the surface of the first slurry layer by the pouring stream in the subsequent application of the second slurry mass. The first slurry layer is also called green body mass or body.

It has further turned out to be useful to carry out the application of the second SiO₂-containing slurry mass by means of a pouring stream guided at an angle in the range of 3° to 15° relative to the free surface of the first slurry layer. Moreover, the pouring stream advantageously has an elongated shape in horizontal cross-section. These two measures have the effect that the forces acting on the free surface of the first slurry layer by way of the pouring stream of the second SiO₂-

containing slurry mass are reduced, so that a substantially flat, horizontal surface is maintained and the pouring stream does not penetrate into the slurry layer formed by the first slurry mass. A rectangular shape with rounded corners is here regarded as an elongated shape. The angle adjustment upon supply of the second

5 SiO₂-containing slurry mass and the elongated form of the cross section of the pouring stream can be achieved with the help of a pouring channel that has a width which is adapted to the surface expansion of the first slurry layer and is at least 50 mm.

The two SiO₂-containing slurry masses preferably contain amorphous SiO₂

10 particles with particle sizes in the range of 1 µm to 50 µm which are present in a splintery and/or spherical form. Particularly preferred are splintery SiO₂ particles in this particle size range and with a wide size distribution for the reason that a good interlock with one another and an advantageous sintering behavior together with a comparatively low shrinkage can thereby be achieved. Moreover, it has been

15 found that in such SiO₂-containing slurry masses a high green strength of the composite-body intermediate product is also obtained.

In a preferred embodiment of the method according to the invention silicon particles and/or carbon particles are contained as an additional component in the SiO₂-containing slurry mass. This leads to the formation of at least one layer of the

20 composite body to be finished from a material of a high silicic acid content with high heat absorption and heat emission. The SiO₂-containing slurry mass that contains silicon and/or carbon particles as additional component forms a layer of the composite body in the form of a composite material that has a matrix which is either opaque, but preferably translucent or transparent. Portions of a phase which

25 contains silicon (Si) in elemental form and is here called "Si phase" or contains carbon particles are intercalated into the matrix with a distribution that is as fine as possible. The fines portions of the intercalated phase act as optical defects in the matrix on the one hand and have the effect that the composite material has a black or gray-blackish visual appearance at room temperature, depending on the

30 thickness, so that this layer is called "black glass". As has already been explained above, the defects also have impacts on the heat absorption or heat emission of the composite material on the whole. Thermal radiation between 2 µm and 8 µm, i.e. in the wavelength range of the infrared radiation, is strongly absorbed. Thus a

bonding of such a material by means of standard welding methods is not possible or is only possible to an inadequate degree.

The heat absorption of the composite material depends on the portion of the Si phase or on the portion of the carbon phase. The greater their portion, the higher are absorption capacity and emission capacity. The weight proportion of the Si phase is preferably at least 0.1% by wt. On the other hand, a particularly high volume fraction of the Si phase may aggravate the production of the composite material. In view of this, the weight proportion of the Si phase is preferably not more than 5%.

- 5 This also applies by analogy to the carbon phase. Its share in the solids content of the first or the second SiO_2 -containing slurry mass is preferably in the range of 0.01% by wt. to 5% by wt.

- As regards the heating of the composite-body intermediate product for the formation of the composite body, it has turned out to be advantageous when this takes place at a temperature of not more than 1,400°C. This maximum temperature is particularly of importance when one of the SiO_2 slurry masses to be processed contains a Si phase because a situation must be avoided where the Si phase fuses. The composite-body intermediate product is kept at the sintering temperature for a period of at least 2 hours.

- 20 The method according to the invention allows the production of a mechanically and thermally stable composite body with two layers that conform to a base body layer connected to a functional layer. Such a base body layer of a material having a high silicic acid content contains an additional component of silicon and/or carbon particles, whereas the second layer can be designated as a functional layer of a material of a high silicic acid content without such an additional component. This functional layer allows hot working, for instance by using a welding torch.

Embodiment

- 30 The invention will now be explained in more detail with reference to a patent drawing and an embodiment. In detail,

Fig. 1 is a flow diagram for explaining the method according to the invention;

Fig. 2 is a sectional view of a composite body produced according to the method according to the invention.

The method according to the invention will now be explained by way of example
5 with reference to the production of a carrier of quartz glass for a reactor for the treatment of a wafer with reference to Fig. 1 and 2.

