

DESCRIPTION

Title of Invention:

AGGREGATE OF FLAKED SILVER PARTICLES AND PASTE CONTAINING
AGGREGATE OF SILVER PARTICLES

Technical Field

[0001]

The present invention relates to an aggregate of flaked silver particles having high light reflectance (hereafter, the aggregate of silver particles is also referred to as a "silver powder"). The silver powder is uniformly dispersed in a resin to form a paste having high reflectance and high glossiness and also having high conductivity. Furthermore, the paste does not readily segregate because of its small silver sedimentation ratio, which makes the viscosity of the paste uniform and achieves high working efficiency of the paste.

Background Art

[0002]

At present, light-emitting diodes (hereafter referred to as "LEDs") are used for many applications such as liquid crystal displays, traffic signals, lighting, and display devices. Since LEDs are energy saving and low cost and thus environmentally friendly, LEDs have promise for expansion into other fields, for example, automobile headlamps.

[0003]

Normally, for example, liquid crystal displays include LED chips fixed using an adhesive having conductivity and a reflecting film for efficiently emitting light from the LED chips to the outside. To more efficiently emit light, such an adhesive for fixing chips desirably has high reflectance.

[0004]

The reflectance of the adhesive is highly affected by the characteristics of a powder dispersed as a conductive filler in the adhesive.

In general, a silver powder is used as a conductive filler for adhesives for fixing LED chips. Therefore, the reflectance of an adhesive increases as the reflectance of a silver powder dispersed increases.

[0005]

The reflectance of the silver powder is higher in an aggregate of flaked silver particles than in an aggregate of spherical particles. The reflectance is also higher in an aggregate of silver particles having a large particle size than in an aggregate of silver particles having a small particle size.

[0006]

However, when a flaked silver powder having a large particle size is dispersed in a resin serving as a raw material for adhesives to increase the reflectance, the silver powder readily settles, which easily causes

segregation.

The segregation reduces the flowability of a portion having a high silver concentration, which makes the viscosity of the paste nonuniform. When the paste is applied using a nozzle, the amount of the paste ejected varies and the nozzle is easily clogged, which deteriorates the working efficiency.

Furthermore, small-diameter nozzles used for fine coating cannot be used because of clogging.

[0007]

A flaked silver powder constituted by fine particles having a small particle size does not readily segregate. However, a desired reflectance is not achieved because such a flaked silver powder originally has low reflectance and irregular reflection of light further decreases the reflectance.

[0008]

Thus, there has been demanded the development of a silver powder that can provide an adhesive which has sufficient reflectance and conductivity, can be suitably used for fixing LED chips, does not readily segregate, and can be used with high working efficiency.

Citation List

Patent Literature

[0009]

PTL 1: Japanese Unexamined Patent Application

Publication No. 2005-15647

Summary of Invention

Technical Problem

[0010]

PTL 1 discloses a paint containing plate-shaped metal fine particles having a thickness of 50 nm or less and a major axis of 5 μm or less.

[0011]

However, the metal powder contained in the paint disclosed in PTL 1 is constituted by very thin plate-shaped metal fine particles. Therefore, such metal fine particles originally have low reflectance and irregular reflection further decreases the reflectance. To achieve high reflectance, a metal thin film needs to be formed by fusing the metal fine particles through heating.

[0012]

The present inventors have set up their technical object as solving the above problems and have made many prototypes and conducted many experiments by trial and error. Consequently, the present inventors have found the following notable finding. In the case where an aggregate of flaked silver particles having a BET specific surface area of 0.5 to less than 1.5 m^2/g and having, according to laser diffractometry, a 50% particle size of 1 to 4 μm and a 75%

particle size/25% particle size ratio of 1.8 or less is used, even if the 50% particle size is small, the content of fine particles is low and thus irregular reflection is suppressed, which increases the reflectance. Therefore, when such an aggregate of flaked silver particles is contained in a resin, a paste having high reflectance and high conductivity is provided. Furthermore, since the silver powder does not readily settle and thus segregation does not easily occur, an adhesive with high working efficiency is provided. Accordingly, the technical object has been achieved.

Solution to Problem

[0013]

The technical object can be achieved by the present invention as follows.

