



(86) Date de dépôt PCT/PCT Filing Date: 1998/10/07
(87) Date publication PCT/PCT Publication Date: 1999/04/15
(45) Date de délivrance/Issue Date: 2002/12/31
(85) Entrée phase nationale/National Entry: 1999/06/07
(86) N° demande PCT/PCT Application No.: JP 1998/004532
(87) N° publication PCT/PCT Publication No.: 1999/017746
(30) Priorité/Priority: 1997/10/07 (9-274842) JP

(51) Cl.Int.⁶/Int.Cl.⁶ A61K 9/20, A61K 47/46, A61K 47/36,
A61K 31/20, A61K 31/355, A61K 31/23

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(54) Titre : PROCEDE DE PREPARATION DE POUDRE EMULSIONNEE
(54) Title: PROCESS FOR THE PREPARATION OF EMULSIFIED POWDERS

(57) **Abrégé/Abstract:**

Emulsified powders are prepared by emulsifying an oil-soluble substance, a chemically modified starch and, if desired, a vegetable gum in water. The emulsified powders obtained according to this process show less disintegration of emulsified particles caused by compressing impact, which leads to less oil exudation. The tableted product of the emulsified powders of the oil-soluble substance is easy in handling, weighing, ingesting and others and thus can be utilized over a wide range of technical fields such as foods and drinks, medicaments or the like.



ABSTRACT

Emulsified powders are prepared by emulsifying an oil-soluble substance, a chemically modified starch and, if desired, a vegetable gum in water. The emulsified powders
5 obtained according to this process show less disintegration of emulsified particles caused by compressing impact, which leads to less oil exudation. The tableted product of the emulsified powders of the oil-soluble substance is easy in handling, weighing, ingesting and others and thus can be
10 utilized over a wide range of technical fields such as foods and drinks, medicaments or the like.

FOP-321-PCT

SPECIFICATION

PROCESS FOR THE PREPARATION OF EMULSIFIED POWDERS

5 TECHNICAL FIELD

10 This invention relates to a process for the preparation of emulsified powders of an oil-soluble substance. The emulsified powders prepared according to the invention are suitable for tableting, because the cells encapsulating oil particles are hardly disintegrated by compressing impact, leading to less oil exudation.

BACKGROUND ART

15 Emulsified powders containing oil-soluble substances such as edible fats have been prepared by emulsifying and dispersing oil-soluble substances in water using an emulsifying agent such as gum arabic, a filler such as dextrin or the like and then drying the emulsion thus
20 obtained by a conventional drying means such as spray drying, drum drying, belt drying, freeze drying or the like. It has been also carried out to emulsify oil-soluble substances using as an emulsifying agent synthetic surfactants such as glycerol fatty acid esters, polyglycerol fatty acid esters, sucrose fatty acid esters and the like.

25 The emulsified powders prepared using gum arabic have a tough coated film and are relatively suited for tableting, but the emulsified particles are difficult to be

finely divided. Further, because gum arabic in an amount of about two or three times that of the oil-soluble substance is required, the oil-soluble substance has a limited content.

5 Although synthetic surfactants are excellent in emulsifying property, a coated film is so weak that the emulsified cells may be disintegrated during the drying step and oil exudation is observed over the surface of powders at a higher rate, and the surfactants are not suited for
10 pulverizing an oil-soluble substance.

 As an alternative to the emulsified powders, there have been proposed mononuclear capsules by microcapsulation using gelatin, but they have the drawbacks that the surface thereof may be converted to a hardened film by formalin
15 treatment or the like, so that there is a high possibility of disintegration in the coated film portion and said portion is disintegrated in the course of tableting.

 As is explained above, when tablets or the like are to be prepared using the emulsified powders of the oil-
20 soluble substance as prepared according to the prior art, oil exudation may be frequently brought about by compressing the emulsified powders having a higher content of the oil-soluble substance, thus the amount thereof to be added is limited. In the case of the emulsified powders having a
25 lower content of the oil-soluble substance, a larger amount of the emulsified powders should be used, but the amount thereof to be added has its limit in view of the composition

of a tablet. Therefore, tablets having a higher content of
the oil-soluble substance are presently not obtainable.
When a large amount of the oil-soluble substance is to be
ingested in the form of tablets as health food, tablets
5 having a higher content of the oil-soluble substance are
desired. Under these circumstances, there have been eagerly
expected emulsified powders of an oil-soluble substance
which do not induce any oil exudation in the course of
tableting.

