



US012389926B2

(12) **United States Patent**  
**Nakajima et al.**(10) **Patent No.: US 12,389,926 B2**(45) **Date of Patent: Aug. 19, 2025**(54) **SOLID COMPOSITION CONTAINING  
DIMETHYL SULFIDE AND DEXTRIN**(71) Applicant: **SUNTORY HOLDINGS LIMITED,**  
Osaka (JP)(72) Inventors: **Takeshi Nakajima**, Kyoto (JP); **Keita  
Kikuchi**, Kyoto (JP); **Daisaku  
Yonezawa**, Kanagawa (JP); **Yuji  
Hirayama**, Kanagawa (JP); **Taisuke  
Osanai**, Kanagawa (JP)(73) Assignee: **SUNTORY HOLDINGS LIMITED,**  
Osaka (JP)(\*) Notice: Subject to any disclaimer, the term of this  
patent is extended or adjusted under 35  
U.S.C. 154(b) by 654 days.(21) Appl. No.: **17/764,673**(22) PCT Filed: **Sep. 25, 2020**(86) PCT No.: **PCT/JP2020/036279**

§ 371 (c)(1),

(2) Date: **Mar. 29, 2022**(87) PCT Pub. No.: **WO2021/065719**PCT Pub. Date: **Apr. 8, 2021**(65) **Prior Publication Data**

US 2022/0330581 A1 Oct. 20, 2022

(30) **Foreign Application Priority Data**

Sep. 30, 2019 (JP) ..... 2019-180912

(51) **Int. Cl.****A23L 2/60** (2006.01)**A23F 3/30** (2006.01)**A23F 3/40** (2006.01)**A23L 2/39** (2006.01)**A23L 2/56** (2006.01)**A23L 29/00** (2016.01)(52) **U.S. Cl.**CPC ..... **A23L 2/60** (2013.01);**A23F 3/30** (2013.01); **A23F 3/40** (2013.01);**A23L 2/39** (2013.01); **A23L 2/56** (2013.01);**A23L 29/055** (2016.08)(58) **Field of Classification Search**CPC ..... **A23L 2/60**; **A23L 2/39**; **A23L 2/56**; **A23L  
2/00**; **A23L 29/30**; **A23L 29/055**; **A23L  
27/2022**; **A23L 27/203**; **A23F 3/30**; **A23F  
3/40**; **A23F 3/405**

USPC ..... 426/590, 535, 597, 594, 596

See application file for complete search history.

(56) **References Cited**

U.S. PATENT DOCUMENTS

4,474,822 A 10/1984 Sato et al.  
5,538,750 A \* 7/1996 Yamaguchi ..... A23L 3/44  
426/597

6,299,925 B1 \* 10/2001 Xiong ..... A61K 36/746

426/477

2007/0009640 A1 \* 1/2007 Hiramoto ..... A23F 3/163

426/597

2009/0029020 A1 \* 1/2009 Strassburger ..... C08B 37/0015

426/302

2009/0155446 A1 6/2009 Reiss et al.

2009/0214446 A1 \* 8/2009 Strassburger ..... A23L 2/06

424/49

2010/0209585 A1 8/2010 Fukuda et al.

2010/0330243 A1 12/2010 Kikuchi et al.

2013/0101697 A1 4/2013 Shimada et al.

2014/0193562 A1 \* 7/2014 Popplewell ..... A23L 27/70

426/534

2014/0326166 A1 \* 11/2014 Mesnager ..... C09D 103/02

106/205.01

2016/0242430 A1 \* 8/2016 Daenzer-Alloncle ..... A23F 5/26

2019/0230951 A1 8/2019 Inoue et al.

## FOREIGN PATENT DOCUMENTS

AU 2015365199 A1 3/2017  
CN 104431092 A 3/2015  
EP 3985087 A1 4/2022  
GB 2074838 A \* 11/1981 ..... A23F 3/14  
JP S57-33545 A 2/1982  
JP 60-210949 A 10/1985  
JP 1-181781 A 7/1989  
JP 3-35898 B2 5/1991  
JP 3-36491 B2 5/1991  
JP 5-304890 A 11/1993  
JP 6-296457 A 10/1994  
JP H09-118626 A 5/1997  
JP 10-179032 A 7/1998  
JP 2005-143467 A 6/2005

(Continued)

## OTHER PUBLICATIONS

Extended (Supplementary) European Search Report dated Sep. 5,  
2023, issued in counterpart EP Application No. 20873292.5. (8  
pages).Tan et al., "Characterisation of key odourants in Japanese green tea  
using gas chromatography-olfactometry and gas chromatography-  
mass spectrometry", LWT-Food Science and Technology, Mar. 22,  
2019, vol. 108, pp. 221-232, table 1, cited in ISR (12 pages).International Search Report dated Nov. 10, 2020, issued in coun-  
terpart International Application No. PCT/JP2020/036279 (3 pages).Narin, N., et al., "Volatile Compounds in Tomato-Based Dried  
Products", Drying Technology, 2010, vol. 28, No. 2, pp. 232-239,  
cited in ISR dated Nov. 10, 2020. (9 pages).International Search Report dated Nov. 10, 2020, issued in Inter-  
national application No. PCT/JP2020/036271 (counterpart to U.S.  
Appl. No. 17/763,038). (3 pages).

(Continued)

Primary Examiner — Erik Kashnikow

Assistant Examiner — Luis Eugenio Diou Berdecia

(74) Attorney, Agent, or Firm — WHDA, LLP

(57)

**ABSTRACT**A solid composition comprises dimethyl sulfide and dextrin.  
The solid composition includes dimethyl sulfide in an  
amount of at least 2.1 µg per gram of the solid composition  
(2.1 µg/g). The dextrin present in the composition has a  
molecular weight of 50,000 or more. The solid composition  
comprises linear dextrin and cyclic dextrin, wherein the  
cyclic dextrin is one or more cyclodextrin(s) selected from  
the group consisting of α-cyclodextrin and γ-cyclodextrin.**13 Claims, No Drawings**

(56)

## References Cited

## FOREIGN PATENT DOCUMENTS

JP	2007-110990	A	5/2007	
JP	2007-289115	A	11/2007	
JP	2008-092817	A	4/2008	
JP	2008148588	A *	7/2008	
JP	2009-17867	A	1/2009	
JP	2009-72188	A	4/2009	
JP	2010207116	A	9/2010	
JP	2010-233559	A	10/2010	
JP	2012-115247	A	6/2012	
JP	2013-153739	A	8/2013	
JP	2013-226111	A	11/2013	
JP	2016-111970	A	6/2016	
JP	2017079668	A	5/2017	
JP	2019-097568	A	6/2019	
JP	2021017415	A	2/2021	
WO	2005/039301	A1	5/2005	
WO	WO-2011047611	A1 *	4/2011	..... A23F 3/18
WO	2015/052161	A1	4/2015	
WO	2015/167221	A1	11/2015	
WO	2016/098388	A1	6/2016	
WO	2017/033850	A1	3/2017	
WO	2018/066613	A1	4/2018	
WO	2020222296	A1	11/2020	

## OTHER PUBLICATIONS

Zhu et al., Determination of Volatile Chemical Constitutes in Tea by Simultaneous Distillation Extraction, Vacuum Hydrodistillation and Thermal Desorption, *Chromatographia*, vol. 68, No. 7-8, 2008, p. 603-610; cited in Extended European Search Report dated Sep. 4, 2023.

Extended (Supplementary) European Search Report dated Sep. 4, 2023, issued in EP Application No. 20872848.5 (counterpart to U.S. Appl. No. 17/763,038). (9 pages).

Office Action dated Feb. 9, 2024, issued in SG Application No. 11202202562 (counterpart to U.S. Appl. No. 17/763,038). (9 pages).

Office Action dated Jun. 17, 2024, issued in JP Application No. 2021-551179 (counterpart to U.S. Appl. No. 17/763,038), with English translation. (8 pages).

Zhou, Ru-juan, et al., "Research of Aroma Components of Green Tea", *The Food Industry*, 2015, vol. 36, No. 12, pp. 270-274; w/English Abstract; Cited in ISR.

Ogita, Hirofumi, "Investigation of High-quality Matcha Flavor and its Application to Flavor Creation. Koryo.", Mar. 20, 2021, No. 289, pp. 103-107; w/English Abstract; Cited in ISR.

Kim, Youngmok, et al., "Youngmok et al. Effects of Different Harvesting Times and Oxidative Fermentation Methods on Phytochemicals. Flavors. and Sensory Properties of Korean Teas." ACS Symposium Series 1303 Chemistry of Korean Foods, and Beverages, 2019, pp. 77-95; Cited in ISR.

English Translation of International Search Report dated May 10, 2022, issued in Application PCT/JP2022/010280 (counterpart to U.S. Appl. No. 18/549,954). (3 pages).

Non-Final Office Action dated Aug. 29, 2024, issued in U.S. Appl. No. 17/763,038. (7 pages).

