

DESCRIPTION

TITLE OF THE INVENTION:

ALUMINA POWDER, PROCESS FOR PRODUCTION OF THE SAME, AND RESIN COMPOSITIONS CONTAINING THE SAME

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TECHNICAL FIELD

The present invention relates to an alumina powder having a high average circularity and an improved flowability at the time of being highly filled, a process for its production and a resin composition employing it.

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BACKGROUND ART

In recent years, along with the progress in high functionality and high speed of heat-generating electronic components such as IC, the amount of heat generation of electronic devices having such components has been increasing, and a high heat dissipating property is desired also for a sealing material for semiconductors. In order to increase the heat dissipating property of a sealing material for semiconductors, an alumina powder having a high thermal conductivity may be incorporated to a rubber or resin. However, by a common Bayer process alumina powder, it was not possible to sufficiently utilize the thermal conductivity of the alumina, because of a remarkable rising viscosity phenomenon at the time of being highly filled.

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In order to solve such a problem, a method has been proposed to spray an aluminum hydroxide powder or an aluminum hydroxide powder slurry into flames from a feed pipe having an intensive dispersing function to obtain spherical alumina powder particles (Patent Document 1). The spherical alumina particles obtained by this method had surface irregularities derived from aluminum hydroxide raw material, even with particles having an average sphericity of at least 0.90, and thus, they are still desired to be improved. Further, also in a case where Bayer process alumina powder is used as a raw material, the product has, on its surface, irregularities derived from the raw material and thus is still desired to be improved.

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PRIOR ART DOCUMENT

PATENT DOCUMENT

DISCLOSURE

OBJECT TO BE ACCOMPLISHED

5 It is an object of the present disclosure to provide an alumina powder having a high average circularity and having an improved flow property at the time of being highly filled, a process for its production, and a resin composition employing it.

MEANS TO ACCOMPLISH THE OBJECT

10 The present disclosure provides the following to accomplish the above object.

In a first aspect of the present disclosure, there is provided an alumina powder which contains an α -phase and a θ -phase as main crystals, and has an α -phase content of at most 40% as measured by the following method, an average circularity of at least 0.95 in each of the particle size range of from 45 to 200 μm and the particle size range
15 of less than 45 μm , and an average particle size of at most 100 μm :

[method for measuring α -phase content]

Powders prepared by mixing α -phase alumina powder and θ -phase alumina powder in mass ratios of 0:10, 5:5 and 10:0, respectively, are subjected to X-ray diffraction measurements, whereupon the integrated intensity of a peak of α -phase detected in the
20 vicinity of $2\theta=43^\circ$ is calculated, and a calibration line of the integrated intensity plotted against the mixing ratio is made; and then, a sample of the alumina powder is subjected to an X-ray diffraction measurement, whereupon the integrated intensity of a peak in the vicinity of $2\theta=43^\circ$ is calculated, and the α -phase content is obtained from the calibration line.

25 In a second aspect of the present disclosure, there is provided a resin composition containing the alumina powder of the first aspect in a resin or rubber.

In a third aspect of the present disclosure, there is provided a sealing material for semiconductors, employing the resin composition of the second aspect.

In a fourth aspect of the present disclosure, there is provided a heat dissipator
30 employing the resin composition of the second aspect.

The present disclosure also provides the following:

(1) An alumina powder which has an α -phase content of at most 40% as measured by the following method, an average circularity of at least 0.95 in each of the particle size range of from 45 to 200 μm and the particle size range of less than 45 μm , and an average particle size of at most 100 μm :

5 [method for measuring α -phase content]

Powders prepared by mixing α -phase alumina powder and θ -phase alumina powder in mass ratios of 0:10, 5:5 and 10:0, respectively, are subjected to X-ray diffraction measurements, whereupon the integrated intensity of a peak of α -phase detected in the vicinity of $2\theta=43^\circ$ is calculated, and a calibration line of the integrated intensity plotted
10 against the mixing ratio is made; and then, a sample of the alumina powder is subjected to an X-ray diffraction measurement, whereupon the integrated intensity of a peak in the vicinity of $2\theta=43^\circ$ is calculated, and the α -phase content is obtained from the calibration line.

(2) A process for producing the alumina powder as defined in the above (1), which
15 comprises heat-treating a pulverized product of fused alumina in flames.

(3) The process for producing the alumina powder according to the above (2), which comprises quenching after the heat-treatment of a pulverized product of fused alumina.

(4) A resin composition containing the alumina powder as defined in the above (1) in a resin or rubber.

20 (5) The resin composition according to the above (4), wherein the resin is an epoxy resin.