Example 1

Preparation of a first SiO₂ slurry mass with additional component

10 For a batch of 10 kg of the first slurry mass (base slurry) 1, 8.2 kg of amorphous quartz glass grains 2, which have been obtained by melting natural quartz raw material and have grain sizes in the range between 250 µm and 650 µm, are mixed with 1.8 kg deionized water 3 with a conductivity of less than 3 µS in a drum mill which is lined with quartz glass and has a volume of about 20 liters. The
15 quartz glass grains 2 were previously purified in a hot chlorination method; attention is paid that the cristobalite content is below 1% by wt.

This mixture is milled by means of grinding balls of quartz glass on a roller block at 23 rpm for a period of 3 days to such an extent that a homogeneous, first base slurry 1 with a solids content of 82% is formed. The SiO₂ particles of the quartz
20 glass grains 2 milled in this way have a splintery grain form. During the grinding process the pH is lowered to about 4 due to the dissolving SiO₂.

Subsequently, the grinding balls are removed from the base slurry 1 obtained in this way, and an additional component in the form of silicon powder 4 with a metallic purity of 99.99% is admixed in an amount until a solids content of 83% by
25 wt. is obtained.

The silicon powder 4 consists of substantially non-spherical powder particles with a narrow particle size distribution, the D₉₇ value of which is about 10 µm and the fines content of which with particle sizes of less than 2 µm was removed before. The silicon powder 4 is uniformly distributed by continued mixing in the base slurry
30 1.

The first slurry mass 5 filled with the silicon powder 4 is homogenized for another 12 hours. The homogeneous first slurry mass 5 obtained thereby has a solids content of 83%. The weight percentage of the silicon powder as an additional component in the total solids content is 2% and the volume fraction is also almost 2% because of the similar specific densities of SiO_2 and Si. The SiO_2 particles 2 in the finished homogenized slurry mass 5 have a splintery form and show a particle size distribution that is distinguished by a D_{50} value of about 8 μm and by a D_{90} value of about 40 μm .

10 Preparation of the second SiO_2 slurry mass without additional component

A second SiO_2 slurry mass 6 is prepared that serves the generation of the functional layer of the composite body 8 and is without an additional component. The SiO_2 base slurry specified above in more detail is here used as the second SiO_2 slurry mass 6.

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Preparation of a plate-shaped composite body

The first slurry mass 5 with the additional component of silicon particles 4 is poured into a trough-shaped plaster mold where it forms the first slurry layer 7 with a free surface of the poured-in first slurry mass 5. The first slurry layer 7 forms a base body layer 10 of the composite body 9. The first slurry mass 5 occupies about 2/3 of the possible filling height of the plaster mold. The cavity mold allows a plate geometry with the green dimensions of 500 mm x 500 mm.

Without any substantial time delay the second slurry mass 6, which besides the SiO_2 particles 2.1 is without any additional component, is subsequently slowly applied to the free surface of the first slurry layer 7. When the second SiO_2 slurry mass 6 is poured out, the pouring stream is guided by use of a pouring channel such that it is applied at an angle of 5° , based on the surface of the first slurry layer 7, to said layer. The pouring channel has a width of 450 mm adapted to the plate geometry and is guided at a minimal distance from the first slurry layer 7 in the range of about one millimeter to 5 mm over said layer. Viewed in the horizontal cross section, the pouring stream produced thereby has the form of a rounded rectangle with the dimensions $450 \times 5 \text{ mm}^2$. It is thereby ensured that the pouring

stream of the second SiO_2 slurry mass 6 does not penetrate the substantially flat surface of the first slurry layer 7, which is formed by the first SiO_2 slurry mass 5, as the boundary. The second SiO_2 slurry mass 6 occupies about half the remaining filling height of the plaster mold and forms a uniform closed slurry layer with a thickness of about 30 mm.

It is also possible to first pour the SiO_2 slurry mass 6 without additional component into the casting mold and then to apply the SiO_2 slurry mass 6 with additional component thereto.

The first slurry layer 7 and the second SiO_2 slurry mass 6 applied thereto jointly form the composite-body intermediate product 8 that after mold casting and removal from the plaster mold is subjected to drying to form a green body. The plaster mold absorbs the dispersing agent up to a residual moisture of about 8%. The composite-body intermediate product 8 is then dried in a ventilated furnace at about 90°C for five days. This is followed by sintering or vitrification in a sintering furnace in air, wherein the composite-body intermediate product 8 is heated to a heating temperature of 1390°C within one hour and kept at this temperature for 5 h. Cooling is carried out with a cooling ramp of $1^\circ\text{C}/\text{min}$ to a furnace temperature of 1000°C and then unregulated in the closed furnace.