International Search Report dated May 11, 2021, issued in Application No. PCT/JP2021/006562 (counterpart to U.S. Appl. No. 17/916,934). (3 pages).

Kawakami, Michiko, "Chapter IV Aroma of shade grown tea, Tea aroma research notes from a diversity perspective in manufacturing", 2000, pp. 49-66, in particular, pp. 63-64; w/ Partial translation; Cited in ISR. (24 pages).

Baba, Ryoko, "Characterization and application to the flavor development of the potent odorants contributing to the characteristic aroma of traditional teas made from *Camellia sinensis* var. *sinensis*." Doctoral Theses of Tokyo University of Agriculture, Dissertation Ph.D. B No. 936; Cited in ISR. (37 pages).

Kawakami, Michiko et al., "Aroma Characteristics of *Kabusecha* (Shaded Green Tea)", 1981, 55(2), pp. 117-123; Cited in ISR. (7 pages).

Hattori, Shoji, "Study of New Approach for Evaluation of Japanese Green tea Using Oasis Method", 2006, 28(1), pp. 85-120; Cited in ISR. (37 pages).

Yamaguchi, K. et al., "Volatile constituents of green tea, Gyokuro (*Camellia sinensis* L. var; *Yabukita*)", *J. Agric. Food Chem.*, 1981, 29(2), pp. 366-370; Cited in ISR. (5 pages).

Office Action dated Jul. 12, 2023, issued in CN Application No. 202180026073.6 (counterpart to U.S. Appl. No. 17/916,934), with English translation. (14 pages).

Wang Mengqi et al., *Food Science*, 2019, vol. 40, No. 22, pp. 219-228; cited in CN Office Action dated Jul. 12, 2023.

Yang Chun et al., *Guizhou Agricultural Sciences*, 2017, vol. 45, No. 1, pp. 122-126; cited in CN Office Action dated Jul. 12, 2023.

Extended (Supplementary) European Search Report dated Apr. 12, 2024, issued in EP application No. 21784981.9 (counterpart to U.S. Appl. No. 17/916,934). (11 pages).

Non-Final Office Action dated Nov. 6, 2024, issued in U.S. Appl. No. 17/916,934. (18 pages).

A. Tiper, "An Introduction to headspace sampling in gas chromatography Fundamentals and Theory", *Perkin Elmer, Inc.*, 2014, pp. 1-35. (35 pages).

Flaig, et al., Characterization of the Key Odorants in a High-Grade Chinese Green Tea Beverage (*Camellia sinensis*; *Jingshan cha*) by Means of the Sensomics Approach and Elucidation of Odorant Changes in Tea Leaves Caused by the Tea Manufacturing Process, *Journal of Agricultural and Food Chemistry*, 2020, vol. 68, pp. 5168-5179. (13 pages) cited in JP Office Action dated Dec. 9, 2024.

Office Action dated Dec. 9, 2024 issued in counterpart JP application No. 2021-060539 with English translation. (6 pages).

Flaig et al., Characterisation of the key aroma compounds in a Longjing green tea infusion (*Camellia sinensis*) by the sensomics approach and their quantitative changes during processing of the tea leaves, *European Food Research and Technology*, 2020, vol. 246, No. 12, p. 2411-2425. cited in EESR dated Feb. 17, 2025 (15 pages).

Extended (Supplementary) European Search Report dated Feb. 17, 2025, issued in counterpart application No. 22779885.7. (7 pages).

Non-Final Office action dated Jun. 18, 2025 issued in U.S. Appl. No. 17/763,038 (32 pages).

\* cited by examiner

1

**SOLID COMPOSITION CONTAINING  
DIMETHYL SULFIDE AND DEXTRIN****TECHNICAL FIELD**

The present invention relates to a solid composition containing dimethyl sulfide and dextrin.

**BACKGROUND ART**

As a typical powdering method for obtaining instant tea, a spray drying method suitable for continuous mass production has been adopted. Spray drying is a method in which fine droplets are ejected into hot air to instantaneously evaporate water, and is a technique in which the residence time exposed to high temperature is short, and the heat quantity given is consumed as latent heat of evaporation, so that the thermal load on the content liquid is suppressed and powder with little deterioration in quality can be obtained. The higher the concentration of the content liquid subjected to spray drying, the higher the retention rate of the aroma component contained in the liquid after spray drying. This can be explained by the selective diffusion theory; in the droplets during drying, when the solute concentration is low, the diffusion coefficient of the aroma component is low, and the aroma component volatilizes simultaneously with the evaporation of water. However, the higher the solute concentration, the higher the diffusion coefficient of the aroma component, and the scent remains due to water evaporating earlier than the migration of the aroma component. Therefore, in order to attain instant tea of high quality, it is important to produce a tea concentrate containing a high concentration of the aroma component.

Methods for concentrating a tea extract, from the viewpoint of the principle of whether water is separated in a solid, liquid, or gas phase, can be classified into three types: freeze concentration (aqueous phase-solid); membrane concentration (aqueous phase-liquid); and evaporation concentration (aqueous phase gas). Among them, in the evaporation concentration, a thermal load applied to a content liquid is large, and thus, it is difficult to obtain a concentrate retaining the aroma component, which is prone to thermal degradation like tea. Furthermore, as for the freeze concentration, in addition to the fact that the concentration tune and cost are large, which is a high hurdle for its practical use, it is difficult to increase the concentration in the first place. For this reason, concentration by any of these schemes has not been often adopted. On the other hand, the membrane concentration is a scheme in which the solute concentration is increased by applying pressure to water as it is through a membrane with fine pores, eliminating evaporation and/or freezing, and thus allowing concentration at low cost without quality change.

Furthermore, as a means for producing a tea concentrate, it is known to add dextrin, which is a kind of excipient and has a molecular structure with sugars linked in a chain, to a tea extract. For example, it has been reported that when acyclic dextrin or cyclic dextrin having an average degree of polymerization of 4 to 10 is added to an extract of tea leaves, roasted grains, or roasted beans before concentration and then subjected to membrane concentration at 40° C., the extract can be concentrated without reducing the concentration efficiency (PTL 1).

Furthermore, it is known that a specific dextrin improves the solubility of instant tea after spray drying. For example, it has been reported that instant tea having excellent flavor and immediate solubility can be produced by spray drying of

2

tea which contains dextrin having an average degree of polymerization of 4 to 10 alone or in combination with cyclic dextrin and which has carbon dioxide dissolved therein (PTL 2). Furthermore, it has been reported that by spray drying of a tea extract with indigestible dextrin added thereto, instant tea excellent in flavor and solubility can be provided (PTL 3).

**CITATION LIST****Patent Literature**

[PTL 1] JP H3-36491 B2  
[PTL 2] JP H3-35898 B2  
[PTL 3] JP 2009-17867 A

**SUMMARY OF INVENTION****Technical Problem**

As a characteristic aroma of so-called high-grade green tea among the tea beverages, there is a so-called green layer scent, and it is known that dimethyl sulfide and the like have the scent of green laver. However, since dimethyl sulfide is prone to volatilizing, in the production of a solid composition containing dimethyl sulfide, dimethyl sulfide has not always been sufficiently retained at a high concentration in the solid composition. Accordingly, an object of the present invention is to provide a solid composition which is excellent in capability of retaining dimethyl sulfide and which has a green layer scent strongly felt when made into a beverage using water, hot water, or the like.

**Solution to Problem**

As a result of intensive studies to solve the above problem, the present inventors focused on dextrin in the production of a solid composition containing dimethyl sulfide, and have found that by incorporating a polymer dextrin into the solid composition, dimethyl sulfide can be retained at a high concentration in the solid composition. Based on this finding, the present inventors have completed the present invention.

The present invention relates to, but is not limited to, the following:

(1) A solid composition containing dimethyl sulfide and dextrin,

wherein a content of dimethyl sulfide in the composition is 2.1 µg or more per gram of the composition (2.1 µg/g), and

the composition contains dextrin having a molecular weight of 50,000 or more.

(2) The composition according to (1), wherein a proportion of the dextrin having a molecular weight of 50,000 or more in molecular weight distribution of the composition is 5 to 35%.

(3) The composition according to (1) or (2), wherein a proportion of the dextrin having a molecular weight of 50,000 to 200,000 in molecular weight distribution of the composition is 2 to 30%.

(4) The composition according to any one of (1) to (3), wherein a proportion of dextrin having a molecular weight of from 200,000 to 350,000 in molecular weight distribution of the composition is 0.3 to 6%.

(5) The composition according to any one of (1) to (4), wherein a proportion of dextrin having a molecular weight of 250,000 or more in molecular weight distribution of the composition is 0.9 to 10%.

(6) The composition according to any one of (1) to (5), wherein a proportion of dextrin having a molecular weight of 350,000 or more in molecular weight distribution of the composition is 0.2 to 4%.

(7) The composition according to any one of (1) to (6), wherein the composition contains two or more kinds of dextrin.

(8) The composition according to any one of (1) to (7), wherein the composition contains linear dextrin and cyclic dextrin.

