



PATENT SPECIFICATION

NO DRAWINGS

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COMPLETE SPECIFICATION

A Homogeneous Crystalline Material and a Process for the Preparation thereof

We, TAKEDA CHEMICAL INDUSTRIES, LTD., of 27 Doshomachi 2-chome, Higashi-ku Osaka, Japan, a body corporate organised under the laws of Japan, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to a homogeneous crystalline material and to a process for the preparation thereof. In particular, the invention relates to a homogeneous crystalline material consisting of the alkali metal or ammonium salts of glutamic acid and of a 5'-nucleotide, and to a process for the preparation thereof.

Hereinafter, glutamic acid is referred to as GA, 5'-inosinic acid is referred to as IMP, 5'-guanylic acid is referred to as GMP, IMP and GMP together are referred to as 5'-nucleotide, and sodium, potassium or ammonium is referred to as alkali.

GA or 5'-nucleotide, or their alkali salts, are known to be effective for improving and enhancing the taste or flavour of foodstuffs and have been employed widely in the food industry and in household cooking. It is also known that GA and IMP and/or GMP have a better taste or flavour when used as a mixture than when GA or a 5'-nucleotide is used independently as a condiment mixed with monosodium glutamate (hereinafter referred to as MSG). At the present time, a mixture of MSG and the disodium salt of 5'-nucleotide is employed as a condiment.

These condiments are prepared by such processes as, for example:

(1) A hydrophilic solvent containing the disodium salt of 5'-nucleotide is sprayed onto the surface of MSG crystals and the MSG crystals coated in this way are dried;

(2) MSG crystals are subjected to a film-

like coating with the disodium salt of 5'-nucleotide in a fluidized bed; and

(3) A granular-type condiment is prepared from a kneaded mixture of MSG and the disodium salt of 5'-nucleotide.

In these processes, MSG and the disodium salt are not always mixed homogeneously and the appearance of the product is poor. Furthermore, the processes themselves are rather complicated, so that they are not particularly suitable for preparing condiments on an industrial scale. Moreover, in these processes, crystalline MSG is one of the starting materials, so that these processes have a limited application in the production of condiments.

We have now found that a homogeneous crystalline material consisting of alkali metal or ammonium salts of GA and 5'-nucleotides can be prepared from an aqueous solution of an alkali metal or ammonium salt of GA and a 5'-nucleotide.

The present invention provides a process for the preparation of a homogeneous crystalline material comprising an alkali metal or ammonium salt of glutamic acid and an alkali metal or ammonium salt of a 5'-nucleotide which comprises preparing an aqueous solution of an alkali metal or ammonium salt of glutamic acid and of an alkali metal or ammonium salt of a 5'-nucleotide and crystallising the solution. The homogeneous crystalline material consists of alkali salts of GA and 5'-nucleotide in general, and the ratio of GA to 5'-nucleotide can be controlled within the fixed limits. The material can be used conveniently as a condiment having a pleasant appearance.

A crystalline material having substantially any molar ratio of alkali salt of 5'-nucleotide to alkali salt of GA is obtainable by suitably controlling such conditions as the molar ratio of the starting material, the concentra-

Fluidised

[Price 4s. 6d.]



tion and pH of the solution of alkali salts of GA and 5'-nucleotide and the type and the volume of a hydrophilic organic solvent.

For instance, the desired homogeneous crystalline material can be obtained by the addition of a hydrophilic organic solvent to an aqueous solution of alkali salts of 5'-nucleotide and alkali salts of GA, in which case, as the concentration of the aqueous solution employed is made lower, the molar ratio of alkali salts of 5'-nucleotide becomes larger. This is also the case when the concentration of the hydrophilic organic solvent employed becomes lower. When the pH of the mixed solution of the aqueous solution and of the hydrophilic organic solvent becomes alkaline, the molar ratio of alkali salts of 5'-nucleotide becomes large.

When the precipitation mentioned above is intended, an inorganic salt such as sodium chloride or phosphate, or an organic acid such as succinic acid, citric acid or an amino acid, or a salt thereof, may be added to obtain various crystal forms of the desired material. A surface-active agent may be used for making the crystal form more homogeneous or of larger-size.

Examples of surface-active agents which may be employed in this invention are anion-active surfactants (such as sodium dodecyl sulphate, sodium hexadecyl sulphate, dodecyl benzene sulphonate, lignin sulphonate, oleic diisobutyl amide sulphate, sodium-p-dodecylphenol triethylene glycol ether sulphonate, sodium 1,3-bis-(2-ethylhexyl)-glycerol ether sulphonate, dodecylaminobutyl lactate sulphonate and octylphosphonic acid diethyl ester), cation-active surfactants (such as cetyl dimethyl benzyl ammonium chloride, dodecyl pyridinium phenyl, sulphate, N-octylmorpholine, methoxyoctylbenzyl pyridinium chloride, 2-octadecylquinoline, dodecylimidazoline, 5-tricosanyl isothiurea and p-toluene sulphonate), non-ionic surfactants (such as polyoxyethylene oleyl phenol ether, the ester of linsed oil fatty acid and polyethylene oxide, dodecyl benzene sulphonamide-N-polyethylene oxide, trimethanol amineethane, bis-(di-hydroxypropyl)-amide, N-2-ethylhexyl-8-gluconic acid amide, glycerol monostearate, hydroxymethylphenol sorbitol ether and stearoyl-maleic acid bis-hydroxyethyl ester).

