

DESCRIPTION

Title of Invention

METHOD FOR PRODUCING GRANULES, AND GRANULE

5 Technical Field

[0001] The present invention relates to a method for producing granules and to a granule.

Background Art

10 [0002] In the production of food products, pharmaceutical products, and the like, it is very important to maintain stability and easy handleability (flowability) of materials upon assuming various environmental changes in temperature, humidity, and the like. Methods for maintaining stability and flowability of a material include powderization, and one of the methods for powderization is spray
15 drying.

[0003] Spray drying is a method of obtaining a dry powder by spraying a solution or a liquid suspension into a gas to rapidly drying the solution or liquid suspension. The drying temperature at the time of spray drying is generally a high temperature. Enzymes, yeast, and bacterial
20 cells are denatured and deactivated when brought into contact with a high-temperature gas, while highly volatile flavorings easily undergo volatilization, and therefore, it is difficult to sufficiently maintain the original characteristics. From the viewpoint of preventing loss caused by such spray drying, suggestions are being made even to the present
25 time (for example, Patent Literature 1).

Citation List

Patent Literature

[0004] Patent Literature 1: Japanese Unexamined Patent Publication No. 2021-106571

Summary of Invention

5 Technical Problem

[0005] In order to produce granules which include a crystalline sugar and/or a crystalline sugar alcohol, and in which sugar and/or sugar alcohol in a non-crystalline state is retained in the gaps formed between sugar and/or sugar alcohols in a crystalline state, it is considered
10 important to completely dissolve the sugar and/or sugar alcohol and then precipitate crystals again before spray drying, from the viewpoints of stability as crystals, easy drying properties during spray drying as well as stability and flowability after spray drying of granules, and the like. Therefore, on the occasion of producing a liquid suspension to be
15 used for spray drying, a method of completely dissolving a sugar and/or a sugar alcohol as a raw material, and then heating and cooling the solution to precipitate some crystals again (crystal precipitation method) has been employed. However, such a crystal precipitation method involves complicated processes, and it is difficult to carry out the
20 method when an apparatus for precipitating crystals is not available. Furthermore, there are occasions in which not only crystals having an intended size but also a large number of microcrystals are generated by a precipitation operation, so that the collection ratio of granules after spray drying may be lowered.

25 [0006] An object of an aspect of the present invention is to provide a method for producing granules conveniently and efficiently, and a

granule obtainable by the method.

Solution to Problem

[0007] The present invention provides each of the following inventions.

[1] A method for producing granules, the method including:

5 a step of obtaining a crystal suspension containing at least one raw material selected from the group consisting of a crystalline sugar and a crystalline sugar alcohol, the crystal suspension including a portion of the raw material in a crystalline state and being obtained without performing dissolution of the entirety of the raw material and
10 crystallization of the raw material after the dissolution; and
a step of spray drying the crystal suspension under low temperature conditions.

[2] The method according to [1], in which the sugar and the sugar alcohol include a monosaccharide, a disaccharide, a trisaccharide, and
15 sugar alcohols of these.

[3] The method according to [1] or [2], in which the raw material is at least one selected from the group consisting of PALATINOSE, sucrose, and trehalose.

[4] The method according to any one of [1] to [3], in which the
20 spray drying is carried out under the conditions of an inlet temperature of 0°C to 60°C.

[5] The method according to any one of [1] to [4], in which the crystal suspension further contains a functional material.

[6] The method according to [5], in which the functional material is
25 an enzyme, a microorganism, or a flavoring.

[7] The method according to any one of [1] to [6], in which the

crystal suspension has a viscosity of 50 to 1550 mPa·s at 25°C.

[8] The method according to any one of [1] to [7], in which an average particle size of the sugar and/or sugar alcohol in the crystalline state in the crystal suspension is 1.0 to 15.9 μm.

5 [9] The method according to any one of [1] to [8], in which a standard deviation of the average particle size of the sugar and/or sugar alcohol in the crystalline state in the crystal suspension is 1.00 to 10.70 μm.

10 [10] The method according to any one of [1] to [9], in which a ratio of an average major axis with respect to an average minor axis of the sugar and/or sugar alcohol in the crystalline state in the crystal suspension is 1.00 to 1.70.

15 [11] The method according to any one of [1] to [10], in which a ratio of an average major axis with respect to an average minor axis of particles constituting the granules is 1.00 to 3.10.

[12] The method according to any one of [1] to [11], in which the granules have an agglomeration degree of less than 10.0% by mass.

20 [13] A granule containing at least one selected from the group consisting of a crystalline sugar and a crystalline sugar alcohol,
in which a portion of the sugar and/or the sugar alcohol is in a crystalline state, while the remaining portion is in a non-crystalline state, and a ratio of an average major axis with respect to an average minor axis of particles constituting the granule is 3.1 or less.

25 [14] The granule according to [13], in which the sugar and the sugar alcohol include a monosaccharide, a disaccharide, a trisaccharide, and sugar alcohols of these.

[15] The granule according to [13] or [14], in which at least one selected from the group consisting of the crystalline sugar and the crystalline sugar alcohol is at least one selected from the group consisting of PALATINOSE, sucrose, and trehalose.

5 [16] The granule according to any one of [13] to [15], further containing a functional material.

[17] The granule according to [16], in which the functional material is an enzyme, a microorganism, or a flavoring.

10 [18] The granule according to any one of [13] to [17], in which an average particle size of particles constituting the granule is 10.00 to 20.00 μm .

[19] The granule according to any one of [13] to [18], in which a standard deviation of the average particle size of the particles constituting the granule is 1.00 to 3.30 μm .

15 [20] The granule according to any one of [13] to [19], in which the granules have an agglomeration degree of less than 10.0% by mass.

Advantageous Effects of Invention

20 [0008] According to an aspect of the present invention, a method for producing granules conveniently and efficiently, and a granule obtainable by the method can be provided.

Brief Description of Drawings

25 [0009] FIG. 1(A) shows a microscopic image of a crystal suspension obtained by the method of Example 1, and FIG. 1(B) shows a microscopic image of a crystal suspension obtained by the method of Comparative Example 1.

FIG. 2(A) shows an electron microscopic image of granules

obtained by the method of Example 3, and FIG. 2(B) shows an enlarged image of FIG. 2(A).

FIG. 3(A) shows an electron microscopic image of granules obtained by the method of Comparative Example 3, and FIG. 3(B) shows an enlarged image of FIG. 3(A).

FIG. 4(A) shows evaluation results for the hygroscopic properties of granules obtained by the method of Example 3, and FIG. 4(B) shows evaluation results for the hygroscopic properties of granules obtained by the method of Comparative Example 3.

Description of Embodiments

[0010] Embodiments of the present invention will be described in detail below. However, the present invention is not intended to be limited to the following embodiments. With regard to numerical value ranges described stepwise in the present specification, the upper limit value or lower limit value of a numerical value range of a certain stage can be arbitrarily combined with the upper limit value or lower limit value of a numerical value range of another stage. With regard to a numerical value range described in the present specification, the upper limit value or lower limit value of the numerical value range may be replaced with a value shown in Examples.

[0011] <Method for producing granules>

A method for producing granules according to the present embodiment includes: a step (mixing step) of obtaining a crystal suspension containing at least one raw material selected from the group consisting of a crystalline sugar and a crystalline sugar alcohol, the crystal suspension including a portion of the raw material in a

crystalline state and being obtained without performing dissolution of the entirety of the raw material and crystallization of the raw material after the dissolution; and a step (spray drying step) of spray drying the crystal suspension under low temperature conditions.

5 [0012] <Mixing step>

In the mixing step, first, a crystal suspension containing at least one raw material selected from the group consisting of a crystalline sugar and a crystalline sugar alcohol, the crystal suspension including a portion of the raw material in a crystalline state, is obtained.

10 [0013] According to the present specification, the term "sugar and/or sugar alcohol in a crystalline state" means a sugar and/or sugar alcohol in a solid form, in which constituent atoms are three-dimensionally regularly repeated. According to the present specification, the term "sugar and/or sugar alcohol in a non-crystalline state" means a sugar
15 and/or sugar alcohol in a solid form or a liquid form, which does not have such regular repetitions.

[0014] The crystalline sugar and crystalline sugar alcohol preferably include a monosaccharide, a disaccharide, a trisaccharide, and sugar alcohols of these, from the viewpoint of enhancing operability in the
20 mixing step.

[0015] Examples of the monosaccharide include glucose, galactose, mannose, fructose, allose, and allulose. Examples of the disaccharide include isomaltulose, sucrose, lactulose, lactose, maltose, trehalose, and cellobiose. Examples of the trisaccharide include nigerotriose,
25 maltotriose, and raffinose. Incidentally, isomaltulose is a disaccharide registered by Mitsui DM Sugar Co., Ltd. under the trademark

"PALATINOSE".

[0016] Examples of the sugar alcohol include sorbitol, erythritol, xylitol, maltitol, lactitol, mannitol, α -glucopyranosyl-1,1-mannitol, and α -glucopyranosyl-1,6-sorbitol.

5 [0017] The above-mentioned crystalline sugars and sugar alcohols may be used singly or in combination of two or more kinds thereof. The crystal suspension may contain two or more kinds of crystalline sugars, may contain two or more kinds of crystalline sugar alcohols, or may contain two or more kinds of crystalline sugars and crystalline sugar
10 alcohols in combination. The method for producing granules according to the present embodiment is a convenient method, and granules in which the ratio of the average major axis with respect to the average minor axis of the constituent particles is sufficiently small (for example, 3.1 or less) even in the case of using two or more kinds of
15 crystalline sugars and alcohols (for example, in the case of using two kinds of sugars), can be obtained. Granules in which the ratio of the average major axis with respect to the average minor axis of constituent particles is sufficiently small, tend to have excellent stability and flowability.

20 [0018] The content of the crystalline sugar and/or sugar alcohol included in the crystal suspension is 30% by mass or more, preferably 40% by mass or more, more preferably 45% by mass or more, and even more preferably 50% by mass or more, based on the total amount of the crystal suspension, from the viewpoint of easily obtaining granules
25 having excellent flowability. From the viewpoint of maintaining satisfactory operability in the mixing step and the spray drying step, the

content of the crystalline sugar and/or sugar alcohol is preferably 90% by mass or less, more preferably 80% by mass or less, and even more preferably 70% by mass or less, based on the total amount of the crystal suspension.