(9) The composition according to any one of (1) to (8), further containing one or more aroma components selected from the group consisting of  $\beta$ -ionone,  $\alpha$ -ionone,  $\beta$ -cyclocitral, (z)-3-hexenol, 1-penten-3-ol, hexanal, and nerolidol.

(10) The composition according to any one of (1) to (9), wherein the composition is a powder composition.

(11) The composition according to any one of (1) to (10), wherein the composition contains a tea leaf extract.

(12) The composition according to (11), wherein the tea leaf extract is a tea leaf extract of sencha.

(13) A food or drink containing the composition according to any one of (1) to (12).

(14) The food or drink according to (13), wherein the food or drink is a beverage.

(15) The food or drink according to (14), wherein the food or drink is a tea beverage.

(16) A food or drink containing dimethyl sulfide and dextrin,

wherein a content of dimethyl sulfide in the food or drink is 20.3 ppb (w/w) or more per 1.0% solid content (Brix), and

the food or drink contains dextrin having a molecular weight of 50,000 or more.

#### Advantageous Effects of Invention

According to the present invention, it is possible to provide a solid composition capable of retaining dimethyl sulfide at a high concentration. In the present invention, dimethyl sulfide contained in a solution before drying in a production process can be retained at a high concentration in a solid composition from the drying. For this reason, through the use of the present invention, a solid composition abundantly containing dimethyl sulfide can be provided.

The solid composition of the present invention can be made into a tea beverage using water or hot water, and can provide a strong green layer scent when drinking. Furthermore, the solid composition of the present invention is much lighter than a beverage, and thus is excellent in convenience during transportation.

Furthermore, the solid composition of the present invention can also be used as a raw material for food. The number and types of foods having tea flavor have been increasing in recent years. The solid composition of the present invention can be used to impart a green layer scent to confectioneries such as cakes, castella, candies, cookies, jellies, puddings, and chocolates.

#### DESCRIPTION OF EMBODIMENTS

##### 1. Solid Composition

One embodiment of the present invention is a solid composition containing dimethyl sulfide and dextrin, a content of dimethyl sulfide in the composition being 2.1  $\mu\text{g}$  or more per gram of the composition (2.1  $\mu\text{g/g}$ ), and the composition containing dextrin having a molecular weight of 50,000 or more. By adopting such a configuration, the

content of dimethyl sulfide in the solid composition can be maintained at a high level, and it is possible to provide a solid composition that has a green layer scent strongly felt when made into a beverage using water, hot water, or the like. The term "green layer scent" as used herein refers to a scent with a roasting aroma, a sweet flavor, and a shore-like smell mixed.

##### 1-1. Dimethyl Sulfide

The solid composition of the present invention contains dimethyl sulfide. Dimethyl sulfide is an organic sulfur compound represented by the composition formula of  $(\text{CH}_3)_2\text{S}$ , and is known as an aroma component exhibiting a green layer scent.

A content of dimethyl sulfide in the solid composition of the present invention is not particularly limited, and is, for example, 2.1  $\mu\text{g}$  or more per gram of the solid composition (2.1  $\mu\text{g/g}$ ), preferably 2.2  $\mu\text{g/g}$  or more, more preferably 2.3  $\mu\text{g/g}$  or more, even more preferably 2.4  $\mu\text{g/g}$  or more, and particularly 2.5  $\mu\text{g/g}$  or more. When the content of dimethyl sulfide is within the above range, a green layer scent will be exhibited. The content of dimethyl sulfide in the solid composition of the present invention does not have a particular upper limit, and is, for example, 50,000  $\mu\text{g}$  or less per gram of the solid composition (50,000  $\mu\text{g/g}$ ), preferably 40,000  $\mu\text{g/g}$  or less, more preferably 25,000  $\mu\text{g/g}$  or less, even more preferably 5,000  $\mu\text{g/g}$  or less, and particularly preferably 2,500  $\mu\text{g/g}$  or less. The content of dimethyl sulfide in the solid composition of the present invention is typically 2.1 to 50,000  $\mu\text{g}$  per gram of the solid composition (2.1 to 50,000  $\mu\text{g/g}$ , preferably 2.2 to 40,000  $\mu\text{g/g}$ , more preferably 2.3 to 25,000  $\mu\text{g/g}$ , even more preferably 2.4 to 5,000  $\mu\text{g/g}$ , and particularly preferably 2.5 to 2,500  $\mu\text{g/g}$ ).

In the present invention, the content of dimethyl sulfide in the solid composition can be measured using gas chromatography (GC). Furthermore, examples of an analyzer therefor include Flash GC Nose HERACLES II (Alpha M.O.S. Japan K.K.). Specifically, a solution is prepared by dissolving the solid composition of the present invention in water, and the contents of dimethyl sulfide can be measured using the solution under the following conditions.

Gas chromatography apparatus: Flash GC Nose HERACLES II

Column 1: MXT-5 (low polarity 10 m, 180  $\mu\text{m}$  ID, 0.4  $\mu\text{m}$ )

Column 2: MXT-WAX (high polarity 10 m, 180  $\mu\text{m}$  ID, 0.4  $\mu\text{m}$ )

Carrier-gas flow rate: hydrogen 1.6 mL/min

Flame ionization detector (FID) temperature: 260° C.

Injector temperature: 200° C.

Oven temperature: 40° C. (5 seconds) to 1.5° C./second to 250° C. (90 seconds)

Injection time: 125 seconds

Trapping temperature: adsorption 50° C., desorption 240° C.

Trapping time: adsorption 130 seconds, preheating 35 seconds

Conditions relating to measurement samples can be set as described in examples below. After the contents of dimethyl sulfide in the solution are measured, the contents of dimethyl sulfide in the solid composition can be obtained by back calculation from the amount of the solid composition dissolved in water.

In the solid composition of the present invention, dimethyl sulfide may be used in a purified or crude form, or an extract containing dimethyl sulfide may be used. For the extract containing dimethyl sulfide, a naturally-derived extract can be used, and both a plant extract and an animal

extract can be used. In the present invention, it is preferable to use a plant extract; and among plants, a tea leaf extract is more preferably used. Here, although not particularly limited, the tea leaf extract containing dimethyl sulfide is preferably a tea leaf extract of a type different from the tea leaf extract described below. That is, when the tea leaf extract described in the section "1-4. Tea Leaf Extract" described below is used as a first tea leaf extract, the tea leaf extract containing dimethyl sulfide can be used as a second tea leaf extract different from the first tea leaf extract.

For raw material tea leaves for the tea leaf extract containing dimethyl sulfide, non-fermented tea is preferably used, and specific examples thereof include green teas such as aracha, sencha, gyokuro, kabusecha, tencha, bancha, and houjicha. The tea leaf extract containing dimethyl sulfide may be extracted from only one kind of tea leaves or may be extracted from a blend of a plurality of kinds of tea leaves. In the present invention, the tea leaf extract containing dimethyl sulfide is preferably a tea leaf extract of green tea, more preferably an extract of tea leaves that have been covered and cultivated with sunlight shielded before plucking, such as kabusecha, gyokuro, or tencha. By using the extract of tea leaves that have been covered and cultivated with sunlight shielded before plucking them, such as kabusecha, gyokuro, or tencha, aroma components other than dimethyl sulfide derived from these tea leaves can be contained, which can impart, to the solid composition, a flavor that is superior as befits high-class tea.

When the tea leaf extract containing dimethyl sulfide is used, the content thereof is not particularly limited, and can be adjusted such that the content of dimethyl sulfide in the solid composition of the present invention will fall within the range indicated above.

#### 1-2. Dextrin

The solid composition of the present invention contains dextrin. Dextrin is a generic name for carbohydrates obtained by hydrolysis of starch or glycogen. In the present invention, the dextrin can be used as an excipient for forming the solid composition, and molecular weight distribution in the solid composition of the present invention can be adjusted according to the weight average molecular weight of the dextrin used and the content thereof in the composition.

A content of the dextrin in the solid composition of the present invention is not particularly limited, and is, for example, 10 to 70 wt %, preferably 20 to 65 wt %, and more preferably 30 to 60 wt %. For the dextrin in the present invention, commercially available products can be used. The content of the dextrin in the solid composition can be measured by performing sugar analysis using a method known to those skilled in the art.

A high-molecular-weight dextrin as used herein refers to dextrin having a molecular weight of 10,000 or more, and a low-molecular-weight dextrin refers to dextrin having a molecular weight of less than 10,000.

The solid composition of the present invention contains dextrin having a molecular weight of 50,000 or more. The presence of such dextrin as having a high molecular weight makes it possible to retain dimethyl sulfide at a high concentration in the solid composition.

On the other hand, when the amount of the dextrin having the high molecular weight is too large, for example, when a proportion of dextrin having a molecular weight of 50,000 or more exceeds 45%, the viscosity of the composition before solidification will be too high to perform a drying treatment, and it tends to be difficult to obtain the solid composition. In the molecular weight distribution of the

solid composition of the present invention, the proportion of the dextrin having a molecular weight of 50,000 or more is preferably 5 to 35%, more preferably 5.5 to 30%, even more preferably 6.0 to 25%, and particularly preferably 6.5 to 23%.