10

DISCLOSURE OF INVENTION

It is an object of the present invention to
provide emulsified powders containing an oil-soluble
substance, wherein the cells containing oil particles are
15 stable against impact and oil exudation is not brought about
in the course of tableting.

We have made earnest studies to solve the above
problems, and as a result, have found that less
disintegrative emulsified powders can be obtained by
20 emulsifying a core substance of an oil-soluble substance
such as edible fats or the like with a chemically modified
starch and drying the resulting emulsified dispersion using
a conventional drying means, upon which the present
invention has been completed. Moreover, we have found that
25 emulsified powders with a superior quality can be obtained
by coating an oil-soluble substance with both a chemically

modified starch having an excellent emulsifying property and a vegetable gum having an excellent coating property.

Accordingly, the invention relates to a process for the preparation of emulsified powders which comprises
5 emulsifying an oil-soluble substance and a chemically modified starch in water and then drying the emulsion.

Also, the invention relates to a process for the preparation of emulsified powders which comprises emulsifying an oil-soluble substance, a chemically modified
10 starch and a vegetable gum in water and then drying the emulsion.

The emulsified powders prepared according to the invention are an aggregate of a great number of particles (cells) in which a core substance of the oil-soluble
15 substance is coated with the chemically modified starch or the like, and a particle diameter thereof is preferably 50-500 μ . The emulsified particles when emulsified in water have preferably a particle diameter of 2 μ or less.

The oil-soluble substance which may be used in the
20 invention includes fat-soluble vitamins such as vitamin A, vitamin E, vitamin A acetate, vitamin E acetate and the like; unsaturated higher fatty acids such as DHA (docosahexaenoic acid), EPA (eicosapentaenoic acid) and the like; oil-soluble flavors such as orange oil, lemon oil,
25 peppermint oil and the like.

The chemically modified starch as used in the invention refers to an ester of a starch with an organic

acid having an emulsifying activity, which is referred to as "Food starch-modified" in the Gazette of the United States, CFR (FDA) 172.89 (d), 1995. Examples of said organic acid may include succinic acid, acetic acid, adipic acid or an alkyl or alkenyl derivative thereof such as octenyl succinate. As the chemically modified starch is particularly preferred octenyl succinate starch, which is a half ester of a starch and octenyl succinate (See U.S. Patent No. 3,971,852), or a salt thereof, whose examples may include "EMULSTAR 30A" (trade mark, manufactured by MATSUTANI KAGAKU KOGYO CO., LTD.), "CAPSULE" (trade mark, manufactured by National Starch & Chemical Nippon NSC LTD.)

The vegetable gum which may be used in the invention includes gum arabic, xanthan gum, guar gum and the like, as well as pullulan, pectin and the like.

In practicing the present process, to a mixture of 150-200 parts (parts by weight, which will be similarly applied hereinafter), 1-90 parts of the chemically modified starch and, if desired, 1-50 parts of the vegetable gum was added 10-60 parts of the oil-soluble substance and then the mixture was stirred at a temperature of 10-60°C at a revolution number of 5000-15000 per minute for 5-20 minutes to prepare an emulsion of the oil-soluble substance. In order to reduce disintegration during tableting, it is desirable that an emulsified particle diameter may be preferably 2 μ or less. The emulsifier to be applied may be suitably a high-speed agitating emulsifier with a higher

shear force such as "CLEARMIX" (trade mark, manufactured by M TECHNIQUE) or a high pressure emulsifier such as a homogenizer, but it is not limited to said emulsifiers. The emulsion thus prepared may be dried as such by any suitable means such as spray drying, drum drying, belt drying, freeze drying or the like to prepare emulsified powders having excellent tableting performances. The emulsified powders obtained by the process of the invention have a good water-dispersion property and also a superior fluidizing property.

10

EXAMPLES

The present invention will be more specifically illustrated by way of the following Examples.

Example 1

15 In 150 g of water were dissolved 20 g of a chemically modified starch (manufactured by MATSUTANI KAGAKU KOGYO CO., LTD.; trade mark of "EMULSTAR*30A"), 30 g of lactose and 20 g of gum arabic and the solution was sterilized by heating at 85-90°C for 15 minutes. This solution was cooled to 40°C or lower, 30 g of vitamin E was added and admixed and then emulsified by means of TK-HOMO-MIXER (manufactured by TOKUSHU KIKA KOGYO CO., LTD.). The emulsified particles were re-emulsified by means of "CLEARMIX" (trade mark of an emulsifier manufactured by M TECHNIQUE) so as to provide finely divided particles. The emulsion thus prepared was spray-dried at an inlet temperature of 140°C and an exhaust temperature of 90°C

25

* Trademark

using a spray drier (manufactured by OHKAWARA KAKOHKI CO., LTD.) to afford 90 g of emulsified powders of vitamin E (Product 1 of the invention).