(6) The resin composition according to the above (4), wherein the resin is a silicone

resin.

(7) The resin composition according to the above (4), wherein the rubber is a silicone rubber.

(8) A sealing material for semiconductors, employing the resin composition as defined
5 in the above (4) or (5).

(9) A heat dissipator employing the resin composition as defined in any one of the above (4) to (7).

ADVANTAGEOUS EFFECTS

According to the present disclosure, it is possible to provide an alumina powder
10 which has a high average circularity and which can maintain the flow property at a high level even at the time of being highly filled in a resin composition and thus is suitable as a filler, and a process for its production. The alumina powder of the present disclosure is useful as a sealing material for semiconductors, a heat dissipator, etc.

BRIEF DESCRIPTION OF THE DRAWING

15 Fig. 1 is a schematic view illustrating an embodiment of the process for producing the alumina powder of the present invention.

BEST MODE FOR CARRYING OUT THE INVENTION

Now, the present invention will be described in detail.

For the alumina powder of the present invention, a fused alumina pulverized
20 product is used as a raw material, whereby it is possible to reduce the surface cracking of alumina powder particles thereby to increase the flowability.

Further, by controlling the α -phase content in the alumina powder to be at most 40%, an improvement can be made also with respect to the surface irregularities of the particles, whereby the flowability can further be increased. The α -phase content
25 should better be close to 0, and it is more preferably at most 30%, particularly preferably from 20 to 0%.

The α -phase content in the alumina powder can be controlled by changing the cooling condition after the heat treatment of a pulverized product of fused alumina. Such cooling can be carried out by spraying water into the furnace at a middle body

portion of the furnace. The cooling condition in the furnace can be adjusted by the amount of water to be sprayed.

In the present invention, the amount of water to be sprayed for cooling is preferably at least 100 L per one hour.

5 The average particle size of the alumina powder of the present invention may be variously selected depending upon the particular purpose. According to the process of the present invention which will be described hereinafter, it is possible to easily produce an alumina powder having an average particle size of at most 100 μm , particularly from 10 to 95 μm . The average particle size can be increased or decreased by controlling
10 the average particle size of the raw material.

In the present invention, the average particle size of the alumina powder is adjusted to be at most 100 μm for such a reason that if the average particle size exceeds 100 μm , it becomes difficult to accomplish an average circularity of at least 0.95. In order to obtain a high flowability in a highly filled region, the average circularity
15 is preferably at least 0.95, and the average circularity is more preferably at least 0.97.

The average particle size can be measured, for example, by using a laser diffraction type particle size distribution measuring apparatus, CILAS GRANULOMETER "Model 920" (manufactured by BECKMAN COULTER). With respect to particles having an average particle size of less than 25 μm , 1 g of a sample, with respect to
20 particles having an average particle size of from 25 to less than 45 μm , 2 g of a sample, or with respect to particles having an average particle size of from 45 to less than 120 μm , 4 g of a sample, is weighed and introduced directly into a sample introduction section of the CILAS GRANULOMETER. The particle size distribution measurement by the CILAS GRANULOMETER was carried out at a pump rotational speed of 60 rpm
25 using water as a solvent and employing the refractive index of alumina (1.768) for setting.

The resin composition of the present invention is one having the alumina powder of the present invention incorporated and filled in a resin or rubber. In the present invention, one having the alumina powder of the present invention incorporated and
30 filled in a resin is referred to as "a resin composition", and one having the alumina powder of the present invention incorporated and filled in a rubber is referred to as "a rubber composition". Further, "the resin composition" in the specification of the present

invention may sometimes be used to cover the resin composition and the rubber composition.

In order to have the alumina powder highly filled in a resin or rubber, the average circularity of the alumina powder is preferably at least 0.95, particularly preferably at least 0.97. The average circularity of the alumina powder may be increased or decreased by changing the amount of a fuel gas (e.g. LPG) to be used for forming flames.

The average circularity can be measured, for example, by using a flow type particle image analyzer "FPIA-3000" manufactured by Sysmex. That is, 40 g of an alumina powder is weighed and put into a 200 ml beaker, and 100 ml of deionized water is introduced, followed by stirring and then by dispersion for 3 minutes by an ultrasonic cleaner (e.g. tradename "Ultrasonic Cleaner Powerful Type VS-150", manufactured by AS ONE Corporation). Into a JIS 45 μm sieve provided with a tray, the slurry in the beaker is introduced, and then 300 ml of deionized water is poured on the sieve, and then, depending upon the particle size, a measurement is carried out as follows. Here, for the average circularity, the perimeter of a projected image of one particle and the perimeter of a circle corresponding to the area of the projected image of the particle are analyzed by the above-mentioned flow-type particle image analyzer "FPIA-3000", the circularity is obtained by the following formula, and an average value per 36,000 particles is automatically calculated.