In the present invention, the distribution of molecular weights in the solid composition can be determined by using a gel permeation chromatography (GPC) analysis method. Specifically, the distribution of molecular weights in the solid composition can be examined under the following conditions.

Apparatus: Sampling injector: 231 XL (GILSON)

Pump: 305 (GILSON)

Column oven: CTO 10AS VP (Shimadzu Corporation)

Detector: RID-10A (Shimadzu Corporation)

Columns: connected in series in the following order

[Pump]→TSKgel Guard Column PWx1 (6.0 mm I.D.×4 cm) (TOSOH CORPORATION)→TSKgel G4000PWx1 (particle size 10 μm, 7.8 mm I.D.×30 cm) (TOSOH CORPORATION)→TSKgel G3000PWx1 (particle size 7 μm, 7.8 mm I.D.×30 cm) (TOSOH CORPORATION)→[Detector]

Analytical Data System: LabSolutions (Shimadzu Corporation)

Flow rate: 1 mL/min

Injection volume: 50 μL

Mobile phases: 0.1 mol/L sodium nitrate solution

Column temperature: 50° C.

An analytical sample can be prepared as described in the examples below. Proportions of components having various molecular weights in molecular weight distribution can also be determined as described in the examples below and can be determined specifically by calculating the proportion of a target peak area to a total value of obtained peak areas (total peak area).

In the molecular weight distribution of the solid composition of the present invention, a proportion of dextrin having a molecular weight of 100,000 or more is not particularly limited, and is, for example, 2.0 to 25%, preferably 3.0 to 20%, and more preferably 4.0 to 15%. Furthermore, in the molecular weight distribution of the solid composition of the present invention, the proportion of dextrin having a molecular weight of 150,000 or more is not particularly limited, and is, for example, 1.0 to 20%, preferably 2.0 to 15%, and more preferably 2.5 to 9.0%. Furthermore, in the molecular weight distribution of the solid composition of the present invention, the proportion of dextrin having a molecular weight of 200,000 or more is not particularly limited, and is, for example, 0.5 to 12%, preferably 1.0 to 9.0%, and more preferably 1.5 to 6.0%. Furthermore, in the molecular weight distribution of the solid composition of the present invention, the proportion of dextrin having a molecular weight of 250,000 or more is not particularly limited, and is, for example, 0.9 to 10%, preferably 1.00 to 5.0%, and more preferably 1.10 to 3.5%. Furthermore, in the molecular weight distribution of the solid composition of the present invention, the proportion of dextrin having a molecular weight of 300,000 or more is not particularly limited, and is, for example, 0.40 to 8.0%, preferably 0.50 to 5.0%, and more preferably 0.60 to 2.0%. Furthermore, in the molecular weight distribution of the solid composition of the present invention, the proportion of dextrin having a molecular weight of 350,000 or more is not particularly limited, and is, for example, 0.2 to 4%, preferably 0.30 to 2.5%, and more preferably 0.40 to 1.5%. Furthermore, in the molecular weight distribution of the solid composition of the present invention, the proportion of

dextrin having a molecular weight of 400,000 or more is not particularly limited, and is, for example, 0.10 to 3.0%, preferably 0.20 to 2.0%, and more preferably 0.30 to 1.0%. Furthermore, in the molecular weight distribution of the solid composition of the present invention, the proportion of dextrin having a molecular weight of 450,000 or more is not particularly limited, and is, for example, 0.05 to 1.5%, preferably 0.10 to 1.0%, and more preferably 0.20 to 0.6%. Furthermore, in the molecular weight distribution of the solid composition of the present invention, the proportion of dextrin having a molecular weight of 500,000 or more is not particularly limited, and is, for example, 0.05 to 1.2%, preferably 0.08 to 0.8%, and more preferably 0.10 to 0.5%.

Furthermore, in the molecular weight distribution of the solid composition of the present invention, the proportion of dextrin having a molecular weight of 50,000 or more and less than 200,000 is not particularly limited, and is, for example, 2 to 30%, preferably 3.5 to 25%, and more preferably 4.5 to 18%. Furthermore, in the molecular weight distribution of the solid composition of the present invention, the proportion of dextrin having a molecular weight of 200,000 or more and less than 350,000 is not particularly limited, and is, for example, 0.3 to 6.0%, preferably 0.50 to 5.0%, and more preferably 0.60 to 4.0%.

In the molecular weight distribution of the solid composition of the present invention, a proportion of a component having a molecular weight of less than 50,000 is not particularly limited. In the molecular weight distribution of the solid composition of the present invention, a proportion of a component having a molecular weight of less than 3,000 is, for example, 35% or more, preferably 40% or more, and more preferably 45% or more. Furthermore, in the molecular weight distribution of the solid composition of the present invention, a proportion of a component having a molecular weight of 3,000 or more and less than 10,000 is, for example, 5.0% or more, preferably 8.0% or more, and more preferably 10% or more. Furthermore, in the molecular weight distribution of the solid composition of the present invention, a proportion of a component having a molecular weight of 10,000 or more and less than 50,000 is, for example, 2.0 to 30%, preferably 4.0 to 25%, and more preferably 6.0 to 20%.

A further constituent proportion of dextrin having a molecular weight of 50,000 or more in the solid composition of the present invention is not particularly limited; and in a molecular weight range of 50,000 or more and less than 500,000, it is preferable that the proportion decreases as the molecular weight increases. That is, preferably, the proportion of dextrin having a molecular weight of 100,000 or more and less than 150,000 is smaller than the proportion of dextrin having a molecular weight of 50,000 or more and less than 100,000, the proportion of dextrin having a molecular weight of 150,000 or more and less than 200,000 is smaller than the proportion of dextrin having a molecular weight of 100,000 or more and less than 150,000, the proportion of dextrin having a molecular weight of 200,000 or more and less than 250,000 is smaller than the proportion of dextrin having a molecular weight of 150,000 or more and less than 200,000, the proportion of dextrin having a molecular weight of 250,000 or more and less than 300,000 is smaller than the proportion of dextrin having a molecular weight of 200,000 or more and less than 250,000, the proportion of dextrin having a molecular weight of 300,000 or more and less than 350,000 is smaller than the proportion of dextrin having a molecular weight of 250,000 or more and less than 300,000, the proportion of dextrin having a molecular weight of 350,000 or more and less than 400,000

is smaller than the proportion of dextrin having a molecular weight of 300,000 or more and less than 350,000, the proportion of dextrin having a molecular weight of 400,000 or more and less than 450,000 is smaller than the proportion of dextrin having a molecular weight of 350,000 or more and less than 400,000, the proportion of dextrin having a molecular weight of 450,000 or more and less than 500,000 is smaller than the proportion of dextrin having a molecular weight of 400,000 or more and less than 450,000.

As described above, in the present invention, the molecular weight distribution in the solid composition can be adjusted according to the type of dextrin used, and the like. The dextrin used in the present invention is not particularly limited, and for example, linear dextrin or cyclic dextrin can be used. The term "linear dextrin" as used herein refers to a dextrin in which glucose molecules are bonded linearly or are bonded in a chain as having a branched chain and do not form a cyclic structure or a helical structure. Furthermore, as used herein, the term "cyclic dextrin" refers to a dextrin in which glucose molecules are bonded to form a ring structure and do not form a helical structure.

The chain dextrin is not particularly limited, and for example, linear dextrin having a dextrose equivalent (DE) of 1 to 25, linear dextrin having a weight average molecular weight of 500 to 160,000, or the like can be used. Furthermore, in the present invention, not only one kind of linear dextrin but also two or more kinds of linear dextrin may be used in combination. A preferred embodiment in the present invention is the use of two kinds of linear dextrin. When two kinds of linear dextrin are used, for example, a combination of a linear dextrin having a DE of 2 to 5 and a linear dextrin having a DE of 16 to 20 or a combination of a linear dextrin having a weight average molecular weight of 90,000 to 140,000 and a linear dextrin having a weight average molecular weight of 600 to 1,200 can be used.

When linear dextrin is used, the content of the linear dextrin in the solid composition of the present invention is, for example, 0 to 65 wt %, preferably 10 to 60 wt %, and more preferably 15 to 55 wt %. When the linear dextrin having a DE of 2 to 5 and the linear dextrin having a DE of 16 to 20 are used as the two types of linear dextrin, a content of the linear dextrin having a DE of 2 to 5 in the solid composition of the present invention is, for example, 0 to 60 wt %, preferably 5 to 50 wt %, and more preferably 10 to 45 wt %, and a content of the linear dextrin having a DE of 16 to 20 is, for example, 0 to 60 wt %, preferably 5 to 50 wt %, and more preferably 10 to 45 wt %. Furthermore, a content ratio (weight ratio) of the linear dextrin having a DE of 2 to 5 to the linear dextrin having a DE of 16 to 20 is, for example, 4:0.5 to 0.5:5, preferably 3:1 to 1:5, and more preferably 2:1 to 1:4.