[0014]

The present invention provides an aggregate of flaked silver particles, the aggregate having a BET specific surface area of 0.5 to less than 1.5 m²/g and having, according to laser diffractometry, a 50% particle size of 1 to 4 μm and a 75% particle size/25% particle size ratio of 1.8 or less.

[0015]

The present invention also provides the aggregate of flaked silver particles according to Claim 1, wherein a reflectance of light having a wavelength of 400 to 800 nm is

40% or more.

[0016]

The present invention also provides a paste containing the aggregate of flaked silver particles according to Claim 1 or 2.

[0017]

The present invention also provides the paste according to Claim 3, wherein the paste contains the aggregate of flaked silver particles in an amount of 50 to 90 wt%.

[0018]

The present invention also provides the paste according to Claim 3 or 4, wherein the paste has a resistivity of 10^{-4} Ω cm or less.

[0019]

The present invention also provides the paste according to any one of Claims 3 to 5, wherein the paste has a silver sedimentation ratio of 0.5% or less.

[0020]

The present invention also provides the paste according to any one of Claims 3 to 6, wherein the paste is an adhesive for fixing a light-emitting diode (LED) chip.

[0021]

The present invention also provides a method for producing the aggregate of flaked silver particles according to Claim 1 or 2.

[0022]

The present invention also provides a method for producing the paste according to any one of Claims 3 to 7.

Advantageous Effects of Invention

[0023]

The silver powder according to the present invention has a small 50% particle size measured by laser diffraction, but has a narrow particle size distribution and thus the percentage of fine particles is low. Therefore, irregular reflection of light due to fine particles is suppressed, and a silver powder having high reflectance is provided.

In particular, a silver powder having a reflectance of 40% or more for light having a wavelength of 400 to 800 nm is provided.

[0024]

The paste prepared by dispersing the silver powder according to the present invention in a resin has high reflectance and high glossiness because the silver powder is uniformly dispersed in a resin.

Furthermore, the silver powder does not readily settle and thus segregation does not easily occur. Therefore, the viscosity of the paste is uniform and ejection failure does not readily occur, which provides a paste having high working efficiency.

[0025]

Since the silver powder according to the present invention is an aggregate of flaked silver particles, the silver particles are in contact with each other in a large area in the paste prepared by dispersing the silver powder in a resin. This provides a paste having high conductivity.

The paste containing the silver powder according to the present invention can have a resistivity of $1 \times 10^{-4} \Omega\text{cm}$ or less.

[0026]

The paste according to the present invention has high reflectance and high glossiness and also has high conductivity. Therefore, the paste can be suitably used as an adhesive for fixing LED chips.

[0027]

The paste according to the present invention has uniform viscosity and the percentage of silver particles having a large particle size is low. Therefore, a nozzle is not easily clogged with the paste and ejection failure does not readily occur. This provides a paste having high working efficiency.

The paste according to the present invention can also be used for small-diameter nozzles for fine coating.

Brief Description of Drawings

[0028]

[Fig. 1] Fig. 1 is a graph illustrating the comparison

of the reflectances of silver powders in Examples and Comparative Examples in the present invention.

[Fig. 2] Fig. 2 is a graph illustrating the comparison of the reflectances of pastes containing silver powders in Examples and Comparative Examples in the present invention.

[Fig. 3] Fig. 3 is a micrograph of a scanning electron microscope (SEM, JSM-6010LA manufactured by JEOL Ltd.) illustrating a silver powder magnified by 5000 times in the present invention.

Description of Embodiments

[0029]

A silver powder according to the present invention has a BET specific surface area of 0.5 to less than 1.5 m²/g. The BET specific surface area is more preferably 0.8 to 1.2 m²/g. If the specific surface area is less than 0.5 m²/g, the number of particles with a large particle size unfavorably increases, which increases the sedimentation ratio and thus facilitates the segregation. If the specific surface area is 1.5 m²/g or more, the number of fine particles unfavorably increases, which decreases the reflectance.

[0030]

The silver particles according to the present invention preferably have a thickness of 180 to 220 nm and an aspect ratio (major axis/thickness) of 7 to 13.

[0031]

Herein, when the silver particles having a specific surface area of 0.5 to less than 1.5 m²/g are calculated as a disc-shaped uniform particle model, the silver particles have a thickness of 140 to less than 620 nm.