The composite body 9 obtained thereby is shown in the sectional view of Fig. 2. It comprises a base body layer 10 of a material of a high silicic acid content with 2 vol.% additional component of Si phase and a functional layer 11 of pure quartz glass formed from the SiO_2 slurry mass 6. The composite body 9 is a plate with the following final dimensions: 450 x 450 x 54 mm.

In a matrix of opaque quartz glass the base body layer 10 of the composite body 9 shows non-spherical portions which are separated from one another and consist of semi-metallic Si phase and are homogeneously distributed, and the size and morphology of which largely correspond to those of the Si powder used. The maximal dimensions of the Si phase portions are on average (median value) in the range of about 1 to $10\text{ }\mu\text{m}$. The intercalated Si phase leads to the black coloration of the base body layer 10 and has the effect that this layer shows a high absorption of thermal radiation at a high temperature. The latter means that a

bonding of the layer with intercalated Si phase by way of hot treatment is not possible because heat introduced by a welding torch is emitted again, so that the material is shining in a brightly glowing manner and will rapidly cool down again by comparison with transparent or opaque quartz glass. This flaw is compensated by the functional layer 11 of the composite body 9 that is arranged on a side surface with a thickness of 20 mm. It consists of opaque quartz glass and has a closed fine porosity of about 5 vol.% with a pore size of $< 20 \mu\text{m}$. This area of the composite body 9 can now be used for bonding by heat treatment.

10 Example 2

For the production of a composite body according to the invention from a first slurry mass 5 containing silicon as the additional component and from a second slurry mass 6 without additional component, two slurry masses are used that, except for the solids content, conform to the first and the second slurry mass of Example 1. The solids content of the two slurry masses is here however about 80% by wt. each time.

The first slurry mass 5 with Si as the additional component is filled into a casting mold for producing a square plate that only comprises an absorbent bottom, but the walls of which consist of non-absorbent material. The casting height is 4 cm. A body which corresponds to the first slurry layer 7 is progressively built up from the bottom to the top in the casting mold due to the absorbent bottom surface. After a waiting period of 3 hours the desired body height of about 3 cm is obtained, but a layer with a thickness of 5 to 10 mm, which consists of a still liquid slurry mass, remains on the surface. This residual slurry mass is sucked off by means of a pump and/or by dabbing with an absorbent soft material, so that a body having a solids content of about 89% by wt. is obtained from the first slurry mass, onto which the second SiO_2 slurry mass 6 (without additional component) is then cast. The casting height of the second slurry mass 6 is 10 cm. The body from the first slurry layer 7 and the second slurry mass 6 applied thereto will jointly form the composite-body intermediate product 8 which, as described in Example 1, is dried and sintered and thereby forms the composite body 9.

Example 3

In this case the first slurry mass 5 contains carbon as the additional component 4. For a batch of 10 kg of the first slurry mass 5, 8.2 kg of amorphous quartz glass grains 2 which were obtained by melting natural quartz raw material are mixed with 1.8 kg deionized water 3 with a conductivity of less than 3 μS in a drum mill which is lined with quartz glass and has a volume of about 20 liters. The quartz glass grains 2 were previously purified in a hot chlorination method; attention is paid that the cristobalite content is below 1% by wt. The quartz glass grains 2 have a mean particle size below 70 μm .

This mixture is milled by means of grinding balls of quartz glass on a roller block at 23 rpm for a period of 3 days to such an extent that a homogeneous, first base slurry 1 with a solids content of 80% is formed. During the grinding process the pH is lowered to about 4.5 due to the dissolving SiO_2 .

Subsequently, the grinding balls are removed from the base slurry 1 obtained thereby, and carbon nanoparticles, such as carbon black, are admixed as additional component 4 in an amount of 1.2% by wt. The solids content of the first SiO_2 slurry mass 5 obtained thereby is then about 81% by wt.

In the preparation of the second SiO_2 slurry mass 6, spherical SiO_2 particles 2.1 are used that are distinguished by a multimodal particle size distribution with a relatively narrow maximum of the size distribution at about 15 μm (D_{50} value). A secondary maximum is in the range of about 2 μm . These SiO_2 particles with a D_{50} value at 15 μm will be called R_{15} in the following. Moreover, further SiO_2 grains are used that have D_{50} values at 5 μm and 30 μm . These grains are called R_5 and R_{30} , respectively, depending on their D_{50} value.

These SiO_2 grains are purified in a hot chlorination process in advance. The impurity content of the purified grains is low and is less than 1 wt. ppm on the whole. Especially the content of Li_2O is less than 10 wt. ppb.