Furthermore, when the linear dextrin having a weight average molecular weight of 90,000 to 140,000 and the linear dextrin having a weight average molecular weight of 600 to 1,200 are used as the two types of linear dextrin, a content of the linear dextrin having a weight average molecular weight of 90,000 to 140,000 in the solid composition of the present invention is, for example, 0 to 65 wt %, preferably 10 to 60 wt %, more preferably 15 to 55 wt %, and a content of the linear dextrin having a weight average molecular weight of 600 to 1,200 is, for example, 5 to 60 wt %, preferably 5 to 50 wt %, more preferably 10 to 45 wt %. Furthermore, a content ratio (weight ratio) of the linear dextrin having a weight average molecular weight of 90,000 to 140,000 to the linear dextrin having a weight average molecular weight of 600 to 1,200 is, for example, 5:1 to 1:3, preferably 3:1 to 1:2, and more preferably 2:1 to 1:1.

As the cyclic dextrin, for example, cyclodextrin can be used. In the present invention, any of  $\alpha$ -cyclodextrin,  $\beta$ -cyclodextrin, and  $\gamma$ -cyclodextrin can be used, and  $\alpha$ -cyclodextrin is preferably used. A weight average molecular weight of the cyclic dextrin used in the present invention is not particularly limited, and is, for example, 700 to 1,300, preferably 800 to 1,200, and more preferably 900 to 1,100. When the cyclic dextrin is used, a content of the cyclic dextrin in the solid composition of the present invention is, for example, 0.5 to 15 wt %, preferably 1 to 12 wt %, and more preferably 3 to 10 wt %.

When the linear dextrin and the cyclic dextrin are used, a content ratio (weight ratio) of the linear dextrin to the cyclic dextrin is, for example, 20:1 to 2:1, preferably 15:1 to 3:1, and more preferably 12:1 to 5:1.

### 1-3. Other Aroma Components

In addition to dimethyl sulfide described above, the solid composition of the present invention may further contain one or more aroma components selected from the group consisting of  $\beta$ -ionone,  $\alpha$ -ionone,  $\beta$ -cyclocitral, (z)-3-hexenol, 1-penten-3-ol, hexanal, and nerolidol. The solid composition of the present invention containing these aroma components will exhibit a more well-balanced green layer scent.

A content of  $\beta$ -ionone in the solid composition of the present invention is not particularly limited, and is, for example, 0.31 to 5,000  $\mu\text{g}$  per gram of the solid composition (0.31 to 5,000  $\mu\text{g/g}$ ), preferably 0.5 to 3,500  $\mu\text{g/g}$ , more preferably 0.65 to 2,000  $\mu\text{g/g}$ , even more preferably 0.8 to 1,500  $\mu\text{g/g}$ , and particularly preferably 1 to 1,000  $\mu\text{g/g}$ .

A content of  $\alpha$ -ionone in the solid composition of the present invention is not particularly limited, and is, for example, 0.01 to 200  $\mu\text{g}$  per gram of the solid composition (0.1 to 200  $\mu\text{g/g}$ ), preferably 0.025 to 150  $\mu\text{g/g}$ , more preferably 0.05 to 100  $\mu\text{g/g}$ , even more preferably 0.075 to 75  $\mu\text{g/g}$ , and particularly preferably 0.1 to 50  $\mu\text{g/g}$ .

A content of the  $\beta$ -cyclocitral in the solid composition of the present invention is not particularly limited, and is, for example, 0.01 to 200  $\mu\text{g}$  per gram of the solid composition (0.1 to 200  $\mu\text{g/g}$ ), preferably 0.025 to 150  $\mu\text{g/g}$ , more preferably 0.1 to 100  $\mu\text{g/g}$ , even more preferably 0.5 to 75  $\mu\text{g/g}$ , and particularly preferably 0.1 to 50  $\mu\text{g/g}$ .

A content of (z)-3-hexenol in the solid composition of the present invention is not particularly limited, and is, for example, 0.1 to 1,000  $\mu\text{g}$  per gram of the solid composition (0.1 to 1,000  $\mu\text{g/g}$ ), preferably 0.5 to 750  $\mu\text{g/g}$ , more preferably 1 to 500  $\mu\text{g/g}$ , even more preferably 5 to 350  $\mu\text{g/g}$ , and particularly preferably 10 to 200  $\mu\text{g/g}$ .

A content of 1-penten-3-ol in the solid composition of the present invention is not particularly limited, and is, for example, 0.1 to 1,000  $\mu\text{g}$  per gram of the solid composition (0.1 to 1,000  $\mu\text{g/g}$ ), preferably 0.5 to 750  $\mu\text{g/g}$ , more preferably 1 to 500  $\mu\text{g/g}$ , even more preferably 5 to 350  $\mu\text{g/g}$ , and particularly preferably 10 to 200  $\mu\text{g/g}$ .

A content of hexanal in the solid composition of the present invention is not particularly limited, and is, for example, 0.01 to 200  $\mu\text{g}$  per gram of the solid composition (0.01 to 200  $\mu\text{g/g}$ ), preferably 0.05 to 100  $\mu\text{g/g}$ , more preferably 0.1 to 50  $\mu\text{g/g}$ , even more preferably 0.5 to 25  $\mu\text{g/g}$ , and particularly preferably 1 to 10  $\mu\text{g/g}$ .

A content of nerolidol in the solid composition of the present invention is not particularly limited, and is, for example, 0.01 to 200  $\mu\text{g}$  per gram of the solid composition (0.1 to 200  $\mu\text{g/g}$ ), preferably 0.025 to 150  $\mu\text{g/g}$  more preferably 0.1 to 100  $\mu\text{g/g}$ , even more preferably 0.5 to 75  $\mu\text{g/g}$ , and particularly preferably 1 to 50  $\mu\text{g/g}$ .

In the present invention, the content of  $\beta$ -ionone in the solid composition can be measured using gas chromatography (GC) in the same manner as in the measurement of the content of dimethyl sulfide described above. Specifically, a solution is prepared by dissolving the solid composition of the present invention in water, and the content of  $\beta$ -ionone can be measured rising the solution under the same conditions as in the measurement of the content of dimethyl sulfide described above.

In the present invention, the contents of  $\alpha$ -ionone,  $\beta$ -cyclocitral, (z)-3-hexenol, 1-penten-3-ol, hexanal, and nerolidol in the solid composition can be measured using gas chromatography mass spectrometry (GC/MS). Specifically, the contents of the above aroma components can be measured under the following conditions.

Apparatus: GC: Agilent Technologies GC7890B

MS: Agilent Technologies 5977A

HS: Gestel MPS

Tube: Tenax TA, Carbon bx1000

Column: HP-INNOWAX 60 m $\times$ 0.25 mm i.d. df=0.75  $\mu\text{m}$   
Temperature condition: 40° C. (4 minutes) to 5° C./minute to 260° C.

Carrier gas flow rate: He 1.5 ml/min

Injecting tanner: splitless

Ion source temperature: 260° C.

Conditions relating to measurement samples can be set as described in examples below.

Since the solid composition of the present invention contains a tea leaf extract, the solid composition of the present invention may contain an aroma component derived from the tea leaves. Examples of Such an aroma component include, but are not limited to, pentanal (valeraldehyde) 2-methylpropanal (isobutyraldehyde), nonanal, trimethylpyrazine, 1-octen-3-ol, 2-ethyl-3,5-dimethylpyrazine, 2-ethyl-3,6-dimethylpyrazine, 2,4-heptadien-6-one, 2,3-diethyl-5-methylpyrazine, 2-methyl-3-n-propylpyrazine, benzaldehyde, ethyl decanoate, acetylthiazoline, ethylacetophenone, and p-cresol.

### 1-4. Tea Leaf Extract

The solid composition of the present invention contains a tea leaf extract. The term "tea leaf extract" as used herein refers to a component extracted from tea leaves. For the tea leaves in the present invention, leaves obtained from a plant of the genus *Camellia* of the family Theaceae (such as *Camellia sineosis* (L) O. Kuntze) can be used. The tea leaves used in the present invention can be classified into unfermented tea, semi-fermented tea and fermented tea according to the processing method. Examples of the unfermented tea include green tea such as aracha, sencha gyokuro, kabusecha, tencha, bancha, houjicha kamairicha, kukicha, boucha, and mecha. Examples of the semi-fermented tea include oolong tea such as tekkannon, shikisuyu, ogonkei, and buigancha. Examples of the fermented tea include black tea such as Darjeeling, Assam, and Sri Lanka. In the present invention, only one kind of tea leaves may be used alone, or a plurality of kinds of tea leaves may be blended and used. Further pore, the tea leaves are not particularly limited as long as they are parts from which an aroma component can be extracted, and leaves, stems, and the like can be used as appropriate, and the form thereof is not limited to large leaves, powder, or the like. In the present invention, a tea leaf extract of green tea is preferably used, and a tea leaf extract of sencha is more preferably used, although not particularly limited thereto.