5 [0019] The crystal suspension supernatant Brix value (Bx) may be 20.00 or more, 25.00 or more, 27.00 or more, 29.00 or more, or 31.00 or more from the viewpoint of easily obtaining granules having excellent flowability, and the supernatant Brix value may be 85.00 or less, 75.00 or less, 65.00 or less, 55.00 or less, 45.00 or less, 40.00 or less, or 35.00 or less from the viewpoint of maintaining satisfactory operability in the mixing step and the spray drying step.

[0020] The Brix value (Bx) according to the present specification means a Refubrix value calculated from the refractive index and can be measured by using a Brix meter (for example, a digital refractometer (RX-5000), manufactured by Atago Co., Ltd.). The method for measuring the supernatant Brix value of the crystal suspension is as described in Examples that will be described below.

20 [0021] The crystallization ratio of the crystal suspension may be 25% by mass or more, 30% by mass or more, 35% by mass or more, 40% by mass or more, 45% by mass or more, 50% by mass or more, 55% by mass or more, or 60% by mass or more, from the viewpoint of easily obtaining granules having excellent flowability. The crystallization ratio may be 90% by mass or less, 85% by mass or less, 80% by mass or less, or 78% by mass or less, from the viewpoint of maintaining satisfactory operability in the spray drying step. The crystallization ratio according to the present specification is measured by the method

described in Examples that will be described below.

[0022] The crystallization ratio can be adjusted by, for example, performing an operation of physically or chemically adding or removing crystals, such as filter filtration, centrifugal separation, gravitational sedimentation, dissolution by hydration and/or heating, or adjustment of consumption of crystal components associated with a chemical reaction. The crystallization ratio can also be adjusted by an operation of increasing crystals, such as charging and mixing of crystal components.

[0023] It is desirable that the crystal suspension contains crystal nuclei, and the size of the crystal nuclei is not particularly limited as long as it is equal to or larger than the size that can be stably present in the crystal suspension. The size of the crystal nuclei may be, for example, a size equal to or larger than that of critical crystal nuclei.

[0024] The viscosity of the crystal suspension at 25°C may be 1550 mPa·s or less, 1400 mPa·s or less, 1300 mPa·s or less, 1200 mPa·s or less, 1100 mPa·s or less, 1000 mPa·s or less, 900 mPa·s or less, 800 mPa·s or less, 700 mPa·s or less, 600 mPa·s or less, or 500 mPa·s or less, and may be 50 mPa·s or more, 100 mPa·s or more, 200 mPa·s or more, 300 mPa·s or more, 400 mPa·s or more, 500 mPa·s or more, 600 mPa·s or more, 700 mPa·s or more, 800 mPa·s or more, 900 mPa·s or more, 1000 mPa·s or more, or 1100 mPa·s or more, from the viewpoints that a decrease in the collection ratio of granules after spray drying is further suppressed, and that generation of agglomeration of granules after spray drying is further suppressed. The viscosity of the crystal suspension may be 50 to 1550 mPa·s at 25°C. The viscosity of the crystal suspension is measured by the method described in Examples

that will be described below.

5 [0025] The average particle size of the sugar and/or sugar alcohol in the crystalline state in the crystal suspension may be 1.00 μm or more, 2.00 μm or more, 3.00 μm or more, 4.00 μm or more, 5.00 μm or more, 6.00 μm or more, 7.00 μm or more, 8.00 μm or more, 9.00 μm or more, 10.00 μm or more, 11.00 μm or more, or 12.00 μm or more, from the viewpoint of maintaining flowability of the granules. The average particle size of the sugar and/or sugar alcohol in the crystalline state may be 30.00 μm or less, 25.00 μm or less, 20.00 μm or less, 18.00 μm or less, 15.90 μm or less, 14.00 μm or less, 13.00 μm or less, 12.00 μm or less, or 11.00 μm or less, from the viewpoint of further suppressing the occurrence of disintegration of the granules. The average particle size of the sugar and/or sugar alcohol in the crystalline state may be, for example, 1.00 to 15.90 μm .

15 [0026] The average particle size of the sugar and/or sugar alcohol in the crystalline state in the crystal suspension can be adjusted by means of addition or removal of a solvent or a solute, solvent temperature, dissolution time, change in the stirring time, pulverization by a stirrer or a pulverizer, fractionation by filtration or the like, a crystal precipitation operation by hydrolysis of sugar or the like.

20 [0027] The "average particle size of the sugar and/or sugar alcohol in the crystalline state" according to the present specification can be measured by means of a digital microscope. For the measurement, for example, SKM-S31B-PC manufactured by SAITOH KOUGAKU Co., Ltd can be used. The details of the method for measuring the average particle size are as described in Examples that will be described below.

[0028] The standard deviation of the average particle size of the sugar and/or sugar alcohol in the crystalline state in the crystal suspension may be 10.70 μm or less, 9.00 μm or less, 8.00 μm or less, 7.00 μm or less, or 6.00 μm or less. A small standard deviation of the average particle size means a small variation in the average particle size. As the standard deviation of the average particle size is in the above-mentioned range, excessive viscosity increase is likely to be suppressed, and as the result, adhesion of granules in a spray drying machine is further suppressed, so that the collection ratio is consequently further improved. The standard deviation of the average particle size of the sugar and/or sugar alcohol in the crystalline state in the crystal suspension may be, for example, 1.0 μm or more, 3.0 μm or more, or 5.0 μm or more. The standard deviation of the average particle size of the sugar and/or sugar alcohol in the crystalline state in the crystal suspension may be, for example, 1.00 to 10.70 μm .

[0029] According to the present specification, the "standard deviation" is unbiased standard deviation determined by the STDEV.S function of Microsoft (registered trademark) Excel.

[0030] The average minor axis of the sugar and/or sugar alcohol in the crystalline state in the crystal suspension may be 1.00 μm or more, 4.00 μm or more, 6.00 μm or more, or 8.00 μm or more, and may be 20.00 μm or less, 15.0 μm or less, or 13.0 μm or less. The average major axis of the sugar and/or sugar alcohol in the crystalline state in the crystal suspension may be 5.00 μm or more, 7.00 μm or more, or 9.00 μm or more, and may be 30.00 μm or less, 25.00 μm or less, or 20.00 μm or less.

[0031] The ratio of the average major axis (unit: μm) with respect to the average minor axis (unit: μm) of the sugar and/or sugar alcohol in the crystalline state in the crystal suspension may be 5.00 or less, 2.50 or less, 2.00 or less, 1.70 or less, or 1.50 or less, and may be 1.00 or more, 1.10 or more, or 1.15 or more, from the viewpoint of maintaining flowability and stability of the granules. The ratio of the average major axis with respect to the average minor axis may be, for example, 1.00 to 1.70. The standard deviation of the ratio of the average major axis (unit: μm) with respect to the average minor axis (unit: μm) of the sugar and/or sugar alcohol in the crystalline state in the crystal suspension may be 0.50 or less, 0.45 or less, or 0.40 or less, and may be 0.05 or more, 0.10 or more, or 0.15 or more.

[0032] The crystal suspension contains a solvent. The solvent may be, for example, an organic solvent such as ethanol, methanol, acetone, or isopropanol; or water.

[0033] The content of the solvent in the crystal suspension may be 10% by mass or more, 20% by mass or more, or 30% by mass or more, and may be 50% by mass or less or 45% by mass or less, based on the total mass of the solvent and the raw material or based on the total mass of the crystal suspension.

[0034] The above-mentioned crystal suspension may be used as it is as a crystal suspension to be spray dried (spray liquid), or may be mixed with other components and then used as a spray liquid. Examples of the other components include a sugar, a sugar alcohol, a solvent, a functional material, and mixtures of these.

[0035] The functional material is not limited as long as it is a material

or a component that exhibits some function in a composition (for
 example, a food product, a pharmaceutical product, or the like)
 obtainable by combining with other materials. The functional material
 may be a material that is affected by the ambient environment such as
 5 moisture, heat, light, acid, oxygen, molecular movement, ultraviolet
 radiation, electrical interaction, and physical stimulation, or may be a
 material that loses the function by heating. More specific examples of
 the functional material include amino acids, peptides (including
 hormones), proteins (including enzymes and antibodies), fatty acids,
 10 vitamins, minerals, microorganisms (for example, bacteria such as
 lactobacillus, butyric acid bacterium, bacillus natto, Lactobacillus
 bifidus, and actinomycetes, fungi, and yeast), hormones other than
 peptides, flavorings, phages, antibiotic substances other than peptides,
 natural raw materials such as herbal medicines, and industrial raw
 15 materials such as metals.

[0036] The crystal suspension to be spray dried (spray liquid) can be
 obtained by, for example, a method including: a step (step A) of mixing
 at least one raw material selected from the group consisting of a
 crystalline sugar and a crystalline sugar alcohol with a solvent to obtain
 20 a crystal suspension A including a portion of the raw material in a
 crystalline state; and a step (step B) of mixing the crystal suspension A
 with a sugar solution to obtain a crystal suspension B (spray liquid) to
 be spray dried, the crystal suspension B including a portion of the raw
 material in a crystalline state.

25 [0037] Mixing of the raw material and a solvent in step A may be
 carried out such that the Brix value (Bx) is 40 or more, 50 or more, 55

or more, or 60 or more and is 85 or less, 75 or less, or 70 or less.

[0038] In step A, the crystal suspension A can be obtained by, for example, mixing the raw material and the solvent and stirring the obtained mixed liquid for 1 to 3 hours while maintaining the mixed liquid at a temperature of 25°C to 35°C. The speed of rotation during stirring may be, for example, 250 to 400 rpm.

[0039] With regard to the crystal suspension A, specific embodiments of the supernatant Brix value, the crystallization ratio, the average particle size of the sugar and/or sugar alcohol in a crystalline state, the average major axis, the average minor axis, the ratio of the average major axis with respect to the average minor axis, standard deviations thereof, and the like may be the same as those mentioned as specific embodiments of the above-described crystal suspension.

[0040] In step B, the crystal suspension A and a sugar solution are mixed. Mixing of the crystal suspension A and a sugar solution may be carried out while maintaining the temperature at 25°C to 35°C.