Circularity=Perimeter of a projected image of a particle/ Perimeter of a circle
corresponding to the area of the projected image of the particle
[Average circularity of particles having particle sizes of from 45 to 200 μm]

From 0.15 to 0.20 g of particles on the sieve are weighed and put into a 5 ml container, and 5 ml of a 25 mass% propylene glycol aqueous solution is added thereto, followed by dispersion for 10 seconds by an ultrasonic cleaner. The dispersion is put in its entire amount into a flow-type particle image analyzer "FPIA-3000" and measured in a HPF mode/quantitative count system (total number counted: 36,000 particles, number of repeated measurement: once), whereupon an analysis is carried out within a particle size range of from 45 to 200 μm (circle-equivalent diameter/based on the number of particles).

[Average circularity of particles having particle sizes of less than 45 μm]

The slurry settled in the above tray is stirred by a stirring rod, and 0.5 ml thereof is sampled into a 5 ml container, and 5 ml of a 25 mass% propylene glycol aqueous solution is added, followed by dispersion for 10 seconds by an ultrasonic cleaner. The dispersion is introduced in its entire amount into a flow-type particle image analyzer "FPIA-3000" and measured in a HPF mode/quantitative count system (total number counted: 36,000 particles, number of repeated measurement: once), whereupon an analysis is carried out within a particle size range of from 1.5 to less than 45 μm (circle-equivalent diameter/based on the number of particles).

The production of the alumina powder of the present invention is carried out preferably by using the installation shown in Fig. 1 by using a pulverized product of fused alumina as a raw material powder.

As outlined, from the top of a furnace, the pulverized product of a fused alumina is sprayed and melted in flames, and from a middle body portion of the furnace, water is sprayed into the furnace for quenching, whereupon the obtained spheroidized particles are transported together with an exhaust gas by a blower to a bag filter and thereby collected. Formation of flames is carried out by spraying a fuel gas such as hydrogen, natural gas, acetylene gas, propane gas or butane and a supporting gas such as air or oxygen from a burner set in the furnace. The temperature of the flames is adjusted to be preferably at least 2,100°C, particularly preferably from 2,100 to 2,300°C.

For the cooling by spraying water into the furnace, a twin-fluid nozzle may be used. In the present invention, it is preferred to carry out the quenching treatment by spraying water by means of Atomax nozzle BN160 Model (manufactured by Atomax Co., Ltd.).

In such quenching treatment by spraying water in the present invention, the temperature in the furnace is preferably lowered by from 200 to 400°C as compared with a case where no water is sprayed.

Such a range of lowering the temperature in the furnace can be attained preferably by the above-mentioned spray amount of water.

In the process for producing an alumina powder of the present invention, the α -phase content of the alumina powder can be adjusted by the spray amount of water (corresponding to setting of the temperature condition in the furnace).

The pulverized product of fused alumina as the raw material to be used in the process for producing an alumina powder of the present invention, is preferably a

pulverized product of a melt-solidified Bayer process alumina. For the melting of Bayer process alumina, an arc furnace may be used. The average particle size of the pulverized product of fused alumina is preferably at most 100 μm , more preferably from 10 to 95 μm .

5 In a case where the alumina powder in the present invention is to be used for a sealing material for semiconductors, it is necessary to reduce ionic impurities in the alumina powder. For the reduction of ionic impurities in the alumina powder, it is preferred to subject the flame-treated product of the pulverized product of fused alumina to washing treatment with water. For such washing treatment with water, a method
10 disclosed in JP-A-2005-281063 may, for example, be used. That is, the treatment is carried out by washing the alumina powder with water having a pH of from 3 to 7 which does not substantially contain Na^+ , K^+ , NH_4^+ , Mg_2^+ or Ca_2^+ , in the presence of an ion exchange resin.

As the resin for the resin composition of the present invention, it is possible to use,
15 for example, an epoxy resin, a phenol resin, a melamine resin, a urea resin, an unsaturated polyester, a fluororesin; a polyamide such as a polyimide, a polyamide imide or a polyether imide; a polyester such as a polybutylene terephthalate or a polyethylene terephthalate; a polyphenylene sulfide, an aromatic polyester, a polysulfone, a liquid crystal polymer, a polyether sulfone, a polycarbonate, a maleimide-
20 modified resin, an ABS resin, an AAS (acrylonitrile/acrylic rubber/styrene) resin, an AES (acrylonitrile/ethylene/propylene/diene rubber/styrene) resin, an EVA (ethylene/vinyl acetate copolymer) resin or a silicone resin. Among them, an epoxy resin, a phenol resin, a fluororesin, a polyimide, a polyphenylene sulfide, a polycarbonate, an ABS resin or a silicone resin is, for example, preferred.