[0032]

The silver powder according to the present invention has, according to laser diffractometry, a 50% particle size of 1 to 4 μm and a 75% particle size/25% particle size ratio of 1.8 or less.

[0033]

If the 50% particle size is less than 1 μm, the number of fine particles unfavorably increases and thus the reflectance decreases. If the 50% particle size is more than 4 μm, the weight per particle unfavorably increases. In the form of paste, the sedimentation ratio increases and thus the segregation is facilitated, which makes the viscosity nonuniform. Consequently, a nozzle is easily clogged with the paste, which causes ejection failure.

[0034]

If the 75% particle size/25% particle size ratio is more than 1.8, the percentage of fine particles and the percentage of silver particles with a large particle size unfavorably increase.

[0035]

The silver powder according to the present invention can

be produced by a method described in Japanese Unexamined Patent Application Publication No. 2003-55701. Specifically, a particulate silver powder having an average particle size of 0.5 to 3 μm is inserted into a ball mill equipped with an impeller. By rotating the impeller, a flaked silver powder can be produced.

[0036]

The centrifuge force exerted on the contents in a container of the mixer ball mill is not particularly limited. The impeller may be rotated so that a centrifuge force of 5 to 300 G is exerted on the contents in the container.

[0037]

Furthermore, well-known metal balls may be charged into the mixer ball mill.

[0038]

The particulate silver powder serving as a raw material is not particularly limited. A particulate silver powder prepared by a well-known method such as an atomizing method, an electrolysis method, or a chemical reduction method can be used.

[0039]

Various solvents and treating agents may be added to the mixer ball mill for the purpose of controlling, for example, the particle size.

[0040]

Non-limiting examples of the solvents added include water, methanol, ethanol, propanol, butanol, pentanol, dimethyl ketone, diethyl ketone, diethyl ether, dimethyl ether, diphenyl ether, toluene, and xylene. These solvents may be used alone or in combination.

[0041]

Non-limiting examples of the treating agents added include nonionic surfactants such as polyoxyethylene alkyl ethers, polyoxyethylene alkyl phenyl ethers, polyoxyethylene fatty acid esters, polyoxyethylene sorbitan fatty acid esters, and sorbitan fatty acid esters.

These surfactants may be used alone or in combination of two or more.

[0042]

Other examples of the treating agents include fatty acids such as oleic acid, stearic acid, and myristic acid. These fatty acids may be used alone or in combination of two or more.

[0043]

When the treating agents are used, the treating agents are preferably added to the mixer ball mill in an amount of 0.001 to 0.05 g in total per 1 m² of the surface area of a flaked silver powder to be obtained.

[0044]

A resin in which the silver powder according to the

present invention is dispersed is not particularly limited as long as the resin does not prevent emission of reflected light from the silver powder. The resin is suitably a transparent resin.

Examples of the resin to which the silver powder according to the present invention is added include silicon resin, epoxy resin, acrylic resin, polyester resin, and melamine resin.

[0045]

The content of the silver powder in the paste according to the present invention is not particularly limited, but is preferably 50 to 90 wt% and more preferably 70 to 85 wt%.

A silver powder content of 50 wt% or less is not preferred because sufficient reflectance and conductivity cannot be achieved. A silver powder content of 90% or more is also not preferred because increases in reflectance and conductivity are not expected and nozzles are easily clogged.

EXAMPLES

[0046]

Examples of the present invention will be described below, but the present invention is not limited thereto.

[0047]

<Example 1>

An appropriate amount of zirconia beads having a diameter of 0.05 mm were charged into a wet-type media

grinding machine including a cylindrical container having a diameter of 107 mm and a height of 320 mm. Then, the peripheral speed of a mixing rotor was set to 7 m/s, and a slurry solution prepared by mixing 700 g of a spherical silver powder having an average particle size of 1.5 μm and 6 L of ethanol was circulated for 1.5 hours.

Subsequently, the contents in the cylindrical container were filtered and ethanol was removed from the resulting filtrate. The resulting product was dried to obtain a flaked silver powder.

[0048]

<Example 2>

A flaked silver powder was obtained in the same manner as in Example 1, except that a spherical silver powder having an average particle size of 2.5 μm was used.