[0041] The sugar solution includes a sugar and/or a sugar alcohol and a solvent. A portion or the entirety of the sugar and sugar alcohol included in the sugar solution may be dissolved in the solvent. Regarding the sugar and the sugar alcohol, those mentioned above can be used. The sugar solution may include one kind or two or more kinds of sugars and/or sugar alcohols. Regarding the sugar and/or sugar alcohol in the sugar solution, a sugar and/or sugar alcohol of the same kind as the sugar and/or sugar alcohol in the crystal suspension A may be included, or a sugar and/or sugar alcohol of different kind may be included.

[0042] The sugar solution can be obtained by, for example, mixing a sugar and/or a sugar alcohol with a solvent. The sugar solution may also be obtained by mixing a sugar and/or a sugar alcohol with a solvent under a temperature condition of 80°C or higher and dissolving the sugar and/or sugar alcohol. The sugar solution may be maintained at 25°C to 35°C after preparation.

[0043] The Brix value (Bx) of the sugar solution may be 50 or more, 55 or more, or 60 or more, and may be 85 or less, 75 or less, or 70 or less.

[0044] The mixing amount of the sugar solution may be 5 parts by mass or more, 10 parts by mass or more, or 15 parts by mass or more, and may be 30 parts by mass or less, 25 parts by mass or less, or 20 parts by mass or less, with respect to 100 parts by mass of the crystal suspension.

[0045] The crystal suspension B includes a portion of at least one raw material selected from the group consisting of a crystalline sugar and a crystalline sugar alcohol, in a crystalline state. The crystal suspension B can be spray dried as it is as a spray liquid.

[0046] With regard to the spray liquid, specific embodiments of the supernatant Brix value, the crystallization ratio, the average particle size of the sugar and/or sugar alcohol in a crystalline state, the average major axis, the average minor axis, the ratio of the average major axis with respect to the average minor axis, standard deviations thereof, and the like may be the same as those mentioned as specific embodiments of the above-described crystal suspension, or may be different from them.

[0047] The content of the sugar and/or sugar alcohol in the spray liquid may be 40% by mass or more, 50% by mass or more, or 60% by mass or more, and may be 90% by mass or less, 80% by mass or less, or 70%

by mass or less, based on the total mass of the solvent and the raw material or based on the total mass of the crystal suspension.

5 [0048] The spray liquid may contain a first sugar raw material selected from the group consisting of a crystalline sugar and a crystalline sugar alcohol, and a second sugar raw material that is selected from the group consisting of a crystalline sugar and a crystalline sugar alcohol and is different from the first sugar raw material. The first sugar raw material may be, for example, isomaltulose. The second sugar raw material may include one kind or two or more kinds of the above-mentioned
10 sugar or sugar alcohol as long as the sugar and sugar alcohol are different from the first sugar raw material, and may include sucrose.

[0049] The content of the first sugar raw material may be 70.0 parts by mass or more, 75.0 parts by mass or more, 80.0 parts by mass or more, 85.0 parts by mass or more, 90.0 parts by mass or more, or 95.0 parts by
15 mass or more, and may be 99.9 parts by mass or less, 95.0 parts by mass or less, or 90.0 parts by mass or less, with respect to 100 parts by mass of the solid content in the spray liquid. According to the present specification, the "solid content" means an amount obtained by subtracting the amount of the solvent (for example, water) from the total
20 amount of the composition.

[0050] When the spray liquid contains a first sugar raw material and a second sugar raw material, the content of the second sugar raw material may be 1.0 part by mass or more, 3.0 parts by mass or more, 5.0 parts by mass or more, 8.0 parts by mass or more, 10.0 parts by mass or more,
25 12.0 parts by mass or more, or 15.0 parts by mass or more, and may be 30.0 parts by mass or less, 25.0 parts by mass or less, 20 parts by mass

or less, 15 parts by mass or less, 10 parts by mass or less, or 5 parts by mass or less, with respect to 100 parts by mass of the solid content in the spray liquid.

5 [0051] When a spray liquid containing a functional material is prepared, the timing for incorporating the functional material into the spray liquid is not particularly limited; however, for example, a spray liquid may be obtained by mixing the functional material and the above-described sugar solution in advance to prepare a mixed liquid, and then mixing the mixed liquid and the crystal suspension A.

10 [0052] The content of the functional material may be 0.01 parts by mass or more, 0.05 parts by mass or more, or 0.1 parts by mass or more, and may be 5 parts by mass or less, 3 parts by mass or less, or 1 part by mass or less, with respect to 100 parts by mass of the above-mentioned crystal suspension. The amount of addition of the functional material
15 can be appropriately adjusted according to the type of the functional material.

[0053] In the mixing step, dissolution of the entirety of the raw material and crystallization of the raw material after dissolution of the entirety of the raw material are not carried out. Dissolution of the entirety of the
20 raw material can be checked by, for example, visual inspection. Regarding another method for checking the dissolution of the entirety of the raw material, for example, a method of observing induction of crystallization with a microscope may be mentioned. In a case where precipitation of crystals is not confirmed when a crystal suspension
25 dropped on a slide glass using a microscope for 5 minutes at room temperature of 25°C, it can be determined that the entirety of the raw

material has been dissolved.

[0054] The dissolution of the entirety of the raw material is usually carried out by heating a liquid including the raw material and the solvent, mixing the heated solvent and the raw material, and the like.

5 The temperature of the liquid at the time of heating or the temperature of the heated solvent is not particularly limited; however, for example, the temperature may be 70°C or higher or 75°C or higher and may be 100°C or lower. In the above-mentioned mixing step, at the time of preparing the crystal suspension, the solvent to be mixed with the raw
10 material is not heated to dissolve the entirety of the raw material.

[0055] The crystallization of the raw material after dissolution of the entirety of the raw material may be carried out by a known method. Examples of a method for crystallizing the raw material after the dissolution of the entirety of the raw material include a method of
15 cooling a dissolution liquid of the raw material (cooling crystal precipitation method) and a reaction crystal precipitation method. In the above-mentioned mixing step, the above-described operation of crystallizing the raw material is not carried out in addition to the operation of dissolving the entirety of the raw material.

20 [0056] When crystallization is conducted by a cooling crystal precipitation method, the temperature (cooling temperature) of the dissolution liquid of the raw material brought by cooling is set according to the type of the crystalline sugar and/or sugar alcohol. The cooling temperature is typically 25°C or lower or 20°C or lower and
25 may be 5°C or higher, 10°C or higher, or 15°C or higher.

[0057] <Spray drying step>

The spray drying step is a step of spray drying the above-mentioned crystal suspension (spray liquid) under low temperature conditions. According to an embodiment, spray drying can be carried out using a spray drying machine (spray dryer). As the
5 spray drying machine, for example, OC-16 manufactured by OHKAWARA KAKOHI CO., LTD. can be used.

[0058] The term low temperature conditions means lower temperature conditions (for example, 60°C or lower) compared with the temperature at which conventional spray drying is carried out (for example, a
10 temperature higher than 60°C). In the present embodiment, since a crystal suspension including a portion of the sugar and/or sugar alcohol in a crystalline state is used, even when spray drying is performed under lower temperature conditions compared to conventional cases, suitable granules can be obtained.

[0059] The low temperature conditions may be temperature conditions to the extent that the function of the functional material is not lost. In the present embodiment, for example, when an enzyme is included as the functional material, deactivation of the enzyme caused by spray drying under high temperature conditions can be suppressed. When a
15 flavoring is included as the functional material, volatilization of the flavoring caused by spray drying under high temperature conditions can be suppressed.

[0060] When it is said that spray drying is carried out under low temperature conditions, it is implied that spray drying is carried out
20 under temperature conditions in which the inlet air temperature (inlet temperature) in the spray drying machine is such as described above.

[0061] According to an embodiment, the inlet air temperature in the spray drying step is preferably 60°C or lower, 55°C or lower, 50°C or lower, 45°C or lower, 40°C or lower, 35°C or lower, 30°C or lower, 25°C or lower, 20°C or lower, or 15°C or lower. The inlet air temperature may be, for example, 0°C or higher, 5°C or higher, or 10°C or higher. That is, the inlet air temperature in the spray drying step may be 0°C to 60°C or may be 0°C to 50°C.

[0062] The outlet air temperature (air exhaust temperature) in the spray drying machine may be, for example, 50°C or lower, 40°C or lower, 35°C or lower, 30°C or lower, 25°C or lower, 20°C or lower, or 15°C or lower and may be 0°C or higher, 5°C or higher, or 10°C or higher.

[0063] The liquid temperature of the crystal suspension in the spray drying step may be, for example, 60°C or lower, 50°C or lower, or 45°C or lower and may be 10°C or higher, 15°C or higher, or 20°C or higher.

[0064] With regard to the spray drying, other conditions such as the amount of supply of the crystal suspension, the ambient temperature, and the ambient humidity may be respectively appropriately adjusted.

[0065] For example, the speed of rotation of the atomizer for the spray drying may be 3000 rpm or more, 5000 rpm or more, or 10000 rpm or more and may be 25000 rpm or less, 20000 rpm or less, or 18000 rpm or less.

[0066] The spray drying step may further include, for example, a post-drying step for the purpose of adjusting the moisture amount of the granules. The post-drying step may be, for example, a step of performing air blowing for a predetermined time against the granules adhering to the tube walls in the spray drying machine and further

volatilizing moisture of the granules. Alternatively, the post-drying step may also be a step of storing the granules obtained by the spray drying in a desiccator or the like containing silica gel for a predetermined time.

5 [0067] The agglomeration degree of the granules may be less than 10.0% by mass or 9.5% by mass or less, and may be 0.1% by mass or more, 0.5% by mass or more, 1.0% by mass or more, 1.5% by mass or more, 2.0% by mass or more, 2.5% by mass or more, 3.0% by mass or more, or 4.0% by mass or more. The agglomeration degree of the
10 granules means the proportion of the portion that does not pass through a sieve having an opening size of 2 mm and a sieve having an opening size of 1 mm among the granules one week after the spray drying, and the agglomeration degree is measured by the method described in Examples that will be described below.

15 [0068] The collection ratio of the granules according to the present embodiment may be 40.0% by mass or more or 45.0% by mass or more and may be 100.0% by mass or less, 90.0% by mass or less, 80.0% by mass or less, or 75.0% by mass or less. The collection ratio of the granules is measured by the method described in Examples that will be
20 described below.