The following formulations have turned out to be useful:

Formulation 1

R_{30}	250 g
R_{15}	500 g
R_5	200 g

The said components are dispersed in water, resulting in a solids content of 86% by wt.

Formulation 2

R₁₅ 400 g

5 R₅ 90 g

Fumed silica: 10 g with BET surface area of 200 m²/g.

The said components are dispersed in the case of formulation 2 in pure ethanol, resulting in a solids content of 84% by wt.

- 10 The highly filled second slurry masses 6 produced in this way show a thixotropic behavior. They can be cast and spread and are therefore well suited for the processing technique according to the invention.

The manufacture of a plate-shaped composite body from the first slurry mass 5 with carbon nanoparticles as the additional component and the second slurry
15 mass 6 with SiO₂ grains according to formulation 1 or 2 is carried out according to the process sequence and by use of the same plaster mold geometry as in Example 1.

In this case the base body layer 10 of the composite body 9 shows carbon
particles which are homogeneously distributed in a matrix of opaque quartz glass
20 and which lead to the black coloration of the base body layer 10 and have the effect that it shows a high absorption of thermal radiation at a high temperature. The latter means that a bonding of the layer with intercalated C phase is not possible by means of hot working as heat introduced by a welding torch is emitted again so that the material is shining in a brightly glowing manner and rapidly cools
25 down again by comparison with transparent or opaque quartz glass. This flaw is compensated by the functional layer 11 of the composite body 9 which is arranged on a side surface with a thickness of 10 mm. It consists of opaque quartz glass. This area of the composite body 9 can now be used for bonding by hot working.

Patent claims

- 5 1. A method for producing a composite body with a first layer of a material of a high silicic acid content with an additional component in a first concentration, connected to a second layer of a material of a high silicic acid content with the additional component in a second concentration that differs from the first concentration, and the first and the second concentration are greater than or
10 equal to zero, comprising the following method steps:
(a) preparing a first slurry layer having a free surface by using a first slurry mass containing a first dispersing agent and, dispersed therein, first SiO₂ particles and the additional component in the first concentration,
(b) providing a second slurry mass containing a second dispersing agent and, dispersed therein, second SiO₂ particles and the additional component
15 in a second concentration that differs from the first concentration,
(c) forming a composite-body intermediate product by applying the second slurry mass to the free surface of the first slurry layer, and
(d) heating the composite-body intermediate product while forming the
20 composite body.
2. The method according to claim 1, characterized in that the first and the second SiO₂-containing slurry mass each have a solids content of at least 65% by wt., preferably of at least 75% by wt.
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3. The method according to claim 1 or 2, characterized in that the first and the second SiO₂-containing slurry mass differ in their solids content from each other by not more than 5% by wt.
- 30 4. The method according to any one of the preceding claims, characterized in that the preparation of the first slurry layer according to method step (a) and the formation of the composite-body intermediate product according to

method step c) are carried out by using an absorbent mold into which the first SiO₂ slurry mass and the second SiO₂ slurry mass are introduced one after the other.

- 5 5. The method according to any one of the preceding claims, characterized in that the preparation of the first slurry layer according to method step (a) comprises a settling phase of at least 30 minutes, after which a supernatant of the first slurry mass is removed from the surface of the first slurry layer.
- 10 6. The method according to any one of the preceding claims, characterized in that the second SiO₂-containing slurry mass is applied by means of a pouring stream guided at an angle in the range of 3° to 15° relative to the free surface of the first slurry layer.
- 15 7. The method according to claim 6, characterized in that the pouring stream in a horizontal cross-section has an elongated shape.
- 20 8. The method according to any one of the preceding claims, characterized in that the first and the second SiO₂-containing slurry mass contains amorphous SiO₂ particles with particle sizes in the range of 1 µm to 50 µm that are present in a splintery and/or spherical form.
- 25 9. The method according to any one of the preceding claims, characterized in that silicon particles and/or carbon particles are contained as the additional component in the SiO₂-containing slurry mass.
- 30 10. The method according to claim 9, characterized in that the Si particles are contained in the SiO₂-containing slurry mass with a solids content in the range of 0.1% by wt. to 5% by wt.
11. The method according to claim 9, characterized in that the carbon particles are contained in the SiO₂-containing slurry mass with a solids content in the

range of 0.01 % by wt. to 3% by wt.

12. The method according to any one of claims 9 to 11, characterized in that the heating process is carried out at a temperature of not more than 1400°C while forming the solid composite body.

13. The method according to any one of claims 9 to 11, characterized in that the SiO₂-containing slurry mass which contains silicon particles and/or carbon particles forms a base body layer of the composite body.