25 Particularly preferred is an epoxy resin or a silicone resin.

The resin composition of the present invention is one having the alumina powder of the present invention incorporated and filled in the above-mentioned resin or rubber. The resin composition of the present invention can be produced by blending predetermined amounts of the respective materials by a blender or a Henschel mixer
30 and then kneaded by e.g. heated rollers, a kneader or a single or twin screw extruder, followed by cooling and then by pulverization. The content of the alumina powder varies depending upon the particular applications, but the present invention is

characterized in that the alumina powder can be highly filled in the resin or rubber, and its content is preferably made to be at least 40 vol%, more preferably from 65 to 90 vol%, based on 100 vol% of the resin composition.

One containing the alumina powder of the present invention in an epoxy resin is suitable as a sealing material for semiconductors. In order to obtain a high heat conductivity exceeding 3 W/m·K as a sealing material for semiconductors, the content of the alumina powder is adjusted to be preferably at least 70 vol%, more preferably from 75 to 90 vol%, based on 100 vol% of the resin composition.

One containing the alumina powder of the present invention in a silicone resin or silicone rubber, is suitable as a heat dissipator. In order to obtain a high heat conductivity as a heat dissipator, the content of the alumina powder is adjusted to be at least 65 vol%, more preferably from 70 to 80 vol%, based on 100 vol% of the resin composition.

EXAMPLES

Now, the present invention will be described in further detail with reference to Examples, but it should be understood that the present invention is by no means limited to such Examples.

Examples 1 to 9 and Comparative Examples 1 to 3 (production of alumina powder)

[Production of pulverized product of fused alumina]

Bayer process calcined alumina powder "D" (A11 manufactured by Light Metal Co., Ltd.) was melted by an arc furnace, followed by cooling and pulverization to obtain pulverized products of fused alumina "A", "B" and "C". The production of "A", "B" and "C" was differentiated by adjusting the pulverization time.

For the pulverization treatment at the time of preparing the raw material, a ball mill (AXB-15, manufactured by Seiwa Giken Co., Ltd.) was used, and as pulverization media, alumina balls (balls with a diameter of 30 mm) were used.

Table 1 shows the types and average particles sizes of the alumina powder raw materials "A", "B", "C" and "D" used in the present invention.

[Production of alumina powder]

[Heat treatment]

The heat treatment was carried out by using the production apparatus shown in Fig. 1. Flames of from 2,150 to 2,500°C were formed by adjusting the spray amounts

of the fuel gas (LPG) and the supporting gas (O_2 gas) to be as shown in Table 2. The alumina powder raw material (supply amount: 30 kg/hr) was accompanied with oxygen gas (supply amount: 20 Nm^3/hr) and sprayed from a nozzle into flames, and the obtained alumina powder was recovered from a bag filter. The temperature of the flames was confirmed by installing a burner outside the furnace and by using a radiation thermometer IS5/F Model manufactured by Impac.

[Quenching treatment]

In order to reduce the α -phase content, cooling treatment by spraying water into the furnace was carried out. For such water spraying, Atomax nozzle BN160 Model (manufactured by Atomax Co., Ltd.) was used. Spraying of water was carried out from 16 holes equally separated around the circumference of the furnace. The temperature of the water to be sprayed was adjusted to 10°C. The amount of water sprayed is shown in Table 2.

Further, with respect to the degree of quenching, the temperature of the furnace wall was measured by inserting an R thermocouple in a horizontal direction to the furnace wall surface in the same horizontal plane as the nozzles for water spraying, and the cooling effects were confirmed by using the furnace wall temperature as a reference. The measured furnace wall temperature is shown in Table 2.

[Water washing treatment]

The obtained alumina powder was subjected to water washing treatment. The water washing treatment was carried out in accordance with the following procedure. That is, for the water washing treatment, the alumina powder was mixed with deionized water of pH=7 wherein Li^+ , Na^+ and K^+ components were not detected by measurement by means of an atomic absorption spectrophotometer, to prepare an aqueous slurry having an alumina powder concentration of 40 mass%, which was stirred for 1 hour by means of a stirring and mixing apparatus (tradename "Stir Disperser RSV175" manufactured by Ashizawa Finetech Ltd.) and then subjected to dehydration treatment by a filter press. The water content of the obtained cake was at most 20 mass% in all cases. This cake was dried at 150°C for 48 hours by a shelf-type drying machine to obtain water washing-treated alumina powder.