[0069] <Granule>

A granule according to the present embodiment contains at least one selected from the group consisting of a crystalline sugar and a crystalline sugar alcohol, and the crystalline sugar and/or sugar alcohol
25 is such that a portion is in a crystalline state while the other portion is in a non-crystalline state. The ratio of the average major axis with

respect to the average minor axis of the particles constituting the granules is 3.1 or less. The granule can be obtained by, for example, the above-mentioned method for producing granules.

[0070] The granule may further include a functional material.

5 Detailed embodiments of the crystalline sugar, the crystalline sugar alcohol, and the functional material are the same as described above, and therefore, further description will not be repeated. The "granule" according to the present specification is an aggregate of particles, and the particles constituting the granule (granular particles) contain one
10 kind or two or more kinds selected from the group consisting of a crystalline sugar and a crystalline sugar alcohol.

[0071] The granule may contain two or more kinds of crystalline sugars, may contain two or more kinds of crystalline sugar alcohols, or may contain two or more kinds of crystalline sugars and crystalline
15 sugar alcohols in combination.

[0072] The granule may contain a first sugar raw material selected from the group consisting of a crystalline sugar and a crystalline sugar alcohol, and a second sugar raw material that is selected from the group consisting of a crystalline sugar and a crystalline sugar alcohol and is
20 different from the first sugar raw material. When the granule contains the first sugar raw material and the second sugar raw material, the flowability of the granule and the stability of functional material and the granule become more excellent.

[0073] The first sugar raw material may be, for example, isomaltulose.

25 The second sugar raw material may include one kind or two or more kinds of the above-mentioned sugars or sugar alcohols as long as they

are different from the first sugar raw material, and may include sucrose.

[0074] The content of the first sugar raw material may be 70.0 parts by mass or more, 75.0 parts by mass or more, 80.0 parts by mass or more, 85.0 parts by mass or more, 90.0 parts by mass or more, or 95.0 parts by mass or more, and may be 99.9 parts by mass or less, 95.0 parts by mass or less, or 90.0 parts by mass or less, with respect to 100 parts by mass of the total amount of the crystalline sugar and crystalline sugar alcohol.

[0075] When the granule contains the second sugar raw material, the content of the second sugar raw material may be 1.0 part by mass or more, 3.0 parts by mass or more, 5.0 parts by mass or more, 8.0 parts by mass or more, 10.0 parts by mass or more, 12.0 parts by mass or more, or 15.0 parts by mass or more, and may be 30.0 parts by mass or less, 25.0 parts by mass or less, 20 parts by mass or less, 15 parts by mass or less, 10 parts by mass or less, or 5 parts by mass or less, with respect to 100 parts by mass of the total amount of the crystalline sugar and the crystalline sugar alcohol.

[0076] The granule according to an embodiment is such that a portion of the crystalline sugar and/or sugar alcohol is in a crystalline state, and the granule is composed of granular particles in which these sugar and/or sugar alcohols in the crystalline state are aggregated. In this case, it is preferable that the other portion (remaining portion) of the crystalline sugar and/or sugar alcohol is in a non-crystalline state and is retained in the gaps formed between the aggregated sugar and/or sugar alcohols in the crystalline state. When the granule includes a functional material, it is preferable that the functional material is also retained in the gaps formed between the sugar and/or sugar alcohols in

the crystalline state.

[0077] Whether the sugar and/or sugar alcohols in the crystalline state are aggregated can be checked by observing the external appearance of granular particles or the morphology of a fracture surface of a granular particle by means of a scanning electron microscope (SEM) or a digital microscope. Whether the sugar and/or sugar alcohol in a non-crystalline state and the functional material are retained in the above-described gaps can be checked by the following method.

(1) The morphology of a granule in the middle of temperature increase is observed with a differential scanning calorimeter (DSC, for example, REALVIEW DSC (TA7000) manufactured by Hitachi High-Tech Science Corporation). As a result, it can be visually recognized that the sugar or sugar alcohol in a non-crystalline state undergoes glass transition as a result of temperature increase.

(2) The difference in polarization properties between the crystalline state and the non-crystalline state is visually recognized with a polarizing microscope (for example, polarizing microscope (MT9200L) manufactured by MEIJI TECHNO CO., LTD.).

[0078] The number of sugar and/or sugar alcohol particles in the crystalline state (number of crystals) included in the particles constituting the granule is, for example, 10 or more, and may be 50 or more or 100 or more. The number of crystals may also be 1000 or less. The number of crystals can be measured by making observation by means of a scanning electron microscope and then counting by visual inspection.

[0079] The shape of the particles constituting the granule may be an

approximately spherical shape from the viewpoint of having more excellent flowability of the granule. The particles constituting the granule may have surface unevenness on the surface of the particles, from the viewpoint of having more excellent flowability.

5 [0080] The average particle size of the granule may be 10.00 μm or more, 20.00 μm or more, 40.00 μm or more, 60.00 μm or more, 80.00 μm or more, or 90.00 μm or more from the viewpoint that the granule are further less likely to agglomerate and have satisfactory flowability, and the average particle size may be 200.00 μm or less, 150.00 μm or less,
10 less, 140.00 μm or less, 135.00 μm or less, or 118 μm or less from the viewpoint of having more satisfactory solubility. The average particle size of the granule may be, for example, 20.00 to 118.00 μm .

[0081] The "average particle size of the granule" according to the present specification can be measured by using an electron microscope.
15 For the measurement, for example, Miniscope TM3030 manufactured by Hitachi High-Tech Co., Ltd. can be used. The details of the method for measuring the average particle size of the granule are as described in Examples that will be described below.

[0082] The standard deviation of the average particle size of the granule
20 may be 40 μm or less or 30 μm or less, and may be 5 μm or more or 10 μm or more, from the viewpoint that the granules are further less likely to agglomerate.

[0083] The average minor axis of the granule may be 80 μm or more or 95 μm or more, and may be 150 μm or less or 130 μm or less. The
25 average major axis of the granule may be 75 μm or more or 95 μm or more, and may be 200 μm or less or 150 μm or less.

[0084] The ratio of the average major axis with respect to the average minor axis of the granule may be 1.40 or less, 1.20 or less, 1.10 or less, or 1.04 or less, and may be 1.00 or more or 1.01 or more, from the viewpoint that the granule has more satisfactory stability and flowability. The ratio of the average major axis with respect to the average minor axis of the granule may be, for example, 1.00 to 1.04. The standard deviation of the ratio of the average major axis with respect to the average minor axis of the granule may be 0.20 or less or 0.12 μm or less and may be 0.01 μm or more, from the viewpoint that the granule is easily dryable, is less likely to agglomerate, and has more satisfactory flowability.

[0085] The average particle size of the particles constituting the granule may be 5.00 μm or more, 10.00 μm or more, or 15.00 μm or more, and may be 50.00 μm or less, 40.00 μm or less, 30.00 μm or less, 25.00 μm or less, or 20.00 or less. The average particle size of the particles constituting the granule may be, for example, 10.00 to 20.00 μm .

[0086] The standard deviation of the average particle size of the particles constituting the granule may be 10.00 μm or less, 8.00 μm or less, 6.00 μm or less, 4.00 μm or less, or 3.3 μm or less, and may be 1.00 μm or more or 2.00 μm or more, from the viewpoint that the granule has more satisfactory stability and flowability. The standard deviation of the average particle size of the particles constituting the granule may be, for example, 1.00 to 3.30 μm .

[0087] The average minor axis of the particles constituting the granule may be 5.0 μm or more or 10.0 μm or more, and may be 30.0 μm or less or 25.0 μm or less. The average major axis of the particles constituting

the granule may be 10.0 μm or more or 15.0 μm or more, and may be 50 μm or less or 35 μm or less.

[0088] The ratio of the average major axis with respect to the average minor axis of the particles constituting the granule is 3.1 or less. When the ratio of the average major axis with respect to the average minor axis of the particles constituting the granule is 3.1 or less, the granule acquires excellent stability and flowability. The ratio of the average major axis with respect to the average minor axis of the particles constituting the granule is 2.8 or less, 2.5 or less, 2.2 or less, or 1.9 or less, and may be 0.5 or more, 1.0 or more, or 1.4 or more, from the viewpoint of obtaining more satisfactory stability and flowability.

[0089] The standard deviation of the ratio of the average major axis with respect to the average minor axis of the particles constituting the granule may be 0.4 or less or 0.3 or less, and may be more than 0.0 or 0.1 or more.

[0090] The total pore volume of the granule may be 0.0265 cm^3/g or more, 0.0270 cm^3/g or more, 0.0275 cm^3/g or more, 0.0280 cm^3/g or more, or 0.0290 cm^3/g or more, and may be 0.0400 cm^3/g or less, 0.0350 cm^3/g or less, 0.0300 cm^3/g or less, or 0.0290 cm^3/g or less.

[0091] The total specific surface area of the granule may be 2.000 m^2/g or more, 4.000 m^2/g or more, 6.000 m^2/g or more, or 8.000 m^2/g or more, and may be 10.000 m^2/g or less, 9.000 m^2/g or more, or 8.5000 m^2/g or less.

[0092] The average pore diameter of the granule may be 60000 $\text{\AA}$ or less, 40000 $\text{\AA}$ or less, 20000 $\text{\AA}$ or less, 10000 $\text{\AA}$ or less, 5000 $\text{\AA}$ or less, 1000 $\text{\AA}$ or less, 800 $\text{\AA}$ or less, or 600 $\text{\AA}$ or less, and may be 200 $\text{\AA}$ or

more, 400 Å or more, or 550 Å or more, from the viewpoint that the granule has more satisfactory stability.

[0093] The porosity of the granule may be 3.80% or more, 3.90% or more, or 3.95% or more, and may be 8.00% or less, 5.00% or less, 5 4.50% or less, or 4.10% or less.

[0094] The aroma retention ratio of granule containing a flavoring may be 50% or more, 60% or more, 70% or more, 80% or more, 90% or more, or 100% or more, and may be 150% or less, 140% or less, or 130% or less.

10 [0095] The total pore volume, the total specific surface area, the average pore diameter, porosity, and the aroma retention ratio of the granule can be measured by the methods described in Examples that will be described below.

[0096] In the method for producing granules according to the present embodiment, a step of completely dissolving a raw material, a step of 15 crystallizing the raw material, a step of performing drying by vacuum freezing, or a step of heating a crystal suspension intended for complete dissolution or crystallization of the raw material before spray drying can be omitted, and therefore, it is possible to produce granules by a method 20 more convenient than conventional methods.