The process of this invention comprises preparing an aqueous solution of salts of GA and 5'-nucleotide; known means of crystallising solutions can be used, and the solution can be left until the homogeneous crystalline material is formed.

In the above method, GA and 5'-nucleotide may be employed in the form of mono- or di-alkali salts or in the form of free acids. The addition of alkali is not necessarily required in the former case, while in the latter case alkali must be added to the aqueous solution of GA and 5'-nucleotide to form the

salts. An alkali hydroxide or water-soluble alkali salt can be used.

The amount of alkali must be sufficient to convert GA and 5'-nucleotide into the mono- or di-alkali salts, preferably mono-alkali salts of GA and dialkali salts of 5'-nucleotide.

The solution of alkali salts of GA and 5'-nucleotide may be prepared by one of the following methods:

(1) Alkali salts of GA and 5'-nucleotide are merely dissolved into water; or (2) GA and 5'-nucleotide are dissolved in water and alkali is added in sufficient quantity to form the salts; or (3) an alkali in a quantity sufficient to convert GA into its alkali salt is added to an aqueous solution of GA and an alkali salt of 5'-nucleotide is then added. The solution of alkali salts of GA and 5'-nucleotide is then subjected to crystallisation, for example, by the addition of a hydrophilic organic solvent.

The concentration of the solution may be varied, depending upon the volume of the hydrophilic organic solvent to be added.

A solution of alkali salts of GA and 5'-nucleotide may be prepared at room temperature, with cooling or with heating. In any case, stirring of the solution promotes the dissolution of free or alkali salts of GA and 5'-nucleotide.

The temperature for the crystallisation may vary from room temperature to an elevated temperature.

Hydrophilic organic solvents which may be employed in this invention include, for example, lower aliphatic alcohols (such as methanol, ethanol and isopropanol), lower aliphatic ketones (such as acetone or methyl-ethyl ketone) or a lower aliphatic ether (such as dioxane). Methanol and ethanol are preferably employed.

The following Examples illustrate the invention:

EXAMPLE 1

30 g. of the disodium salt of IMP were dissolved in a solution of 570 g. of MSG in 400 millilitres of hot water at a temperature of from 90° C. to 100° C. The solution was then cooled to room temperature while stirring gently, whereupon beautiful prisms were precipitated. Yield: 270 g.

EXAMPLE 2

Sodium chloride was dissolved in a solution containing MSG and then disodium salt of IMP dissolved under the same conditions as in Example 1 to make a solution of 10% concentration. Cooling to room temperature then took place while stirring gently, whereupon beautiful prisms containing 5.95% by weight of the disodium salt of IMP were precipitated. Yield: 498 g.

aminomethane

amine

EXAMPLE 3

0.5% by weight of sodium dodecyl sulphate were added to a solution containing MSG and the disodium salt of IMP formed under the same conditions as in Example 1. Crystallisation was then allowed to take place, whereupon larger prisms than those obtained in Example 1, containing 8.1% by weight of the disodium salt of IMP were precipitated. Yield: 256 g.

EXAMPLE 4

34.2 G. of crude ribonucleotide containing 16.15 g. of the disodium salt of IMP, 9.5 g. of the disodium salt of GMP, 0.05 g. of the disodium salt of 5'-uridilic acid and 0.05 g. of 5'-cytidilic acid were added to 974 g. of a solution containing 424.4 g. of MSG, followed by the addition of 825 millilitres of methanol while stirring gently, whereupon fine needles precipitated. Yield: 390.2 g. The needles obtained contained 3.57% of IMP and 2.1% of GMP.

EXAMPLE 5

31.5 G. of the disodium salt of GMP were dissolved in a solution containing 415.5 g. of MSG in 550 millilitres of water 300 millilitres of ethanol were added to the resulting solution, whereupon needles containing 7.2% of GMP were precipitated. Yield: 334 g.

WHAT WE CLAIM IS:—

1. A process for the preparation of a homogeneous crystalline material comprising an alkali metal or ammonium salt of glutamic acid and an alkali metal or ammonium salt of a 5'-nucleotide, which comprises preparing an aqueous solution of an alkali metal or ammonium salt of glutamic acid and of an alkali metal or ammonium salt of a 5'-nucleotide and crystallising the solution.