A content of the tea leaf extract in the power composition of the present invention is not particularly limited, and is, for example, 30 to 90 wt % preferably 33 to 80 wt %, and more

preferably 40 to 70 wt %. When the content of the tea leaf extract is within the above range, the flavor derived from the tea leaves can be sufficiently felt.

#### 1-5. Other Additives

In addition to the various components described above, it is possible to add additives commonly used in food or drink to the solid composition of the present invention, such as antioxidants, preservatives, pH adjusters, sweeteners, nutritional fortifiers, thickening stabilizers, emulsifiers, dietary fibers, and quality stabilizers, as long as the effect of the present invention is not impaired.

#### 1-6. Applications of Solid Composition

The solid composition of the present invention can be contained in food or drink (beverage or food). That is, according to the present invention, food or drink containing the above-described solid composition can be provided. The solid composition of the present invention is preferably contained in a liquid to prepare a beverage, and is most preferably dissolved in water or hot water to prepare a tea beverage. In this regard, the solid composition of the present invention can be provided as instant tea. The term "instant tea" as used herein refers to a product obtained by drying a solution using an extract of tea leaves as a raw material and processing the dried solution into a solid form. Examples of the tea beverage include unfermented teas (such as green tea), semi-fermented teas (such as oolong tea), and fermented tea (such as black teas), to be more specific, steamed unfermented teas (green tea) such as sencha, bancha, houjicha, gyokuro kabusecha, tencha; unfermented teas such as pan-fried tea such as ureshinocha, aoyagicha, and various Chinese teas; semi-fermented teas such as baozhong tea, tekkannon tea, and oolong tea; and fermented teas such as black tea, awa-bancha, and pu-erh tea. The tea beverage in which the solid composition of the present invention is utilized is preferably green tea. That is, the solid composition of the present invention can be provided as instant green tea.

When the solid composition of the present invention is contained in a liquid such as water or hot water, a content of the solid composition in the liquid is not particularly limited, is, for example, 0.01 to 20 wt %, preferably 0.05 to 10 wt %, and more preferably 0.1 to 5.0 wt %. Also, the content of the solid composition in the solution may be adjusted so that the content of dimethyl sulfide in the solution is, for example, 1 to 2,500 ppb (w/w), preferably 5 to 2,000 ppb (w/w), more preferably 10 to 1,000 ppb (w/w), even more preferably 100 to 500 ppb (w/w), or the content of  $\beta$ -ionone in the solutions is, for example, 0.05 to 100 ppb, preferably 0.2 to 80 ppb, more preferably 0.4 to 60 ppb, and even more preferably 4 to 40 ppb.

The solid composition of the present invention can also be added to food. Examples of such food include cakes, castella, candies, cookies, jellies, puddings chocolates, etc. as confectionery, ice cream, ice candy, sherbet, etc. as frozen desserts, and snacks, regardless of whether they are Japanese sweets or Western sweets; and the solid composition can also be used for breads or dairy products. When the solid composition of the present invention is added to food, the amount added can be appropriately set according to the type of the food or the like.

When the solid composition of the present invention is added to food, the amount added can be appropriately set according to the type of the food or the like. The solid composition of the present invention can be added to the food so that the content of the solid composition in the food is 0.01 to 20 wt %, preferably 0.05 to 10.0 wt %, more preferably 0.1 to 5.0 wt %, even more preferably 0.5 to 5.0

wt %. For example, the solid composition of the present invention can be added to food so that the content of  $\beta$ -ionone in the food is 0.05 to 100 ppb, preferably 0.2 to 80 ppb, more preferably 0.4 to 60 ppb, even more preferably 4 to 40 ppb.

Furthermore, the amount of the solid composition of the present invention added to food may be set using the content of dimethyl sulfide as an index. For example, the solid composition of the present invention can be added to food so that the content of dimethyl sulfide in the food is 1 to 2,500 ppb (w/w), preferably 5 to 2,000 ppb (w/w), more preferably 10 to 1,000 ppb (w/w), even more preferably 100 to 500 ppb (w/w).

Further more, in one embodiment, the present invention is a food or drink (beverage and food) containing dimethyl sulfide and dextran, wherein the food or drink contains dextrin having a molecular weight of 50,000 or more. A content of dimethyl sulfide in the food or drink is preferably 20.3 ppb (w/w) or more per 1.0% solid content (Brix), more preferably 20.8 to 485,000 ppb (w/w) 21.3 to 380,000 ppb (w/w) 22.3 to 242,100 ppb (w/w), 23.1 to 48,400 ppb (w/w) per 1.0% Brix, and even more preferably 24.1 to 24,200 ppb (w/w) per 1.0% Brix. Note that the solid content (Brix) value is a value obtained by converting a refractive index measured at 20° C. using a glucometer, a refractometer, or the like into a mass/mass percent of a sucrose solution based on the conversion table provided by the International Commission for Uniform Methods of Sugar Analysis (ICUMSA), and represents a soluble solid content in a composition. Although the type of the food or drink (beverage and food) is not particularly limited, examples of the beverage include steamed unfermented teas (green tea) such as sencha, bancha, houjicha, gyokuro, kabusecha, and tencha, unfermented teas such as pan-fried tea such as ureshinocha, aoyagicha, and various Chinese teas; semi-fermented teas such as baozhong tea, tekkannon tea, and oolong tea; and fermented teas such as black tea, awa-bancha, and pu-erh tea. Furthermore, examples of the food include confectionery such as cakes, castella candies, cookies, jellies, puddings, chocolates, etc., frozen desserts such as ice cream, ice candy, sherbet, etc., snacks, breads and dairy products.

#### 2. Producing Method

The solid composition of the present invention can be produced through a step of preparing a solution containing dimethyl sulfide and dextran described above and a step of drying the obtained solution. The solution may contain the tea leaf extract and various components as described above in addition to dimethyl sulfide and dextran. The amount of any of the components incorporated can be appropriately set as long as the effect of the present invention is not impaired, and the order of incorporating the various components is not particularly limited. Furthermore, as a solvent of the solution, water may be used, or the extract of tea leaves may be used as it is. Note that the amount of dextrin incorporated into the solution before solidification can be appropriately adjusted so that the content of dextrin in the soluble solid of the solution will be the content in the solid composition described above.

The solution can be dried by a method conventionally known to those skilled in the art. Examples of such a method include spray drying, freeze drying, hot air drying, and vacuum drying, and it is preferable to use spray drying in the present invention. Note that conditions such as temperature and time in the spray drying are not particularly limited, and can be appropriately adjusted for solidifying of the solution.

In producing the solid composition of the present invention, in addition to the steps described above, a step of



concentrating the solution containing dimethyl sulfide and dextrin, a step of sterilizing the solution, or the like can be included. Any of the steps can be carried out using a method conventionally known to those skilled in the art.

Furthermore, for dimethyl sulfide, the tea leaf extract containing dimethyl sulfide (the second tea leaf extract) may be used as described above. By mixing the first tea leaf extract (the tea leaf extract described in the section of "1-4. Tea Leaf Extract" above) and the second tea leaf extract, a solution containing dimethyl sulfide and dextrin can be prepared. Although not particularly limited, the tea leaf extract containing dimethyl sulfide (the second tea leaf extract) can be produced through a step of distilling the tea leaves. The tea leaves used as a raw material in the second tea leaf extract are as described above, and tea leaves that have been covered and cultivated with sunlight shielded before plucking, such as kabusecha, gyokuro, or tencha can be preferably used as a raw material in the present invention.

As a method for distilling tea leaves to obtain the tea leaf extract containing dimethyl sulfide (the second tea leaf extract), a steam distillation method is typically used. The steam distillation method is a method in which steam is passed through a raw material (tea leaves) and an aroma component distilled out along with the steam is cooled and condensed. As the steam distillation method, schemes such as atmospheric pressure steam distillation, reduced pressure steam distillation, and gas-liquid multistage countercurrent contact distillation (spinning cone column) may be adopted, and in the present invention, the atmospheric pressure steam distillation scheme is preferably used. Furthermore, in the steam distillation of tea leaves, a blow-in steam distillation is typically performed. The blow-in steam distillation is a method in which steam is contacted with a raw material directly (tea leaves) placed in a container such as a basket, and the steam obtained though the raw material is recovered and cooled to obtain a distillate.

Regarding the tea leaf extract containing dimethyl sulfide (the second tea leaf extract), the concentration of various aroma components can be increased through a step of further concentrating the extract obtained by the steam distillation method. As such a concentrating method, distillation concentration is typically performed. In the distillation concentration, for example, a scheme in which a tea leaf extract is introduced into a distillation pot, and heated from the bottom to boil the tea leaf extract, and aroma components together with steam are recovered may be adopted. In the distillation concentration method, both the atmospheric pressure distillation concentration and the reduced pressure distillation concentration may be adopted, and in the present invention, the scheme of the reduced pressure distillation concentration is preferably adopted. Furthermore, when distillation concentration is performed, an operation called salt precipitation may be performed. By performing the salt precipitation treatment, the polarity of the salt draws water molecules in the distillate introduced into the distillation pot, and the volatilization of the organic compound can be promoted. The salt precipitation treatment can be performed by making the distillate targeted for concentration contain a salt such as sodium chloride.