The average circularity of the alumina powder was measured by the following method. The results are shown in Table 3.

[Average circularity]

The average circularity was measured, for example, by using a flow type particle image analyzer "FPIA-3000" manufactured by Sysmex. That is, 40 g of an alumina powder was weighed and put into a 200 ml beaker, and 100 ml of deionized water was introduced, followed by stirring and then by dispersion for 3 minutes by an ultrasonic cleaner (e.g. tradename "Ultrasonic Cleaner Powerful Type VS-150", manufactured by AS ONE Corporation). Then, into a JIS 45 μm sieve provided with a tray, the slurry in the beaker was introduced, and then 300 ml of deionized water was poured on the sieve, and then, depending upon the particle size, a measurement was carried out as follows. Here, for the average circularity, the perimeter of a projected image of one particle and the perimeter of a circle corresponding to the area of the projected image of the particle were analyzed by the above-mentioned flow-type particle image analyzer "FPIA-3000", the circularity was obtained by the following formula, and an average value per 36,000 particles was automatically calculated.

$$\text{Circularity} = (\text{Perimeter of a projected image of a particle}) / (\text{Perimeter of a circle corresponding to the area of the projected image of the particle})$$

[Average circularity of particles having particle sizes of from 45 to 200 μm]

From 0.15 to 0.20 g of particles on the sieve were weighed and put into a 5 ml container, and 5 ml of a 25 mass% propylene glycol aqueous solution was added thereto, followed by dispersion for 10 seconds by an ultrasonic cleaner. The dispersion was put in its entire amount into a flow-type particle image analyzer "FPIA-3000" and measured in a HPF mode/quantitative count system (total number counted: 36,000 particle, number of repeated measurement: once) whereupon an analysis was carried out within a particle size range of from 45 to 200 μm (circle-equivalent diameter/based on the number of particles).

[Average circularity of particles having particle sizes of less than 45 μm]

The slurry settled in the above tray was stirred by a stirring rod, and 0.5 ml thereof was sampled into a 5 ml container, and 5 ml of a 25 mass% propylene glycol aqueous solution was added, followed by dispersion for 10 seconds by a ultrasonic cleaner.

The dispersion was introduced in its entire amount into a flow-type particle image analyzer "FPIA-3000" and measured in a HPF mode/quantitative count system (total number counted: 36,000 particles, number of repeated measurement: once) whereupon

an analysis was carried out within a particle size range of from 1.5 to less than 45 μm (circle-equivalent diameter/based on the number of particles).

The average particle size was measured, for example, by using a laser diffraction type particle size distribution measuring apparatus, CILAS GRANULOMETER "Model 920" (manufactured by BECKMAN COULTER). With respect to particles having an average particle size of at most 25 μm , 1 g of a sample, with respect to particles having an average particle size of from 25 to 45 μm , 2 g of a sample, or with respect to particles having an average particle size of from 45 to 120 μm , 4 g of a sample, was weighed and introduced directly into a sample introduction section of the CILAS GRANULOMETER. The particle size distribution measurement by the CILAS GRANULOMETER was carried out at a pump rotational speed of 60 rpm using water as a solvent and employing the refractive index of alumina (1.768) for setting.

The α -phase content was measured by the following method. The results are shown in Table 3.

The α -phase content can be obtained by a calibration line method of X-ray diffraction. For the measurement of the X-ray diffraction, JDX-3500 Model X-ray diffraction apparatus (manufactured by JEOL Ltd.) was used.

[Method for measuring α -phase content]

Powders prepared by mixing α -phase alumina powder AA-05 (manufactured by Sumitomo Chemical Co., Ltd.) and θ -phase alumina powder TAIMICRON TM-100D (manufactured by TAIMEI CHEMICALS Co., Ltd.) in mass ratios of 0:10, 5:5 and 10:0, respectively, were subjected to X-ray diffraction measurements, whereupon the integrated intensity of a peak of α -phase detected in the vicinity of $2\theta=43^\circ$ was calculated, and a calibration line of the integrated intensity plotted against the mixing ratio was made.

Then, a sample of the alumina powder was subjected to an X-ray diffraction measurement, whereupon the integrated intensity of a peak in the vicinity of $2\theta=43^\circ$ was calculated, and the α -phase content was obtained from the calibration line.