[0097] In the granule according to the present embodiment, the occurrence of agglomeration is suppressed while the granule can be efficiently produced by a more convenient operation. Furthermore, while the granule according to the present embodiment can be 25 efficiently produced by a more convenient operation, deterioration of hygroscopic properties, flowability, and jet flow properties are also

suppressed.

[0098] Since granules are obtained by spray drying under low temperature conditions, when a granule contains a functional material, the function of the included functional material is less likely to be lost by heat. That is, even after this granule is subjected to a drying step, the granule still satisfactorily maintains the function of the functional material. Usually, when a granule is obtained by spray drying, it is difficult to obtain a suitable granule without performing spray drying under higher temperature conditions. However, with regard to the granule according to the present embodiment, a suitable granule can be easily obtained even under low temperature conditions.

[0099] The granule according to the above-mentioned embodiment can be used as, for example, a material that is added to food products, food additives, pharmaceutical products, cosmetic products, quasi-pharmaceutical products or pharmaceutical products, livestock feed, fertilizers, antibiotic substances, and soil improving agents.

Examples

[0100] Hereinafter, the present invention will be described more specifically by way of Examples. However, the present invention is not intended to be limited to the following Examples.

[0101] <Example 1>

(Preparation of crystal suspension)

Into a metal container having a volume of 8 L, 3.25 kg of PALATINOSE (registered trademark of Mitsui DM Sugar Co., Ltd.) (Mitsui DM Sugar Co., Ltd., PST-N) and 1.75 kg of water were introduced and diluted to a Brix (Bx) of 65, and a crystal suspension

was obtained. Thereafter, while the temperature of the crystal suspension was maintained at 30°C, the crystal suspension was stirred for 2 hours with a stirrer (Shinto Scientific Co., Ltd., three-one motor, BL-1200) at 350 rpm, and crystal suspension A1 was obtained.

5 [0102] (Preparation of spray liquid)

900 g of granulated sugar (Mitsui DM Sugar Co., Ltd., GN) and 300 g of ion-exchanged water warmed to 80°C or higher were introduced into a 3L plastic Jug, and the mixture was stirred until the sugar was completely dissolved. The obtained sugar solution was
10 diluted with ion-exchanged water until the Bx reached 60, and 1.5 L of a sugar solution with a Bx of 60 was prepared. The prepared sugar solution with a Bx of 60 was maintained at a temperature of 30°C in a water bath (TAITEC CORPORATION, PERSONAL-11). The crystal suspension A1 was mixed with 0.96 kg of the above-mentioned sugar
15 solution with a Bx of 60, and a spray liquid having a solid content ratio of PALATINOSE : sucrose of 85 : 15 was prepared.

[0103] (Spray drying)

About 4 kg of the spray liquid maintained at 30°C was spray dried by using a spray drying machine (OHKAWARA KAKOHKI CO.,
20 LTD., OC-16 (dry type)). The conditions for spray drying were set to the conditions shown below.

Air blowing temperature: 40°C

Air exhaust temperature: 28°C

Blower inverter frequency: 60 Hz

25 Air exhaust volume: 38 Hz

Controlled pressure in the tower: Slight positive pressure

Speed of rotation of atomizer: 14000 rpm (setting: 33 Hz)

Raw material supply rate: 50 mL/min, 4.0 kg/h (setting: 8.5 Hz)

Apparatus: OC-16 (dry type)

[0104] <Example 2>

5 (Preparation of crystal suspension)

Crystal suspension A2 was obtained in the same manner as in the procedure for obtaining the crystal suspension A1 of Example 1, except that 6.5 kg of PST-N and 3.5 kg of water were used, and that the speed of rotation of the stirrer was changed to 300 rpm.

10 [0105] (Preparation of spray liquid)

1800 g of granulated sugar (Mitsui DM Sugar Co., Ltd., GN) and 600 g of ion-exchanged water warmed to 80°C or higher were introduced into a 3L plastic Jug, and the mixture was stirred until the sugar was completely dissolved. The sugar solution was diluted with ion-exchanged water until the Bx reached 60, and 1.5 L of a sugar solution with a Bx of 60 was prepared. The prepared sugar solution with a Bx of 60 was maintained at a temperature of 30°C in a water bath (TAITEC CORPORATION, PERSONAL-11). 1.9 kg of the sugar solution with a Bx of 60 and 90.0 g of ion-exchanged water were mixed, and a sugar solution without flavoring was prepared. The crystal suspension A2 was mixed with 1.90 kg of the above-mentioned sugar solution without flavoring, and a spray liquid having a solid content ratio of PALATINOSE : sucrose of 85 : 15 was prepared.

[0106] (Spray drying)

25 While about 10 kg of the spray liquid was maintained at 30°C, the entire amount was spray dried by using a Mohno pump (HEISHIN

Ltd., 3NTL08PUL). The conditions for the spray drying were set to the conditions shown below.

Air blowing temperature: 40°C

Air exhaust temperature: 28°C to 30°C

5 Air blow volume: 60 Hz

Air exhaust volume: 38 Hz

Static pressure in the tower: Slight positive pressure

Speed of rotation of atomizer: 14000 rpm (setting: 33 Hz)

Type of atomizer: Disc type atomizer

10 Raw material supply rate: 50 mL/min, 4.0 kg/h (setting: 8.5 Hz)

Apparatus: OC-16 (dry type)

[0107] <Example 3>

(Preparation and spray drying of crystal suspension)

15 19.0 g of a muscovado flavoring (TAKASAGO INTERNATIONAL CORPORATION, KOKUTO MICRON H-80210) and 71.0 g of ion-exchanged water were mixed, and an aqueous solution of the muscovado flavoring was prepared. 1.9 kg of the sugar solution with a Bx of 60 obtained by the method described in "(Preparation of spray liquid)" of Example 2 and 90.0 g of the aqueous solution of the muscovado flavoring were mixed, and a flavoring-containing sugar solution was prepared. 1.90 kg of the flavoring-containing sugar solution was mixed with the crystal suspension A2 obtained by the method of Example 2, and a spray liquid having a solid content ratio of PALATINOSE : sucrose of 85 : 15 was prepared. Spray drying was performed under the same conditions as in Example 2, by using the obtained spray liquid.

[0108] <Examples 4 to 7>

(Preparation of crystal suspension)

3.25 kg of PST-N and 2.4 kg of water were introduced into a metal container having a volume of 8 L, and the mixture was diluted to a Bx of 57.5. Thereafter, while the diluted substance was maintained at a temperature of 30°C, the diluted substance was stirred for 2 hours with a stirrer (Shinto Scientific Co., Ltd., three-one motor, BL-1200) at 300 rpm, and a crystal suspension (c1) was obtained.

[0109] (Preparation of spray liquid)

1800 g of granulated sugar (Mitsui DM Sugar Co., Ltd., GN) or 1800 g of PST-N was introduced into a 3L plastic Jug, 1200 g of ion-exchanged water warmed to 80°C or higher was introduced thereinto, and the mixture was stirred until the sugar was completely dissolved. Each of the obtained sugar solutions was diluted with ion-exchanged water until the Bx reached 60, and as a result, a sucrose syrup with a Bx of 60 including granulated sugar and a PALATINOSE syrup with a Bx of 60 including PST-N were prepared as sugar solutions (a) with a Bx of 60. The prepared sugar solutions (a) with a Bx of 60 were maintained at a temperature of 30°C by using a water bath (TAITEC CORPORATION, PERSONAL-11).

[0110] An aqueous solution of muscovado flavoring (b) was prepared by mixing 9.5 g of a muscovado flavoring (TAKASAGO INTERNATIONAL CORPORATION, KOKUTO MICRON H-80210) and 40.5 g of ion-exchanged water.

[0111] The sugar solution (a) with a Bx of 60 and the aqueous solution of muscovado flavoring (b) were mixed so as to obtain the composition

shown in Table 1, and the resulting mixed liquid was mixed with the crystal suspension (c1). As a result, a spray liquid having the composition shown in Table 2 was prepared.

[Table 1]

		Example 7 sugar 0%	Example 6 sugar 5%	Example 5 sugar 10%	Example 4 sugar 15%
Crystal suspension (c1)		5650 g	5650 g	5650 g	5650 g
Aqueous solution of muscovado flavoring (b)		50 g	50 g	50 g	50 g
Sugar solution (a) with Bx of 60	Sucrose syrup (Bx 60.0)	0 g	320 g	640 g	960 g
	PALATINOSE syrup (Bx 60.0)	960 g	640 g	320 g	0 g
Total		6660 g	6660 g	6660 g	6660 g

[Table 2]

	Example 7 sugar 0%		Example 6 sugar 5%		Example 5 sugar 10%		Example 4 sugar 15%	
	Solid content (g)	Ratio (%)	Solid content (g)	Ratio (%)	Solid content (g)	Ratio (%)	Solid content (g)	Ratio (%)
PALATINOSE	3825	99.75	3633	94.76	3441	89.75	3249	84.7
Sucrose	0	0.00	192	5.01	384	10.01	576	15.02
Muscovado flavoring	9.5	0.25	9.5	0.25	9.5	0.25	9.5	0.25
Total	3834	100.0	3834	100.0	3834	100.0	3834	100.0

[0112] (Spray drying)

While the spray liquid was maintained at 30°C, the entire
5 amount (6.7 kg) thereof was spray dried by using a Mohno pump
(HEISHIN Ltd., 3NTL08PUL) under the conditions shown below.

Air blowing temperature: 30°C

Air exhaust temperature: 28.1°C to 24.0°C

Air blow volume: 60 Hz

10 Air exhaust volume: 38 Hz

Static pressure in the tower: Slight positive pressure

Speed of rotation of atomizer: 14000 rpm (setting: 33 Hz)

Type of atomizer: Disc type atomizer

Raw material supply rate: 50 mL/min, 4.0 kg/h (setting: 8.5 Hz)

15 Apparatus: OC-16 (dry type)

[0113] After completion of spraying, without changing the operation

conditions for the spray drying machine, dry air was continuously blown for 40 minutes as it was, and the inside of the drying machine was dried. Thereafter, a dried substance was collected by scraping off.