Furthermore, the tea leaf extract containing dimethyl sulfide (the second tea leaf extract) may be produced through a process of performing an activated carbon treatment. By performing the activated carbon treatment, the amount of unnecessary aroma components can be reduced. The term "activated carbon" as used herein refers to a porous material having carbon as the main ingredient that is pro-

duced from a carbon material such as wood through an activation reaction at high temperature.

### 3. Method for Enhancing Green Layer Scent in Food or Drink

The solid composition of the present invention obtained as described above may be added to food or drink to increase the green layer scent in the food or drink. Therefore, in another embodiment, the present invention is also a method for increasing the green layer scent in food or drink, including a step of adding the solid composition obtained through the above procedure to the food or drink.

## EXAMPLES

Hereinafter, the present invention will be described based on examples, but the present invention is not limited to these examples.

### 1. Preparation of Solid Composition

#### (i) Preparation of Solid Composition Stock Solution

A solid composition stock solution was prepared using tea leaves extract solid raw material (tea solid law material; the product name GT204S, raw material: green tea (made in China)), linear dextrin A (Matsutani Chemical Industry Co., Ltd., TK-16, weight average molecular weight: 910, DE: 18), linear dextrin B (Sanwa Starch Co., Ltd., Sandec #30, weight average molecular weight: 120,000, DE: 2 to 5), and cyclic dextrin (CycloChem Co., Ltd.,  $\alpha$ -cyclodextrin, weight average molecular weight: 973). Specifically, each raw material was incorporated in proportions shown in Table 1 below to prepare a powder mixture of 6,000 g in total, and 14,000 g of water was added thereto to prepare 20,000 g in total (Brix 30%) of a solid composition stock solution. Note that the above described tea leaves extract solid raw material (tea solid law material) was obtained by drying and solidifying an extract of tea leaves, and did not contain dextrin.

TABLE 1

	Tea Solid Law Material [wt %]	Linear Dextrin A [wt %]	Linear Dextrin B [wt %]	Cyclic Dextrin [wt %]
Sample 1	50	45	0	5
Sample 2	50	31	14	5
Sample 3	50	17	28	5
Sample 4	50	0	45	5

#### (ii) Preparation of Distillate Containing Dimethyl Sulfide at a High Concentration

Separately from (i) above, a tea leaf extract containing dimethyl sulfide at a high concentration was prepared. Specifically, 12 kg of commercially-available kabusecha tea leaves were weighed and mixed with 12 kg of Water, stirred by hand so as not to cause unevenness in the water content of the tea leaves, thereby performing a wetting treatment on the tea leaves. Then, the tea leaves were introduced into a basket placed in a steam distillation pot, and a blow-in steam distillation was performed under the conditions of a steam pressure of 0.25 MPa, a steam flow rate of 16 kg/hr, and a steam temperature of 100° C. (atmospheric pressure). Condensation was performed with the cooling refrigerant temperature set to forward 4° C. and backward 6° C., and at a refrigerant flow rate of 30 L/min to recover a distillate. The recovery time of the distillate was 30 minutes from the start of the distillation. This operation was performed four times to obtain 28 kg in total of the distillate.

1.52 g of powdered activated carbon derived from wood having an average pore size of 3 nm (Osaka Gas Chemical,

## 15

SHIRASAGI WP-Z) was added to 22.4 kg of the obtained distillate and stirred by a stirrer for 10 minutes to perform an activated carbon treatment. The activated carbon in the distillate was removed using a filter paper (ADVANTEC, No. 2). Note that the treatment temperature in the activated carbon treatment was set to 5.7° C. Furthermore, the liquid without the activated carbon treatment and the liquid treated with the activated carbon treatment were mixed at a ratio of 1:4 to obtain 28 kg of a distillate.

## (iii) Preparation of Solid Composition

6 L of the distillate obtained in (ii) above was added to and mixed with 14 L of the solid composition stock solution obtained in (i) above. A portion of the pre-drying stock solution for preparing the solid composition prepared as described above was taken out as a sample for measuring the aroma component concentration, and then the pre-drying stock solution was subjected to spray drying treatment using a spray dryer to prepare a solid composition. Note that the

## 16

G3000PWx1 (particle size 7 µm, 7.8 mm ID.x30 cm) (TOSOH CORPORATION)→[Detector]

Analytical Data System: LabSolutions (Shimadzu Corporation)

Flow rate: 1 mL/min

Injection volume: 50 µL

Mobile phases: 0.1 mol/L sodium nitrate solution

Column temperature: 50° C.

Using STANDARD P-82 (Shodex, Showa Denko K.K.) as a standard solution, retention times of eight kinds of molecular weights were first detected and a calibration curve was prepared based on the detection results. Then, from the total value of the peak areas detected at each retention time in the sample for analysis, the proportion of the peak area (proportion to the total peak area) was calculated for each size of the molecules contained in the sample. The result of the molecular weight distribution of each sample is Shown in Table 2.

TABLE 2

	Molecular Weight Distribution (%)							
	less than 200	200 or more and less than 500	500 or more and less than 1,000	1,000 or more and less than 3,000	3,000 or more and less than 10,000	10,000 or more and less than 30,000	30,000 or more and less than 50,000	50,000 or more and less than 100,000
Sample 1	0.00	7.22	38.26	39.90	12.77	1.84	0.01	0.00
Sample 2	2.79	5.25	33.94	34.21	12.15	4.89	1.82	2.39
Sample 3	5.42	5.59	30.07	27.69	11.12	7.92	4.05	4.21
Sample 4	8.38	5.54	25.79	19.54	10.02	11.67	6.12	7.02

  

	Molecular Weight Distribution (%)							
	150,000 or more and less than 200,000	200,000 or more and less than 250,000	250,000 or more and less than 300,000	300,000 or more and less than 350,000	350,000 or more and less than 400,000	400,000 or more and less than 450,000	450,000 or more and less than 500,000	500,000 or more
Sample 1	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Sample 2	1.01	0.66	0.40	0.25	0.16	0.11	0.07	0.13
Sample 3	2.06	1.33	0.78	0.46	0.29	0.19	0.12	0.20
Sample 4	3.20	2.05	1.19	0.70	0.43	0.28	0.19	0.35

45

drying conditions were such that the inlet hot air temperature was 160° C. and the outlet hot air temperature was 110° C.

## 2. Molecular Weight Distribution in Solid Composition

For each solid composition described above, the molecular weight distribution of soluble molecules contained in the composition was measured. Each solid composition was diluted with 0.1 mol/L sodium nitrate solution to a concentration of 1% (w/v) to prepare a sample for analysis, and the Molecular weight distribution was measured using gel permeation chromatography (GPC). Note that the conditions of the gel permeation chromatography analysis were as follows.

Apparatus: Sampling Injector: 231 XL (GILSON)

Pump: 305 (GILSON)

Column oven: CTO-10AS VP (Shimadzu Corporation)

Detector: RID-10A (Shimadzu Corporation)

Columns: connected in series in the following order

[Pump]→TSKgel Guard Column PWx1 (6.0 mm I.D.x4 cm) (TOSOH CORPORATION)→TSKgel G4000PWx1 (particle size 10 µm, 7.8 mm I.D.x30 cm)(TOSOH CORPORATION)→TSKgel

60

65

Furthermore, Table 3 shows the result of calculating the proportion of dextrin having a molecular weight of 50,000 or more, the proportion of dextrin having a molecular weight of 100,000 or more, the proportion of dextrin having a molecular weight of 150,000 or more, the proportion of dextrin having a molecular weight of 200,000 or more, the proportion of dextrin having a molecular weight of 250,000 or more, the proportion of dextrin having a molecular weight of 300,000 or more, the proportion of dextrin having a molecular weight 350,000 or more, the proportion of dextrin having a molecular weight of 400,000 or more, the proportion of dextrin having a molecular weight of 450,000 or more, and the proportion of dextrin having a molecular weight of 500,000 or more in the molecular weight distribution shown in Table 2. Furthermore, Table 3 also shows the result of calculating the proportion of dextrin having a molecular weight of 50,000 or more and less than 200,000 and the proportion of dextrin having a molecular weight of 200,000 or more and less than 350,000 in the molecular weight distribution described above.