Example 2

5 In 150 g of deaerated water were dissolved 30 g of a chemically modified starch (manufactured by National Starch & Chemical Nippon NSC LTD.; trade mark of "CAPSULE"), 14 g of lactose and 25 g of gum arabic and the solution was sterilized by heating at 85-90°C for 15 minutes. This
10 solution was cooled to 40°C or lower, 30 g of DHA oil and 1 g of vitamin E were added and admixed and then emulsified by means of TK-HOMO-MIXER. The emulsified particles were re-emulsified by means of "CLEARMIX" (trade mark of an emulsifier manufactured by M TECHNIQUE) so as to provide
15 finely divided particles. The emulsion thus prepared was spray-dried at an inlet temperature of 140°C and an exhaust temperature of 90°C using a spray drier (manufactured by OHKAWARA KAKOHKI CO., LTD.) to afford 90 g of emulsified powders of DHA (Product 2 of the invention).

20 Example 3

 Following the same procedure as described in Example 2 except that 30 g of peppermint oil was used instead of 30 g of the DHA oil, there was obtained 90 g of emulsified powders of peppermint oil.

25 Example 4

 In 150 g of water were dissolved 20 g of the chemically modified starch as used in Example 1 and 50 g of

dextrin (DE 10) and the solution was sterilized and then 30 g of vitamin E was added. Then, the same procedure as described in Example 1 was repeated to afford 90 g of emulsified powders of vitamin E (Product 4 of the invention).

Example 5

In 150 g of deaerated water were dissolved 30 g of the chemically modified starch as used in Example 2 and 39 g of dextrin (DE 10) and the solution was sterilized. Then, 30 g of DHA oil and 1 g of vitamin E were added. The same procedure as described in Example 1 was repeated to afford 90 g of emulsified powders of DHA (Product 5 of the invention).

Example 6

In 15 kg of purified water was dissolved 5 kg of a chemically modified starch (manufactured by MATSUTANI KAGAKU KOGYO CO., LTD.; "EMULSTAR^{*}30A"). To the solution was added 5 kg of vitamin E acetate (manufactured by EISAI CO., LTD.) and then the mixture was emulsified by means of the TK-HOMO-MIXER and a pressure emulsifier. The emulsion was spray-dried at an inlet temperature of 160°C and an exhaust temperature of 80°C by means of a spray drier (manufactured by OHKAWARA KAKOHKI CO., LTD.) to prepare emulsified powders of vitamin E. To 990 g of the emulsified vitamin E powders thus prepared was added 10 g of hydrous silicon dioxide (manufactured by FUJI SILYSIA CHEMICAL LTD.; "SILYSIA^{*}355")

* Trademark

as a fluidizing agent and admixed to afford emulsified powders of vitamin E (Product 6 of the invention).

Reference Example 1

5 Following the same procedure as described in Example 4 except that an equal amount of gum arabic was used instead of the chemically modified starch, there was obtained 90 g of emulsified powders of vitamin E (Reference Product 1).

Reference Example 2

10 Following the same procedure as described in Example 5 except that an equal amount of gum arabic was used instead of the chemically modified starch, there was obtained 90 g of emulsified powders of DHA (Reference Product 2).

15 Comparative Example 1

 Tablets were prepared by tableting the following formulations according to a conventional method. The tablets thus prepared were stored in polyethylene bags at 40°C for one week.

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Table 1Examples of formulations

| | <u>No. 1</u> | <u>No. 2</u> | <u>No. 3</u> | <u>No. 4</u> | <u>No. 5</u> | <u>No. 6</u> |
|-------------------------------|--------------|--------------|--------------|--------------|--------------|--------------|
| Powder sugar | 93.0 | 93.0 | 93.0 | 93.0 | 93.9 | 93.0 |
| 5 Sucrose fatty acid ester | 1.5 | 1.5 | 1.5 | 1.5 | 1.5 | 1.5 |
| Powdered peppermint flavor | 0.5 | 0.5 | 0.5 | 0.5 | 0.5 | 0.5 |
| 10 Product 1 of the invention | 5.0 | - | - | - | - | - |
| Product 2 of the invention | - | 5.0 | - | - | - | - |
| Product 4 of the invention | - | - | 5.0 | - | - | - |
| 15 Reference Product 1 | - | - | - | 5.0 | - | - |
| Product 5 of the invention | - | - | - | - | 5.0 | - |
| <u>Reference Product 2</u> | <u>-</u> | <u>-</u> | <u>-</u> | <u>-</u> | <u>-</u> | <u>5.0</u> |
| Total (g) | 100.0 | 100.0 | 100.0 | 100.0 | 100.0 | 100.0 |