EXAMPLES 11 to 19 and COMPARATIVE EXAMPLES 11 to 13

In Examples 11 to 18 and Comparative Examples 11 to 13 in Table 3, 30 parts by volume of the resin blend shown in Table 5 and 70 parts by volume of the alumina powder obtained in one of the above Examples 1 to 8 and Comparative Examples 1 to 3,

were mixed to prepare an epoxy resin composition. With respect to the respective resin compositions of such Examples and Comparative Examples, the flowability and the heat conductivity were evaluated as follows.

Further, in Example 19, 20 parts by volume of the resin blend shown in Table 5
5 and 80 parts by volume of the alumina powder of Example 9 were mixed to prepare an epoxy resin composition, and its flowability and heat conductivity were evaluated as follows.

Here, preparation of a heat dissipator used in the measurement of the heat conductivity was carried out as follows. A material for semiconductors, prepared by
10 heating and kneading by a twin screw extrusion kneader was cast into a plate-forming mold of 25 mm × 25 mm × 3 mm by means of a transfer molding machine to prepare a sample for evaluation of the heat conductivity.

[Flowability]

Using a spiral flow mold, a spiral flow value of the sealing material for
15 semiconductors, prepared by heating and kneading by the twin screw extrusion kneader, was measured by means of a transfer molding machine provided with a spiral flow measuring mold in accordance with EMMI-66 (Epoxy Molding Material Institute: Society of Plastic Industry). The transfer molding conditions were such that the mold temperature was 175°C, the molding pressure was 7.4 MPa (gauge pressure), and the
20 pressure holding time was 90 seconds.

EXAMPLES 21 to 29 and COMPARATIVE EXAMPLES 21 to 23

[Viscosity evaluation]

In Examples 21 to 28 and Comparative Examples 21 to 23 in Table 4, 40 parts by volume of the liquid silicone rubber and the 70 parts by volume of the alumina powder
25 obtained in one of the above Examples 1 to 8 and Comparative Examples 1 to 3, were mixed to prepare a silicone rubber composition. With respect to the respective silicone rubber compositions of such Examples and Comparative Examples, the viscosity and the heat conductivity were evaluated as follows.

Further, in Example 29, 30 parts by volume of the liquid silicone rubber and 70
30 parts by volume of the alumina powder obtained in Example 9 were mixed to prepare a silicone rubber composition, and its viscosity and heat conductivity were evaluated as follows. Further, as the liquid silicone rubber, YE5822A (manufactured by Momentive

Material) was used.

The obtained results are shown together with the production conditions for the alumina powders (Examples 1 to 9 and Comparative Examples 1 to 3) in Table 4.

[Viscosity measurement]

5 For the viscosity measurement, heat-treated alumina powder was introduced into the liquid silicone rubber YE5822A manufactured by Momentive Material, followed by mixing by means of a stirring machine NZ-1100, manufactured by TOKYO RIKAKIKAI Co., Ltd.. The viscosity of the mixed silicone rubber composition was measured, after vacuum deaeration, by B Model viscosity TVB-10, manufactured by TOKI SANGYO CO., LTD. For the viscosity measurement, No. 7 spindle was used, and the measurement was carried out at a rotational speed of 20 rpm at a room temperature of 20°C.

Further, preparation of the heat dissipator using the silicone rubber composition was carried out as follows.

That is, to the silicone rubber composition comprising the liquid silicone rubber YE5822A and the alumina powder, prepared in the above viscosity measurement, liquid silicone rubber YE5822B was added in a ratio of 10:1 by mass, to YE5822A (i.e. YE5822A:YE5822B=10:1). After the addition of YE5822B, the mixture was molded and heat-treated in an atmosphere of 120°C to prepare a heat dissipator.

[Heat conductivity measurement]

20 The heat dissipator of the epoxy resin composition or the silicone rubber composition prepared as described above was molded into a size of 25 mm × 25 mm (length × width) and a thickness of 3 mm, and the molded member was sandwiched between a 15 mm × 15 mm copper heater case and a copper plate and set under a clamping torque of 5 kgf/cm. Then, a power of 15 W was applied to the copper heater case and maintained for 4 minutes, whereupon the temperature difference between the copper heater case and the copper plate was measured, and the thermal resistance was calculated by the following formula.

Thermal resistance (°C/W)=

Temperature difference (°C) between copper heater case and copper plate/

30 Power (W) applied to heater

The heat conductivity can be calculated by the following formula from the thermal resistance (°C/W), the heat transfer area [area of copper heater case] (m²) and the

thickness (m) of the molded product under a clamping torque of 5 kgf/cm.