TABLE 3

	Molecular Weight Distribution (%)									
	50,000 or more	100,000 or more	150,000 or more	200,000 or more	250,000 or more	300,000 or more	350,000 or more	400,000 or more	450,000 or more	500,000 or more
Sample 1	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Sample 2	6.76	4.37	2.79	1.78	1.12	0.72	0.47	0.31	0.20	0.13
Sample 3	13.57	9.35	5.42	3.35	2.03	1.25	0.79	0.50	0.31	0.20
Sample 4	21.32	14.30	8.38	5.19	3.14	1.95	1.25	0.52	0.54	0.35

  

Molecular Weight Distribution (%)		
	50,000 or more and less than 200,000	200,000 or more and less than 350,000
Sample 1	0.00	0.00
Sample 2	4.98	1.31
Sample 3	10.21	2.58
Sample 4	16.13	3.93

As shown in Tables 2 and 3 above, it was shown that a peak of dextrin having a molecular weight of 50,000 or more was confirmed in Samples 2 to 4, each of which contains linear dextrin B that is high-molecular-weight dextrin. Note that since Sample 1 not containing linear dextrin B, which is high-molecular-weight dextrin, had no peak observed in the region of a molecular weight of 50,000 or more (see Tables 2 and 3), the peak detected in the region of a molecular weight of 50,000 or more in this test is considered to be due to high-molecular-weight dextrin.

### (3) Evaluation of Aroma Component Retention

Each of the pre-drying stock solution and the solid composition obtained as described above was diluted or dissolved with water so as to have a solid content (Brix) value of 4%. Note that the solid content (Brix) value is a value obtained by converting a refractive index measured at 20° C. using a glucometer, a refractometer, or the like into a mass/mass percent of a sucrose solution based on the conversion table provided by the International Commission for Uniform Methods of Sugar Analysis (ICUMSA), and represents a soluble solid content in a composition. 10 mL of the resultant solution was placed in a vial containing 3 g of sodium chloride, and the vial was sealed and introduced into a gas chromatography analyzer (Flash GC Nose HERACLES II, Alpha M.O.S. Japan K.K.). Dimethyl sulfide in each solution was analyzed under the conditions indicated below.

Incubation: 60° C., 15 min

Syringe: temperature: 70° C. post-injection washing: 90 seconds

Headspace injection: 5000 µl at 250 µl/sec

Column 1: MXT-5 (low polarity 10 m, 180 µm ID, 0.4 µm)

Column 2: MIT-WAX (high polarity 10 m, 180 µm ID, 0.4 µm)

Carrier-gas flow rate: hydrogen 1.6 mL/min

Flame ionization detector (FID) temperature: 260° C.

Injector temperature: 200° C.

Oven temperature: 40° C. (5 seconds) to 1.5° C./second to 250° C. (90 seconds)

Injection time: 125 seconds

Trapping temperature: adsorption 50° C., desorption 240° C.

Trapping time: adsorption 130 seconds, preheating 35 seconds

The retention rate of dimethyl sulfide before and after spray drying were calculated from the integrated value of all peak areas from the data obtained through the analysis. The content of dimethyl sulfide in the solid composition after drying was also determined.

TABLE 4

	Retention Rate of Dimethyl Sulfide	Content in Solid Composition (µg/g)
Sample 1	27%	2.021
Sample 2	30%	2.519
Sample 3	34%	2.920
Sample 4	38%	3.000

As shown in the above results, Samples 2 to 4 (see Table 3), which contain high molecular weight dextrin having a molecular weight of 50,000 or more, exhibited a higher dimethyl sulfide retention rate as compared with Sample 1, which does not contain high molecular weight dextrin having a molecular weight of 50,000 or more (see Table 4). This result shows that the retention rate of dimethyl sulfide in the solid composition improves when the solid composition contains dextrin having a molecular weight of 50,000 or more.

### 4. Sensory Evaluation

0.58 g of the solid composition of Samples 1 to 4 obtained as described above was dissolved in water and adjusted to amount to 100 g, thereby obtaining samples for sensory evaluation. Note that the Brix value of the sample for sensory evaluation was about 0.6%. Converted values of the dimethyl sulfide contents in the samples for sensory evaluation prepared as described above are shown in Table 5.

TABLE 5

	Dimethyl Sulfide Content in Solid Composition (µg/g)	Dimethyl Sulfide Content in Sample for Sensory Evaluation (ppb (w/w) (per Brix0.6)	Dimethyl Sulfide Content in Sample for Sensory Evaluation (ppb (w/w) (in terms of Brix1.0)
Sample 1	2.021	11.72	19.54
Sample 2	2.519	14.61	24.35

19

TABLE 5-continued

	Dimethyl Sulfide Content in Solid Composition ( $\mu\text{g/g}$ )	Dimethyl Sulfide Content in Sample for Sensory Evaluation (ppb (w/w) (per Brix0.6)	Dimethyl Sulfide Content in Sample for Sensory Evaluation (ppb (w/w) (in terms of Brix1.0)
Sample 3	2.920	16.94	28.23
Sample 4	3.000	17.40	29.00

Sensory evaluation was performed on each prepared sample by five panelists who were well trained for flavor evaluation. For the sensory evaluation, the degree of a green layer scent felt in the sample was evaluated in the following five grades, and scored in increments of 0.1 points, and finally the average value of the evaluation points was calculated. Note that in the sensory evaluation, the evaluation point of the base sample (Sample 1) was set to three points, and each sample was evaluated.

- 1: Almost no green layer scent felt.
- 2: Green layer scent felt not so much.
- 3: Green layer scent felt.
- 4: Green layer scent lasting (excellent green layer scent).
- 5: Green layer scent lasting longer (very excellent green layer scent).

TABLE 6

Sensory Evaluation Result						
	Average	Panelist 1	Panelist 2	Panelist 3	Panelist 4	Panelist 5
Sample 1	3.0	3.0	3.0	3.0	3.0	3.0
Sample 2	3.4	3.2	3.8	3.4	3.3	3.2
Sample 3	3.7	4.0	4.0	3.6	3.5	3.2
Sample 4	4.0	5.0	4.2	3.8	3.5	3.5

As described above, it was shown that Samples 2 to 4, which contain high molecular weight dextrin having a molecular weight of 50,000 or more and have a high retention rate of dimethyl sulfide, had the green layer scent lasting longer than Sample 1, which does not contain high molecular weight dextrin having a molecular weight of 50,000 or more. Also, from the above results, it was shown that a beverage containing 12.2 ppb or more of dimethyl sulfide per 0.6% of solid content (Brix) or 20.3 ppb or more of dimethyl sulfide in terms of 1.0% of solid content (Brix), had the green layer scent lasting longer.

The invention claimed is:

1. A solid composition comprising dimethyl sulfide and dextrin, wherein a content of dimethyl sulfide in the solid composition is 2.1  $\mu\text{g}$  or more per gram of the solid composition (2.1  $\mu\text{g/g}$ ), and wherein the solid composition comprises dextrin having a molecular weight of 50,000 or more, wherein the solid composition comprises linear dextrin and cyclic dextrin, and wherein the cyclic dextrin is one or more cyclodextrin(s) selected from the group consisting of  $\alpha$ -cyclodextrin and  $\gamma$ -cyclodextrin,

20

wherein a proportion of the dextrin having a molecular weight of 50,000 or more in molecular weight distribution of the solid composition is 5 to 35%, and wherein a proportion of the dextrin having a molecular weight of 50,000 to 200,000 in molecular weight distribution of the solid composition is 2 to 30%.

2. The solid composition according to claim 1, wherein a proportion of dextrin having a molecular weight of from 200,000 to 350,000 in molecular weight distribution of the solid composition is 0.3 to 6%.

3. The solid composition according to claim 1, wherein a proportion of dextrin having a molecular weight of from 250,000 or more in molecular weight distribution of the solid composition is 0.9 to 10%.

4. The solid composition according to claim 1, wherein a proportion of dextrin having a molecular weight of 350,000 or more in molecular weight distribution of the solid composition is 0.2 to 4%.

5. The solid composition according to claim 1, wherein the solid composition comprises two or more kinds of dextrin.

6. The solid composition according to claim 1, further comprising one or more aroma components selected from the group consisting of  $\beta$ -ionone,  $\alpha$ -ionone,  $\beta$ -cyclocitral, (z)-3-hexenol, 1-penten-3-ol, hexanal, and nerolidol.

7. The solid composition according to claim 1, wherein the solid composition is a powder composition.

8. The solid composition according to claim 1, wherein the solid composition comprises a tea leaf extract.

9. The solid composition according to claim 8, wherein the tea leaf extract is a tea leaf extract of sencha.

10. A food or drink comprising the solid composition according to claim 1.

11. The food or drink according to claim 10, wherein the food or drink is a beverage.

12. The food or drink according to claim 11, wherein the food or drink is a tea beverage.

13. A food or drink comprising dimethyl sulfide and dextrin,

wherein a content of dimethyl sulfide in the food or drink is 20.3 ppb (w/w) or more per 1.0% solid content (Brix), and

the food or drink comprises dextrin having a molecular weight of 50,000 or more,

wherein the food or drink comprises linear dextrin and cyclic dextrin, and

wherein the cyclic dextrin is one or more cyclodextrin(s) selected from the group consisting of  $\alpha$ -cyclodextrin and  $\gamma$ -cyclodextrin,

**21**

wherein a proportion of the dextrin having a molecular weight of 50,000 or more in molecular weight distribution of the food or drink is 5 to 35%, and wherein a proportion of the dextrin having a molecular weight of 50,000 to 200,000 in molecular weight distribution of the food or drink is 2 to 30%.

\* \* \* \* \*

**22**