[0114] <Comparative Example 1>

5 (Preparation of suspension for comparison)

3.25 kg of PST-N and 1.75 kg of water were introduced into a metal container having a volume of 8 L, and the mixture was heated in a water bath for one hour at 80°C. After it was confirmed that the sugar was completely dissolved, water was introduced into the metal container to adjust the mixture to a Bx of 65. The suspension in the metal container was cooled to reach 20°C by using a water bath containing water at 7°C. While being stirred at 300 rpm, the suspension was irradiated for 15 minutes by means of an ultrasonic wave oscillator (SMT Co., Ltd., ULTRA SONIC HOMOGENIZER, UH-600S) (memory 8, continuous irradiation). Thereafter, while the temperature of the content was maintained at 30°C, the content was stirred for 1 hour and 45 minutes with a stirrer at 300 rpm, and a suspension for comparison B1 was obtained.

[0115] (Preparation and spray drying of spray liquid)

20 Preparation and spray drying of the spray liquid were carried out in the same manner as in Example 1, except that the suspension for comparison B1 was used instead of the crystal suspension A1.

[0116] <Comparative Example 2>

(Preparation of suspension for comparison)

25 A suspension for comparison B2 was obtained by the same procedure for obtaining the suspension for comparison B1 of

Comparative Example 1, except that 6.5 kg of PST-N and 3.5 kg of water were used.

[0117] (Preparation and spray drying of crystal suspension)

5 Preparation and spray drying of the spray liquid were carried out in the same manner as in Example 2, except that the suspension for comparison B2 was used instead of the crystal suspension A2.

[0118] <Comparative Example 3>

(Preparation and spray drying of suspension for comparison)

10 The suspension for comparison B1 was mixed with 1.90 kg of the above-mentioned flavoring-containing sugar solution, and a spray liquid having a solid content ratio of PALATINOSE : sucrose of 85 : 15 was prepared. Spray drying was carried out under the same conditions as in Example 2 by using the obtained spray liquid.

[0119] <Comparative Examples 4 to 7>

15 (Preparation of suspension for comparison)

3.25 kg of PST-N and 2.4 kg of water were introduced into a metal container having a volume of 8 L, and the mixture was heated in a water bath for one hour at 80°C. After it was confirmed that the sugar was completely dissolved, water was introduced into the metal container to adjust the mixture to a Bx of 57.5. The metal container having a volume of 8 L was put into a water bath containing water at 7°C to cool the content until the temperature reached 20°C. While being stirred at 300 rpm, the suspension was irradiated for 15 minutes by means of an ultrasonic wave oscillator (SMT Co., Ltd., ULTRA SONIC HOMOGENIZER, UH-600S) (memory 8, continuous irradiation).
25 Thereafter, while the temperature of the content was maintained at

30°C, the content was stirred for 1 hour and 45 minutes with a stirrer at 300 rpm, and a suspension for comparison (c2) was obtained.

[0120] (Preparation and spray drying of spray liquid)

5 Preparation and spray drying of the spray liquid were carried out in the same manner as in Examples 4 to 7, except that the suspension for comparison (c2) was used instead of the crystal suspension (c1).

[Table 3]

	Presence or absence of crystallization step after raw material dissolution	Presence or absence of flavoring addition	Sugar %	Suspension Bx	Inlet temperature °C
Example 1	Absent	Absent	15	65	40
Example 2	Absent	Absent	15	65	40
Example 3	Absent	Present	15	65	40
Example 4	Absent	Present	15	57.5	30
Example 5	Absent	Present	10	57.5	30
Example 6	Absent	Present	5	57.5	30
Example 7	Absent	Present	0	57.5	30
Comparative Example 1	Present	Absent	15	65	40
Comparative Example 2	Present	Absent	15	65	40
Comparative Example 3	Present	Present	15	65	40
Comparative Example 4	Present	Present	15	57.5	30
Comparative Example 5	Present	Present	10	57.5	30
Comparative Example 6	Present	Present	5	57.5	30
Comparative Example 7	Present	Present	0	57.5	30

[0121] <Brix value (Bx)>

The Bx was measured with a Bx refractometer (Atago Co., Ltd., RX-5000α).

[0122] <Crystallization ratio>

5 The crystallization ratio was calculated by introducing 1 g of a crystal suspension or a spray liquid into an Eppendorf tube having a capacity of 1.5 mL, performing centrifugation using a centrifuge (Sakuma Seisakusho Co., Ltd., M150IV) at 16000 rpm for one minute, removing the supernatant to obtain a residual crystal mass, and applying
10 the residual crystal mass and the mass of the crystal suspension or spray liquid into the following formula.

Crystallization ratio (% by mass) = Residual crystal mass (g)/(mass of crystal suspension or spray liquid (g)) × 100

[0123] <Viscosity>

15 Regarding the viscosity, the viscosity at a liquid temperature of 25°C was measured by using a B type viscometer (TOKYO KEIKI INC., BL type). The viscometer was equipped with adaptor No. 3, and the measuring unit was immersed in the object substance at a speed of rotation of 12 rpm for 15 seconds. Thereafter, the reading value was
20 recorded. Based on the conversion coefficient of the B type viscometer, the viscosity (mPa·s) of the object substance was calculated by multiplying the reading value by 100.

[0124] <External appearance of granules>

25 Evaluation of the external appearance of the obtained granules was carried out by using an electron microscope (Hitachi High-Tech Co., Ltd., TM3030-Plus).

[0125] <Evaluation of product collection ratio (yield)>

The product collection ratio (yield) (% by mass) was determined from the proportion of the granule weight (g) after spray drying with respect to the solid content weight (g) in the spray liquid.

5 [0126] <Evaluation of water activity>

The water activity (%) was measured by means of a moisture activity measuring apparatus (Meter Japan, Inc., Dew Point water activity Meter AquaLAB Series 4TE) by using 5 g or 10 mL of granules.

[Table 4]

	Crystal suspension			Spray liquid			Granules	
	Viscosity (mPa·s)	Supernatant Bx	Crystallization ratio (% by mass)	Viscosity (mPa·s)	Supernatant Bx	Crystallization ratio (% by mass)	Collection ratio (yield) (% by mass)	Water activity (Aw)
Example 1	-	34.95	73.4	-	-	-	48.4	0.24
Example 2	450	32.64	76.1	300	-	-	67.3	0.33
Example 3	600	32.73	76.0	400	-	-	69.1	0.36
Example 4	1100	32.80	65.9	600	42.11	48.3	54.8	0.72
Example 5	1200	33.90	64.1	600	38.87	55.1	57.9	0.81
Example 6	1100	32.64	66.1	400	36.29	59.9	61.0	0.73
Example 7	1100	33.60	64.6	200	34.11	63.7	53.7	0.45
Comparative Example 1	-	35.46	72.8	-	-	-	21.9	0.30
Comparative Example 2	2000	33.01	75.7	1300	-	-	26.4	0.32
Comparative Example 3	3700	35.53	72.7	1700	-	-	27.5	0.35
Comparative Example 4	1800	33.90	64.1	1000	41.46	49.8	32.6	0.78
Comparative Example 5	1600	34.01	63.9	800	38.08	56.6	39.9	0.72
Comparative Example 6	1700	32.23	66.8	600	35.08	62.1	23.7	0.72
Comparative Example 7	2000	32.01	67.1	300	33.41	64.9	25.0	0.48

[0127] Even when the Bx and the crystallization ratio are equal, in the method of Examples, crystals having an appropriately small crystal diameter in the crystal suspension and the spray liquid are uniformly produced, the viscosity was low, and the collection ratio of the granules after spray drying was satisfactory. In the methods of Examples and the Comparative Examples, the water activity was equal.

[0128] <Evaluation of agglomeration degree of granules>

(1) Sieve evaluation (only some of samples)

1 to 2 kg of the obtained granules were placed in an aluminum pack (length below chunks 340 mm × 240 mm), the aluminum pack was hermetically sealed and stored for one week at room temperature of 25°C, subsequently the granules were passed through each of a sieve having an opening size of 2 mm and a sieve having an opening size of 1 mm, the weights the granules remaining on the sieve and the granules passing through the sieve were measured, and the agglomeration degree was evaluated according to the following criteria.

Weak: Less than 10% of the granules remain on the sieve.

Medium: 10% or more and less than 30% of the granules remain on the sieve.

Strong: 30% or more of the granules remain on the sieve.

[0129] (2) Checking by visual inspection

500 g each of the granules that had been placed in an aluminum pack, hermetically sealed, and stored for one month or longer at room temperature of 25°C were spread on black paper, and the agglomeration degree was checked by visual inspection according to the following criteria. Table 6 shows the evaluation results for the granules obtained

by the methods of Example 1 and Comparative Example 1.

Weak: Less than 10% of the granules agglomerate in a lump-like form.

5 Medium: 10% or more and less than 30% of the granules agglomerate in a lump-like form.

Strong: 30% or more of the granules agglomerate in a lump-like form.

[Table 5]

	Agglomeration degree after one week of storage			
	2 mmON (%)	1 mmON (%)	2 mmON + 1 mmON (%)	Agglomeration degree: sieve evaluation
Example 4	2.9	6.7	9.5	Weak
Example 5	2.7	5.4	8.1	Weak
Example 6	3.4	6.0	9.4	Weak
Example 7	1.0	3.9	4.9	Weak
Comparative Example 4	0.8	12.8	13.6	Medium
Comparative Example 5	3.3	30.1	33.3	Medium
Comparative Example 6	13.2	27.5	40.7	Medium
Comparative Example 7	0.0	4.2	4.2	Weak

[Table 6]

	Agglomeration degree upon storage for one month or after one month: checking by visual inspection
Example 1	Weak
Comparative Example 1	Medium

10 [0130] As shown in Table 5, a significant number of the granules obtained by the method of Comparative Examples of a conventional method (crystal precipitation method) exhibited agglomeration of a medium degree, whereas agglomeration was hardly recognized in the granules produced by the method of Examples.

15 [0131] <Crystal diameter of particles in crystal suspension and spray liquid>

A crystal suspension or a spray liquid was dropped on a slide glass, a cover glass was mounted thereon, and measurement was made at a magnification ratio of 1000 times. In the method for measuring the average particle size, for any ten or more sugar crystals, an average of the particle sizes of each ten or more sugar crystals constituting those sugar crystals was obtained with a digital microscope (SAITOH KOUGAKU Co., Ltd, SKM-S31B-PC). For ten or more particles as the objects of measurement in the crystal suspension or spray liquid, the distance in a first direction and the distance in a second direction when the distance in the first direction and the distance in the second direction orthogonally intersecting the first direction intersect each other at the halfway point of each distance, were measured as transverse width and longitudinal width, respectively. The first direction is the same direction for all the particles taken as the objects of measurement. Between the measured transverse width and longitudinal width, the longer one was designated as major axis, the shorter one was designated as minor axis, and the averages for the particles as the objects of measurement were designated as average major axis and average minor axis. FIG. 1(A) and FIG. 1(B) respectively show microscopic images of the crystal suspensions obtained by the methods of Example 1 and Comparative Example 1.