Heat conductivity (W/m·k)=Thickness (m) of molded product/

{Thermal resistance (°C/W) × heat transfer area (m²)}

Tables 3 and 4 show the results of measurement of the physical properties of the resin compositions and the rubber compositions in the respective Examples and Comparative Examples.

Further, "filler filling rate" in Tables 3 and 4 is meant for the filling rate (vol%) of the alumina powder obtained in each Example or Comparative Example.

TABLE 1

Symbol for alumina powder raw material	Type of raw material	Average particle size (μm)
A	Pulverized product of fused alumina	46.4
B	Pulverized product of fused alumina	95.6
C	Pulverized product of fused alumina	124.3
D	Calcined alumina powder	48.9

10

TABLE 2

	Symbol for alumina powder raw material	Amount of LPG (Nm ³ /hr)	Amount of supporting O ₂ (Nm ³ /hr)	Amount of water sprayed (L/(hr·nozzle))	Temperature of flames (°C)	Temperature of furnace wall (°C)
Ex. 1	A	25	125	0	2150	1046
Ex. 2	A	25	125	5	2150	983
Ex. 3	A	25	125	15	2150	921
Ex. 4	A	25	125	20	2150	853
Ex. 5	B	40	200	0	2300	1351
Ex. 6	B	40	200	5	2300	1236
Ex. 7	B	40	200	15	2300	1125
Ex. 8	B	40	200	20	2300	1038
Ex. 9	A	25	125	20	2150	853
Comp. Ex. 1	C	50	250	0	2400	1390
Comp. Ex. 2	C	60	300	0	2500	1455
Comp. Ex. 3	D	25	125	0	2150	1026

TABLE 3

	Alumina powder used	Average particle size (μm)	Circularity		α -phase content (%)	Filler filling rate (vol%)	Spiral flow (cm)	Heat conductivity ($\text{W/m}\cdot\text{K}$)
			Particles of 45-200 μm	Particles of less than 45 μm				
Ex. 11	Ex. 1	49.2	0.99	0.99	30.5	70	104.8	4.2
Ex. 12	Ex. 2	48.4	0.98	0.99	25.6	70	110.3	4.1
Ex. 13	Ex. 3	47.9	0.98	0.99	21.4	70	116.8	4.1
Ex. 14	Ex. 4	48.8	0.98	0.99	17.5	70	123.1	4
Ex. 15	Ex. 5	98.7	0.97	0.99	38.2	70	113.8	4.4
Ex. 16	Ex. 6	97.1	0.97	0.98	34.6	70	119.6	4.3
Ex. 17	Ex. 7	97.3	0.96	0.97	31.4	70	117.1	4.2
Ex. 18	Ex. 8	96.6	0.96	0.97	28.9	70	118.8	4.2
Ex. 19	Ex. 9	48.8	0.98	0.99	17.5	80	85.6	5.3
Comp. Ex. 11	Comp. Ex. 1	127.9	0.89	0.99	51.6	70	51.8	4.1
Comp. Ex. 12	Comp. Ex. 2	129.6	0.93	0.99	42.8	70	83.7	4.1
Comp. Ex. 13	Comp. Ex. 3	52.1	0.97	0.98	45.9	70	84.3	4.1

TABLE 4

	Alumina powder used	Amount of LPG (Nm ³ /hr)	Amount of supporting O ₂ (Nm ³ /hr)	Amount of water sprayed (L/(hr·nozzle))	Filler filling rate (vol%)	Viscosity (cP)	Heat conductivity (W/m·K)
Ex. 21	Ex. 1	25	125	0	60	106590	3.4
Ex. 22	Ex. 2	25	125	5	60	97410	3.3
Ex. 23	Ex. 3	25	125	15	60	92820	3.3
Ex. 24	Ex. 4	25	125	20	60	88740	3.2
Ex. 25	Ex. 5	40	200	0	60	90984	3.5
Ex. 26	Ex. 6	40	200	5	60	84456	3.4
Ex. 27	Ex. 7	40	200	15	60	80376	3.4
Ex. 28	Ex. 8	40	200	20	60	75888	3.4
Ex. 29	Ex. 9	25	125	20	70	144420	4.3
Comp. Ex. 21	Comp. Ex. 1	50	250	0	60	159630	3.3
Comp. Ex. 22	Comp. Ex. 2	60	300	0	60	136170	3.3
Comp. Ex. 23	Comp. Ex. 3	25	125	0	60	191250	3.3