Fig. 3 is a micrograph of a scanning electron microscope (SEM) illustrating the flaked silver powder in Example 2 magnified by 5000 times.

[0049]

<Comparative Examples 1 to 7>

Into a ball mill including a cylindrical container having a diameter of 150 mm and a height of 190 mm, 700 g of a particulate silver powder prepared by a chemical reduction method and having an average particle size of 0.1 to 3 μm , 1 L of ethanol, and an appropriate amount of well-known metal

balls having a diameter of 1 to 2 mm were charged. The ball mill was operated at a container rotation speed of 40 to 80 rpm for 5 to 20 hours and then the rotation was stopped. Subsequently, the contents in the cylindrical container were filtered and ethanol was removed from the resulting filtrate. The resulting product was dried to obtain a flaked silver powder in each of Comparative Examples 1 to 7.

[0050]

<Comparative Example 8>

A spherical silver powder prepared by an atomizing method and having an average particle size of 1.5 μm was used for measurement without performing flaking.

[0051]

Each of the silver powders in Examples and Comparative Examples was evaluated by the following methods.

[0052]

(BET specific surface area)

The BET specific surface area of each silver powder was measured with a flow-type specific surface area automatic analyzer FlowSorb II 2100 (manufactured by SHIMADZU CORPORATION).

[0053]

(Particle size)

The 25% particle size, 50% particle size, and 75% particle size of each silver powder was measured with a

laser diffraction particle size analyzer SALD-3100
(manufactured by SHIMADZU CORPORATION).

[0054]

(Reflectance: silver powder)

The reflectance of each silver powder was measured with a spectrophotometric color difference meter SE6000 (manufactured by NIPPON DENSHOKU INDUSTRIES CO., LTD.) at a measurement wavelength of 550 nm.

The reflectances of each silver powder at 380 to 780 nm were also measured with a spectrophotometric color difference meter SE6000 (Fig. 1).

[0055]

(Conductivity)

A paste prepared by mixing 87 wt% of the silver powder into an epoxy resin was applied onto a glass substrate and then treated at 200°C for 30 minutes to obtain a coating film. The resistivity (ρ) was calculated from the resistance (R) and cross-sectional area (S) of the coating film.

$$\rho = R \cdot S / L$$

(R: resistance (Ω), S: cross-sectional area (cm^2), L: distance between electrodes (cm))

[0056]

(Silver sedimentation ratio)

A 10 cc syringe was filled with the same paste as that used for evaluating the conductivity and left in an upright

position at 25°C for 3 days. The difference in silver content between 1 cc of the uppermost portion and 1 cc of the lowermost portion was measured. The silver content was calculated from an ignition loss obtained by treating the paste at 800°C for 60 minutes.

[0057]

(Reflectance: paste)

A paste prepared by mixing 87 wt% of the silver powder in each of Example 1 and Comparative Examples 1, 7, and 8 into an epoxy resin was applied onto paper using a bar coater and then treated at 200°C for 30 minutes to obtain a coating film. The reflectance of the coating film at a measurement wavelength of 550 nm was measured with a spectrophotometric color difference meter SE6000 (manufactured by NIPPON DENSHOKU INDUSTRIES CO., LTD.).

The reflectances of the paste containing the silver powder at 380 to 780 nm were also measured with a spectrophotometric color difference meter SE6000 (Fig. 2).

[0058]

(Glossiness)

A paste prepared by mixing 87 wt% of the silver powder in each of Example 1 and Comparative Examples 1, 7, and 8 into an epoxy resin was applied onto paper using a bar coater and then treated at 200°C for 30 minutes to obtain a coating film. The glossiness of the coating film at a

reflection angle of 60° was measured with a gloss meter GM-268 Plus (manufactured by KONICA MINOLTA, INC.).

[0059]

Table 1 shows the evaluation values in Examples and Comparative Examples.