[0132] <Crystal diameters of granules and particles constituting granules>

The granule diameter and the crystal diameter of the particles constituting the granules were measured by using an electron microscope (Hitachi High-Tech Corporation, Miniscope TM3030).

Ten each of the granules obtained by the methods of Examples and Comparative Examples were observed, the longer width and the shorter width of each granule were measured, and the average value of the granule diameters was determined. The longer widths and the shorter widths of three to ten crystals included in the granules were respectively measured, and the average value of the crystal diameter in the granules was determined. FIG. 2 shows an electron microscopic image of the granules obtained by the method of Example 3. FIG. 3 shows an electron microscopic image of the granules obtained by the method of Comparative Example 3.

[0133] <Standard deviation>

The standard deviations of the average particle size, the average major axis, the average minor axis, and the ratio of the average major axis with respect to the average minor axis were calculated by the STDEV.S function of Microsoft (registered trademark) Excel.

[Table 7]

	Crystal suspension							
	Average particle size (μm)		Average minor axis (μm)		Average major axis (μm)		Ratio of average major axis (μm) with respect to average minor axis (μm)	
	Average value	Standard deviation	Average value	Standard deviation	Average value	Standard deviation	Average value	Standard deviation
Example 1	-	-	-	-	-	-	-	-
Example 2	-	-	-	-	-	-	-	-
Example 3	-	-	-	-	-	-	-	-
Example 4	11.12	5.08	9.07	3.03	13.16	6.00	1.41	0.35
Example 5	14.78	5.56	12.81	4.36	16.75	6.13	1.31	0.18
Example 6	9.23	2.09	8.57	2.28	9.89	1.74	1.18	0.18
Example 7	13.14	5.49	12.04	5.29	14.24	5.74	1.22	0.23
Comparative Example 1	-	-	-	-	-	-	-	-
Comparative Example 2	-	-	-	-	-	-	-	-
Comparative Example 3	-	-	-	-	-	-	-	-
Comparative Example 4	20.05	10.81	15.06	6.50	25.04	12.20	1.79	0.80
Comparative Example 5	16.01	15.03	9.02	4.60	22.99	18.64	2.36	0.87
Comparative Example 6	16.44	10.96	10.76	6.51	22.12	11.80	2.34	0.96
Comparative Example 7	22.43	21.74	10.40	5.25	34.46	25.47	3.58	2.22

[Table 8]

	Spray liquid							
	Average particle size (μm)		Average minor axis (μm)		Average major axis (μm)		Ratio of average major axis (μm) with respect to average minor axis (μm)	
	Average value	Standard deviation	Average value	Standard deviation	Average value	Standard deviation	Average value	Standard deviation
Example 1	-	-	-	-	-	-	-	-
Example 2	12.58	6.53	10.19	6.30	14.97	6.14	1.70	0.73
Example 3	10.54	4.45	9.37	3.48	11.70	5.16	1.25	0.23
Example 4	11.93	6.41	10.79	5.62	13.07	7.22	1.19	0.12
Example 5	12.20	6.28	10.47	4.69	13.92	7.40	1.31	0.18
Example 6	10.71	6.64	8.90	3.60	12.51	8.53	1.35	0.51
Example 7	10.02	7.78	9.25	7.66	10.79	8.23	1.23	0.24
Comparative Example 1	-	-	-	-	-	-	-	-
Comparative Example 2	10.30	7.13	6.05	2.62	14.54	7.78	2.87	2.39
Comparative Example 3	7.70	3.92	5.40	2.19	9.99	3.98	1.90	0.64
Comparative Example 4	16.01	13.98	10.96	10.21	21.06	15.87	2.24	1.24
Comparative Example 5	18.46	13.86	14.96	9.62	21.97	16.91	1.38	0.36
Comparative Example 6	17.51	14.98	15.83	20.42	19.20	7.11	2.10	1.22
Comparative Example 7	20.01	10.09	14.37	2.64	25.66	11.71	1.80	0.80

[Table 9]

	Granules							
	Average particle size (μm)		Average minor axis (μm)		Average major axis (μm)		Ratio of average major axis (μm) with respect to average minor axis (μm)	
	Average value	Standard deviation	Average value	Standard deviation	Average value	Standard deviation	Average value	Standard deviation
Example 1	131.75	23.81	127.16	24.74	136.34	23.65	1.08	0.08
Example 2	107.10	15.71	102.79	14.97	111.41	17.58	1.08	0.09
Example 3	122.96	9.73	117.58	8.50	128.34	13.10	1.09	0.09
Example 4	102.00	19.12	96.63	16.13	107.38	22.51	1.11	0.07
Example 5	103.92	24.94	102.71	25.45	105.13	24.48	1.03	0.03
Example 6	101.39	12.53	100.18	12.41	102.59	12.73	1.02	0.02
Example 7	98.66	13.54	97.05	13.87	100.27	13.27	1.04	0.03
Comparative Example 1	119.90	14.55	114.53	16.84	125.27	16.21	1.11	0.17
Comparative Example 2	127.34	30.45	116.92	28.82	137.76	33.88	1.18	0.14
Comparative Example 3	121.06	14.63	118.06	14.19	124.07	15.41	1.05	0.04
Comparative Example 4	166.84	29.50	145.89	37.92	187.79	25.93	1.34	0.29
Comparative Example 5	213.95	77.24	179.02	71.49	248.87	89.85	1.44	0.38
Comparative Example 6	176.93	100.73	140.39	53.71	213.47	151.94	1.45	0.41
Comparative Example 7	133.82	13.88	119.27	19.22	148.37	19.44	1.27	0.26

[Table 10]

	Particles constituting granules (μm)							
	Average particle size (μm)		Average minor axis (μm)		Average major axis (μm)		Ratio of average major axis (μm) with respect to average minor axis (μm)	
	Average value	Standard deviation	Average value	Standard deviation	Average value	Standard deviation	Average value	Standard deviation
Example 1	23.8	5.6	18.8	4.8	28.8	6.6	1.7	0.2
Example 2	15.8	3.2	12.8	1.9	18.7	5.0	1.4	0.2
Example 3	19.6	3.0	14.7	1.9	24.5	4.1	1.7	0.1
Example 4	18.0	2.1	13.9	1.8	22.0	2.6	1.6	0.1
Example 5	19.5	4.9	15.5	4.1	23.4	5.9	1.6	0.2
Example 6	19.1	3.4	15.5	2.8	22.7	4.6	1.5	0.2
Example 7	17.1	3.5	13.7	2.5	20.5	4.6	1.5	0.1
Comparative Example 1	20.4	3.4	10.2	1.6	30.5	5.7	3.2	0.5
Comparative Example 2	29.3	5.2	12.4	2.4	46.2	8.9	4.0	0.6
Comparative Example 3	21.5	3.9	7.6	1.0	35.5	7.1	5.0	0.7
Comparative Example 4	32.5	5.8	12.2	2.0	52.8	10.3	4.7	0.8
Comparative Example 5	55.2	24.1	22.8	12.1	87.6	38.7	4.8	1.5
Comparative Example 6	51.8	17.5	27.6	10.7	76.0	25.6	3.2	0.9
Comparative Example 7	39.0	8.4	15.9	3.1	62.1	15.3	4.2	1.0

[0134] <Evaluation of flowability, jet flow properties, and bulk density>

The “flowability index” and “jet flow properties index” proposed by R.L. Carr were calculated for the granules of Examples and Comparative Examples (Carr, R.L., “Evaluating flow properties of solids.” Chem. Eng. (1965) 72 (163-168)). The angle of repose (°), the spatula angle (°), the compression degree (%), and the degree of uniformity (-) of the granules were determined by using a multifunctional-type powder property measuring instrument (Seishin Enterprise Co., Ltd., Multi-Tester MT-02), and an index corresponding to each measured value was obtained based on Carr’s theory. The flowability index was obtained by summing the respective indices of the measured values. The angle of rupture (°) and the angle of convergence (°) of the granules, and the dispersity (%) were determined with the multifunctional-type powder property measuring instrument, and the index corresponding to each of the measured values obtained based on Carr’s theory and the index based on the flowability index were added to each other to obtain the jet flow property index. The temperature and humidity at the time of measurement were 25°C and 50%.

[0135] The flowability and jet flow properties of the granules were evaluated based on Carr’s evaluation criteria shown below.

Evaluation criteria for flowability

Flowability index value	Degree of flowability
90 to 100	Most satisfactory
80 to 89	Satisfactory

	70 to 79	Fairly satisfactory
	60 to 69	Ordinary
	40 to 59	Not so good
	0 to 19	Very bad
5	[0136] Evaluation criteria for jet flow properties	
	Jet flow property index value	Degree of jet flow properties
	80 to 100	Very strong
	60 to 79	Fairly strong
	40 to 59	Inclined
10	25 to 39	Possibly present
	0 to 24	Absent
15	[0137] The granules obtained by the method of Examples were obtained by a more convenient operation and had satisfactory flowability and jet flow properties similarly to those obtained by the method of the Comparative Examples.	
20	[0138] For the angle of repose and the degree of uniformity, flowability was separately evaluated based on Carr's flowability index table described in the Glossary of Powder Technology (The Society of Powder Technology, Japan, last modified on May 17, 2021, URL: http://www.sptj.jp/powderpedia/words/10382/).	
25	[0139] For the granules of Examples and Comparative Examples, the bulk densities (loose bulk density, tight bulk density, and dynamic bulk density) were determined with a multifunctional-type powder property measuring instrument. The granules obtained by the method of Examples showed a bulk density similar to that of the granules obtained by the method of the Comparative Examples.	