20

Surface of the above tablets was microscopically observed, while one tablet (1.5 g) was dissolved in 30 g of water and the emulsified particles were microscopically observed. As a result, no oil exudation over the tablet

25 surface was observed in the formulations No. 1 and No. 2 in which Products 1 and 2 of the invention were incorporated respectively, and microscopic observation showed that the emulsified particles after being dispersed in water had a maximum size of around 3 μ and any disintegration of

30 emulsified particles caused by compressing was not observed.

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In the formulation No. 3 or No. 5 in which Product 4 or 5 of the invention was incorporated, slight oil exudation over the surface of tablets was observed and partial disintegration of the emulsified particles was also observed, but each formulation was sufficiently applicable to practical use. On the other hand, in the formulation No. 4 or No. 6 in which Reference Product 1 or 2 was incorporated, partial oil exudation over the surface of tablets was observed and coalesced emulsified particles were observed by microscopic observation after being dispersed in water. After storing the tablets at 40°C for one week, organoleptic tests for their flavor were carried out by ten special panelists. It was judged that the formulations No. 1 and No. 2 in which Products 1 and 2 of the invention were respectively incorporated smelt of no deterioration odor during storage. On the other hand, it was judged by all the panelists that the formulation No. 4 or No. 6 in which Reference Product 1 or 2 was respectively incorporated smelt of noticeable deterioration odor during storage. When the formulations were evaluated prior to tableting according to the same method as described above, no significant difference was observed in the formulations No. 1 to No. 6. It may be considered from the above results that deterioration odor during storage could be caused by disintegration of emulsified particles in the course of tableting.

It is demonstrated from the foregoing that powders coated with the chemically modified starch and vegetable gums show less disintegration in tableting and excellent tableting property and thus are extremely useful.

5

INDUSTRIAL APPLICABILITY

According to the present invention, there is provided emulsified powders of an oil-soluble substance which show less disintegration of emulsified particles caused by compressing impact. The tableted product of the emulsified powders of the oil-soluble substance is easy in handling, weighing, ingesting and others and then can be utilized over a wide range of technical fields such as foods and drinks, medicaments or the like.

10

CLAIMS

1. A process for the preparation of emulsified powders which comprises emulsifying, at a temperature ranging from 10 to 60°C for 5 to 20 minutes, an oil-soluble substance and a chemically modified starch in water and drying, by spray drying, drum drying, belt drying or freeze drying, the emulsion thus formed, said chemically modified starch which represents 0,32 to 35,85 weight per cent of the reaction mixture being an ester of a starch with succinic acid, acetic acid, adipic acid or an alkyl or an alkenyl derivative thereof.
2. A process for the preparation of emulsified powders which comprises emulsifying, at a temperature ranging from 10 to 60°C for 5 to 20 minutes, an oil-soluble substance, a chemically modified starch and a vegetable gum in water and drying, by spray drying, drum drying, belt drying or freeze drying, the emulsion thus formed, said chemically modified starch represents 0,32 to 35,85 and said vegetable gum represents 0,28 to 23,69 weight per cent of the reaction mixture, the chemically modified starch being an ester of a starch with succinic acid, acetic acid, adipic acid or an alkyl or alkenyl derivative thereof and the vegetable gum being selected in the group constituted by gum arabic, xanthan gum, guar gum, pullulan and pectin.
3. The process for the preparation of emulsified powders as claimed in claim 1 or 2

wherein said oil-soluble substance is an edible fat.

4. The process for the preparation of emulsified powders as claimed in claim 3 wherein said edible fat is a medicament or a health food.

5. The process for the preparation of emulsified powders as claimed in claim 3 wherein said edible fat is vitamin E or docosahexaenoic acid.

6. The process for the preparation of emulsified powders as claimed in claim 1 or 2 wherein said chemically modified starch is an ester of a starch with an alkenyl succinate.

7. The process for the preparation of emulsified powders as claimed in claim 6 wherein said alkenyl succinate is octenyl succinate.

8. The process for the preparation of emulsified powders as claimed in claim 6 wherein said chemically modified starch is octenyl succinate starch or sodium octenyl succinate starch.

9. The process for the preparation of emulsified powders as claimed in claim 2 wherein said vegetable gum is gum arabic.