TABLE 5

Type of material	Variety	Blend (mass%)
Epoxy resin	Orthocresol novolac type ("EOCN-1020" manufactured by Nippon Kayaku Co., Ltd.)	63.8
Curing agent	Phenol novolac resin ("PMS-4261" manufactured by Gun Ei Chemical Industry Co., Ltd.	32.1
Curing accelerator	Triphenylphosphine (manufactured by HOKKO CHEMICAL INDUSTRY CO., LTD.	0.6
Releasing agent	Montanoic acid ester ("WaxEflakes" manufactured by Clariant Japan)	3.5
Silane coupling agent	Organosilane ("KBM-403" manufactured by Shin-Etsu Chemical Co., Ltd.	0.5 Part by mass to 100 parts by mass of alumina powder

As is evident from Tables 3 and 4, the resin compositions using the alumina powders of the present invention have low viscosities and high spiral flow values, and thus, the flowability is remarkably improved.

The resin compositions using the alumina powders of the present invention are suitable for application to a sealing material for semiconductors or a heat dissipator.

INDUSTRIAL APPLICABILITY

The alumina powder of the present invention is useful as a filler for a resin composition and further useful for a molding compound or a heat dissipating sheet for e.g. automobiles, portable electronic equipments, industrial equipments or household electric appliances.

Further, the sealing material for semiconductors obtainable from the resin composition of the present invention is useful as a component of which the heat dissipation property is important, such as graphic chips.

The entire disclosure of Japanese Patent Application No. 2008-118246 filed on April 30, 2008 including specification, claims, drawings and summary is incorporated herein by reference in its entirety.

MEANING OF SYMBOLS

1: Melting furnace

- 2: Burner
- 3: Fuel gas supply pipe
- 4: Supporting gas supply pipe
- 5: Raw material powder supply pipe
- 5 6: Water supply pipe
- 7: Bag filter
- 8: Blower
- 9: R thermocouple

CLAIMS:

1. An alumina powder which contains an α -phase and a θ -phase as main crystals, and has an α -phase content of at most 40% as measured by the following method, an average circularity of at least 0.95 in each of the particle size range of from 45 to 200 μm and the particle size range of less than 45 μm , and an average particle size of at most 100 μm :

[method for measuring α -phase content]

Powders prepared by mixing α -phase alumina powder and θ -phase alumina powder in mass ratios of 0:10, 5:5 and 10:0, respectively, are subjected to X-ray diffraction measurements, whereupon the integrated intensity of a peak of α -phase detected in the vicinity of $2\theta=43^\circ$ is calculated, and a calibration line of the integrated intensity plotted against the mixing ratio is made; and then, a sample of the alumina powder is subjected to an X-ray diffraction measurement, whereupon the integrated intensity of a peak in the vicinity of $2\theta=43^\circ$ is calculated, and the α -phase content is obtained from the calibration line.

2. The alumina powder according to Claim 1, wherein the α -phase content is at most 30%.

3. The alumina powder according to Claim 1 or 2, wherein the α -phase content is at most 20%.

4. A resin composition containing the alumina powder as defined in Claim 1 in a resin or rubber.

5. The resin composition according to Claim 4, wherein the resin is an epoxy resin or a silicone resin.

6. The resin composition according to Claim 4, wherein the rubber is a silicone rubber.

7. A sealing material for semiconductors, employing the resin composition as defined in Claim 4 or 5.

8. A heat dissipator employing the resin composition as defined in any one of Claims 4 to 6.

ALUMINA POWDER, PROCESS FOR PRODUCTION OF THE SAME, AND RESIN
COMPOSITIONS CONTAINING THE SAME

ABSTRACT:

5 To provide an alumina powder having an improved flowability, a process for its
production, and a resin composition employing it.

 An alumina powder which has an α -phase content of at most 40% as measured
by the following method, an average circularity of at least 0.95 in each of the particle
size range of from 45 to 200 μm and the particle size range of less than 45 μm , and an
10 average particle size of at most 100 μm :

[method for measuring α -phase content]

Powders prepared by mixing α -phase alumina powder and θ -phase alumina powder in
mass ratios of 0:10, 5:5 and 10:0, respectively, are subjected to X-ray diffraction
measurements, whereupon the integrated intensity of a peak of α -phase detected in the
15 vicinity of $2\theta=43^\circ$ is calculated, and a calibration line of the integrated intensity plotted
against the mixing ratio is made; and then, a sample of the alumina powder is subjected
to an X-ray diffraction measurement, whereupon the integrated intensity of a peak in the
vicinity of $2\theta=43^\circ$ is calculated, and the α -phase content is obtained from the calibration
line.

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[Figure 1]