2. A process as claimed in claim 1, wherein the 5'-nucleotide is 5'-inosinic acid.

3. A process as claimed in claim 1, wherein the 5'-nucleotide is 5'-guanylic acid.

4. A process as claimed in claim 1, wherein the 5'-nucleotide is a mixture of 5'-inosinic acid and 5'-guanylic acid.

5. A process as claimed in any of claims 1 to 4, wherein a hydrophilic organic solvent is added to the solution before crystallisation.

6. A process as claimed in claim 5, wherein the hydrophilic organic solvent is methanol.

7. A process as claimed in any of the preceding claims, wherein the aqueous solution is heated.

8. A process as claimed in any of the preceding claims, wherein a surfactant is added to the solution.

9. A process as claimed in claim 1, substantially as herein described with reference to any of the Examples.

10. A homogeneous crystalline material comprising salts of glutamic acid and 5'-nucleotide when produced by the process claimed in any of the preceding claims.

11. A homogeneous crystalline material comprising an alkali metal or ammonium salt of glutamic acid and an alkali metal or ammonium salt of a 5'-nucleotide.

12. A homogeneous crystalline material as claimed in claim 11, wherein the 5'-nucleotide is 5'-inosinic acid.

13. A homogeneous crystalline material as claimed in claim 11, wherein the 5'-nucleotide is 5'-guanylic acid.

14. A homogeneous crystalline material as claimed in claim 11, wherein the 5'-nucleotide is a mixture of 5'-inosinic acid and 5'-guanylic acid.

ELKINGTON & FIFE,
Chartered Patent Agents,
High Holborn House,
52/54, High Holborn, London, W.C.1,
Agents for the Applicants.

EXAMPLE 3

0.5% by weight of sodium dodecyl sulphate were added to a solution containing MSG and the disodium salt of IMP formed under the same conditions as in Example 1. Crystallisation was then allowed to take place, whereupon larger prisms than those obtained in Example 1, containing 8.1% by weight of the disodium salt of IMP were precipitated. Yield: 256 g.

EXAMPLE 4

34.2 G. of crude ribonucleotide containing 16.15 g. of the disodium salt of IMP, 9.5 g. of the disodium salt of GMP, 0.05 g. of the disodium salt of 5'-uridylic acid and 0.05 g. of 5'-cytidylic acid were added to 974 g. of a solution containing 424.4 g. of MSG, followed by the addition of 825 millilitres of methanol while stirring gently, whereupon fine needles precipitated. Yield: 390.2 g. The needles obtained contained 3.57% of IMP and 2.1% of GMP.

EXAMPLE 5

31.9 G. of the disodium salt of GMP were dissolved in a solution containing 415.5 g. of MSG in 550 millilitres of water 300 millilitres of ethanol were added to the resulting solution, whereupon needles containing 7.2% of GMP were precipitated. Yield: 334 g.

WHAT WE CLAIM IS:—

1. A process for the preparation of a homogeneous crystalline material comprising an alkali metal or ammonium salt of glutamic acid and an alkali metal or ammonium salt of a 5'-nucleotide, which comprises preparing an aqueous solution of an alkali metal or ammonium salt of glutamic acid and of an alkali metal or ammonium salt of a 5'-nucleotide and crystallising the solution.

2. A process as claimed in claim 1, wherein the 5'-nucleotide is 5'-inosinic acid.

3. A process as claimed in claim 1, wherein the 5'-nucleotide is 5'-guanylic acid.

4. A process as claimed in claim 1, wherein the 5'-nucleotide is a mixture of 5'-inosinic acid and 5'-guanylic acid.

5. A process as claimed in any of claims 1 to 4, wherein a hydrophilic organic solvent is added to the solution before crystallisation.

6. A process as claimed in claim 5, wherein the hydrophilic organic solvent is methanol.

7. A process as claimed in any of the preceding claims, wherein the aqueous solution is heated.

8. A process as claimed in any of the preceding claims, wherein a surfactant is added to the solution.

9. A process as claimed in claim 1, substantially as herein described with reference to any of the Examples.

10. A homogeneous crystalline material comprising salts of glutamic acid and 5'-nucleotide when produced by the process claimed in any of the preceding claims.

11. A homogeneous crystalline material comprising an alkali metal or ammonium salt of glutamic acid and an alkali metal or ammonium salt of a 5'-nucleotide.

12. A homogeneous crystalline material as claimed in claim 11, wherein the 5'-nucleotide is 5'-inosinic acid.

13. A homogeneous crystalline material as claimed in claim 11, wherein the 5'-nucleotide is 5'-guanylic acid.

14. A homogeneous crystalline material as claimed in claim 11, wherein the 5'-nucleotide is a mixture of 5'-inosinic acid and 5'-guanylic acid.

ELKINGTON & FIFE,

Chartered Patent Agents,

High Holborn House,

52/54, High Holborn, London, W.C.1,

Agents for the Applicants.

nucleotide