[0060]

[Table 1]

	Shape	Specific surface area (m ² /g)	50% particle size (μm)	25% particle size (μm)	75% particle size (μm)	75%/25% ratio	Reflectance (silver powder) (%)	Conductivity (Ω·cm)	Silver sedimentation ratio (paste) (%)	Reflectance (paste) (%)	Glossiness (paste)
Example 1	Flake	1.06	2.08	1.65	2.60	1.58	45.3	2.5×10 ⁻⁵	0.14	50.5	23.5
Example 2	Flake	1.11	2.33	1.86	2.90	1.56	43.8	3.0×10 ⁻⁵	0.23	-	-
Comparative Example 1	Flake	0.25	6.81	3.93	10.72	2.73	52.1	3.0×10 ⁻⁵	1.54	58.0	4.4
Comparative Example 2	Flake	0.27	6.88	4.10	10.47	2.55	49.8	2.6×10 ⁻⁵	0.75	-	-
Comparative Example 3	Flake	0.31	8.45	5.23	12.72	2.43	49.8	3.5×10 ⁻⁵	0.98	-	-
Comparative Example 4	Flake	0.38	5.53	2.89	9.17	3.17	38.8	5.5×10 ⁻⁵	0.69	-	-
Comparative Example 5	Flake	0.76	2.60	1.49	4.37	2.93	39.8	5.9×10 ⁻⁴	0.21	-	-
Comparative Example 6	Flake	0.91	3.57	2.07	6.24	3.01	38.8	2.3×10 ⁻⁵	0.44	-	-
Comparative Example 7	Flake	1.05	2.71	1.47	4.74	3.22	29.1	4.8×10 ⁻⁵	0.16	36.7	2.1
Comparative Example 8	Sphere	0.65	1.71	1.34	2.14	1.60	29.8	4.1×10 ⁻³	0.75	34.1	0.9

[0061]

Fig. 1 illustrates the reflectances of each silver powder at 380 to 780 nm.

Fig. 2 illustrates the reflectances of the paste containing the silver powder in each of Example 1 and Comparative Examples 1, 7, and 8 at 380 to 780 nm.

[0062]

It was confirmed from Table 1 and Fig. 1 that the silver powder according to the present invention was a silver powder having a small 50% particle size by laser diffraction of 1 to 4 μm , but having high reflectance.

[0063]

It was confirmed from Table 1 and Fig. 2 that the paste containing the silver powder according to the present invention was a paste having high reflectance, high glossiness, a small silver sedimentation ratio, and high conductivity.

It was also confirmed that the glossiness of the paste according to the present invention was higher than that of the pastes in Comparative Examples to a degree that the glossiness could be distinguished through visual inspection.

Industrial Applicability

[0064]

The silver powder according to the present invention has a small 50% particle size measured by laser diffraction, but

has a narrow particle size distribution and thus has high reflectance. The paste containing the silver powder according to the present invention has low irregular reflection and thus has high reflectance and high glossiness, and also has high conductivity. Furthermore, the paste has uniform viscosity because the segregation is suppressed, and thus has high working efficiency. Therefore, the paste can be suitably used as an adhesive for LED chips.

Accordingly, the present invention is highly industrially applicable.

CLAIMS

[Claim 1]

An aggregate of flaked silver particles, the aggregate having a BET specific surface area of 0.5 to less than 1.5 m²/g and having, according to laser diffractometry, a 50% particle size of 1 to 4 μm and a 75% particle size/25% particle size ratio of 1.8 or less.

[Claim 2]

The aggregate of flaked silver particles according to Claim 1, wherein a reflectance of light having a wavelength of 400 to 800 nm is 40% or more.

[Claim 3]

A paste comprising the aggregate of flaked silver particles according to Claim 1 or 2.

[Claim 4]

The paste according to Claim 3, wherein the paste contains the aggregate of flaked silver particles in an amount of 50 to 90 wt%.

[Claim 5]

The paste according to Claim 3 or 4, wherein the paste has a resistivity of 10⁻⁴ Ωcm or less.

[Claim 6]

The paste according to any one of Claims 3 to 5, wherein the paste has a silver sedimentation ratio of 0.5% or less.

[Claim 7]

The paste according to any one of Claims 3 to 6, wherein the paste is an adhesive for fixing a light-emitting diode (LED) chip.

[Claim 8]

A method for producing the aggregate of flaked silver particles according to Claim 1 or 2.

[Claim 9]

A method for producing the paste according to any one of Claims 3 to 7.