[Table 11]

	Flowability								Jet flow properties					Bulk density		
	Compression degree [%]	Angle of repose [°]		Spatula angle [°]	Degree of uniformity [-]		Flowability		Angle of rupture [°]	Angle of convergence [°]	Dispersion [%]	Jet flow properties		Bulk density		
	Measured value	Measured value	Evaluation	Measured value	Measured value	Evaluation	Total	Evaluation	Measured value	Measured value	Measured value	Total	Evaluation	Loose bulk density	Tight bulk density	Dynamic bulk density
Example 1	13.3	36.3	Fairly satisfactory	48.8	3.3	Most satisfactory	79.5	Good	25.4	10.9	-	-	-	0.45	0.51	0.46
Example 2	24.9	32.0	Satisfactory	48.2	2.5	Most satisfactory	75.0	Slightly good	20.4	11.5	30.9	77.5	Strong	0.45	0.60	0.49
Example 3	18.0	34.2	Satisfactory	54.1	2.7	Most satisfactory	78.0	Slightly good	24.1	10.1	30.4	74	Strong	0.47	0.57	0.49
Example 4	27.4	40.2	Fairly satisfactory	52.6	-	-	-	-	28.3	11.9	30.1	-	-	0.46	0.64	0.51
Example 5	27.9	38.6	Fairly satisfactory	50.5	-	-	-	-	25.4	13.2	28.9	-	-	0.47	0.65	0.52
Example 6	27.9	35.3	Satisfactory	46.1	-	-	-	-	23.4	11.9	33.4	-	-	0.50	0.65	0.54
Example 7	24.3	35.0	Satisfactory	42.6	-	-	-	-	22.4	12.7	32	-	-	0.51	0.67	0.55
Comparative Example 1	20.8	34.4	Satisfactory	47.0	2.0	Most satisfactory	77.0	Slightly good	23.0	11.5	-	-	-	0.51	0.65	0.54
Comparative Example 2	20.1	35.3	Satisfactory	44.3	2.4	Most satisfactory	78.5	Slightly good	23.4	11.9	23.2	74	Strong	0.40	0.50	0.42
Comparative Example 3	22.0	34.8	Satisfactory	47.6	2.4	Most satisfactory	75.0	Slightly good	23.5	11.3	23.2	74	Strong	0.37	0.48	0.40
Comparative Example 4	14.6	35.4	Satisfactory	42.7	-	-	-	-	25.2	10.2	9.4	-	-	0.46	0.54	0.48
Comparative Example 5	11.8	34.3	Satisfactory	36.7	-	-	-	-	24.1	10.2	8.8	-	-	0.50	0.57	0.51
Comparative Example 6	13.2	36.6	Fairly satisfactory	40.3	-	-	-	-	25.1	11.5	8.8	-	-	0.47	0.54	0.47
Comparative Example 7	21.9	33.4	Satisfactory	34.2	-	-	-	-	21.0	12.3	26.8	-	-	0.45	0.58	0.48

[0140] <Evaluation of hygroscopic properties of granules>

5 The hygroscopic properties (adsorption-desorption isotherm curve) of the granules obtained by each method of Example 3 and Comparative Example 3 were measured by using a dynamic water vapor adsorption measuring apparatus (Surface Measurement Systems, Ltd., DVS Intrinsic). 10 mg of a sample was used, and the relative humidity was increased from 0% to 90% at an increment of 5% and then was decreased to 0% by the same procedure. The relative humidity was maintained constant until the rate of change in water content (dm/dt) of the sample was below 0.02 for 10 minutes, or until after 30 minutes even when the rate of change in water content did not reach 0.02%/min, and the temperature was maintained at 25°C at all times. The evaluation results for the hygroscopic properties of the granules obtained by the method of Example 3 are shown in FIG. 4(A).
10 The evaluation results for the hygroscopic properties of the granules obtained by the method of Comparative Example 3 are shown in FIG. 4(B).

[0141] The granules obtained by the method of Examples exhibited an adsorption-desorption isotherm curve similar to that of the granules obtained by the method of the Comparative Examples, and it was confirmed that the hygroscopic properties were equal.

[0142] <Evaluation of average pore diameter, total pore volume, porosity, and total specific surface area of granules>

25 The average pore diameter, the total pore volume, the porosity, and the total specific surface area of the granules obtained by each of the methods of Example 1 and Comparative Example 1 were evaluated

by using a mercury intrusion porosimeter (Quantachrome Corporation, POREMASTER 60GT). 0.4 g of a sample was supplied to the mercury intrusion porosimeter, and measurement was made at 20°C.

Average pore diameter = 50% cumulative pore size

5 Total pore volume = Total amount of intruded mercury (cm³)/sample weight (g)

Porosity = Pore volume of sample (cm³) × sample bulk density (g/cm³)/sample weight (g) × 100

10 Total specific surface area = Specific surface area when the pores are assumed to be cylindrical open systems (calculated from pore distribution and volume)

[Table 12]

	Total pore volume (cm ³ /g)	Total specific surface area (m ² /g)	Average pore diameter (Å)	Porosity (%)
Comparative Example 1	0.0263	1.640	69960	3.73
Example 1	0.0284	8.430	584.5	4.01

15 [0143] The granules obtained by the method of Example 1 showed results that the average pore diameter was smaller, and the total pore volume, the total specific surface area, and the porosity were higher, compared to the granules obtained by the method of Comparative Example 1.

[0144] <Evaluation of aroma retention ratio of granules>

20 The aroma retention ratio of the granules obtained by each of the methods of Examples 4 and 6 and Comparative Examples 4 and 6 was measured by using a portable type odor sensor (New Cosmos Electric Co., Ltd., XP-329IIIR). 50 g of a sample was introduced into an aluminum pack (SEISANNIPPONSHA LTD., LAMIZIP Flat Bag Bottom Opening type AL-G) and hermetically sealed. Furthermore, an

aluminum pack hermetically sealed without introducing a sample was prepared as a zero value setting, and an aluminum pack in which a mixture of 42.4 g of PALATINOSE (Mitsui DM Sugar Co., Ltd., PST-N), 7.5 g of granulated sugar (Mitsui DM Sugar Co., Ltd., GN), and 0.125 g of a muscovado flavoring (TAKASAGO INTERNATIONAL CORPORATION, KOKUTO MICRON H-80210) was introduced and hermetically sealed was prepared as a standard article. Each of the aluminum packs was left to stand for 3 hours at room temperature of 25°C, subsequently the intensity of aroma within the aluminum pack was measured for 5 minutes in the monitoring mode of a portable-type odor sensor, the highest intensity of aroma for 5 minutes was designated as the intensity of aroma of the sample or the standard article, and the aroma retention ratio of each sample was calculated by the following formula.

$$\text{Aroma retention ratio (\%)} = \text{Intensity of aroma of sample} / \text{intensity of aroma of standard article} \times 100$$

Incidentally, the intensity of aroma inside the aluminum pack for zero value setting was designated as intensity zero.

[Table 13]

	Aroma retention ratio (%)
Example 4	131
Comparative Example 4	125
Example 6	120
Comparative Example 6	102

[0145] The granules obtained by the method of Examples exhibited aroma retention ratios equal to or higher than those of the granules obtained by the method of the Comparative Examples having the same sugar content percentage, and it was confirmed that the aroma retention

properties were equal or higher.

CLAIMS

[Claim 1] A method for producing granules, the method comprising:

5 a step of obtaining a crystal suspension comprising at least one raw material selected from the group consisting of a crystalline sugar and a crystalline sugar alcohol, the crystal suspension comprising a portion of the raw material in a crystalline state and being obtained without performing dissolution of the entirety of the raw material and crystallization of the raw material after the dissolution; and

10 a step of spray drying the crystal suspension under low temperature conditions,

wherein the spray drying is performed under conditions with an inlet temperature of 0°C to 60°C.

[Claim 2] The method according to claim 1, wherein the sugar and
15 the sugar alcohol include a monosaccharide, a disaccharide, a trisaccharide, and sugar alcohols of these.

[Claim 3] The method according to claim 1 or 2, wherein the raw material is at least one selected from the group consisting of PALATINOSE, sucrose, and trehalose.

20 [Claim 4] The method according to claim 1 or 2, wherein the crystal suspension further comprises a functional material.

[Claim 5] The method according to claim 4, wherein the functional material is an enzyme, a microorganism, or a flavoring.

[Claim 6] The method according to claim 1 or 2, wherein the
25 crystal suspension has a viscosity of 50 to 1550 mPa·s at 25°C.

[Claim 7] The method according to claim 1 or 2, wherein an

average particle size of the sugar and/or sugar alcohol in a crystalline state in the crystal suspension is 1.00 to 15.90 μm .

5 [Claim 8] The method according to claim 1 or 2, wherein a standard deviation of an average particle size of the sugar and/or sugar alcohol in a crystalline state in the crystal suspension is 1.00 to 10.70 μm .

[Claim 9] The method according to claim 1 or 2, wherein a ratio of an average major axis with respect to an average minor axis of the sugar and/or sugar alcohol in a crystalline state in the crystal suspension is 10 1.00 to 1.70.

[Claim 10] The method according to claim 1 or 2, wherein a ratio of an average major axis with respect to an average minor axis of particles constituting the granule is 3.1 or less.

15 [Claim 11] The method according to claim 1 or 2, wherein the granule has an agglomeration degree of less than 10.0% by mass.

[Claim 12] A granule comprising at least one selected from the group consisting of a crystalline sugar and a crystalline sugar alcohol,
wherein the sugar and/or the sugar alcohol is such that a portion thereof is in a crystalline state while the remaining portion is in a non-crystalline state, and
20 a ratio of an average major axis with respect to an average minor axis of particles constituting the granule is 3.1 or less.

[Claim 13] The granule according to claim 12, wherein the sugar and the sugar alcohol include a monosaccharide, a disaccharide, a trisaccharide, and sugar alcohols of these.
25

[Claim 14] The granule according to claim 12 or 13, wherein at least

one selected from the group consisting of the crystalline sugar and the crystalline sugar alcohol is at least one selected from the group consisting of PALATINOSE, sucrose, and trehalose.

5 [Claim 15] The granule according to claim 12 or 13, further comprising a functional material.

[Claim 16] The granule according to claim 15, wherein the functional material is an enzyme, a microorganism, or a flavoring.

10 [Claim 17] The granule according to claim 12 or 13, wherein an average particle size of particles constituting the granule is 10.00 to 20.00 μm .

[Claim 18] The granule according to claim 12 or 13, wherein a standard deviation of an average particle size of particles constituting the granule is 1.00 to 3.30 μm .

15 [Claim 19] The granule according to claim 12 or 13, wherein the granule has an agglomeration degree of less than 10.0% by mass.