

PROCESS FOR THE EXTRACTION OF ESTERS OF XANTHOPHYLL AND OF CAROTENOIDS

5 The present invention relates to a process for the isolation of the trans-xanthophyll esters and carotenoids and in particular to a process for the isolation of the trans esters of lutein through the extraction of plant material.

US patent No. 6191293 discloses a method for the extraction of lutein from marigold flowers by contacting the flower with a hydrocarbon solvent to extract the ester from the flower. After solvent removal, the crude extracted esters
10 are then mixed with an alcohol where they crystallise of, the purified esters are then collected by filtration. The process provides esters with 40 to 70% purity but the great disadvantage of this process is that the recovered yield generally does not exceed 50% yield (the crystallisation step leads to a dramatic loss of product).

We have found that the yield of purified ester product can be
15 significantly improved, up to 100%, by pre-treating the flower prior to the extraction with a hydrocarbon solvent.

According, to the present invention there is provided a process for the extraction of a trans-xanthophyll ester concentrate from a plant material, which process comprises

20 (a) contacting the plant material containing the xanthophyll ester with an alcohol or a nitrile solvent for a time sufficient to extract any non-xanthophyll compound from the plant material;

(b) separating the alcohol or nitrile solvent and the non-xanthophyll compound dissolved therein from the remaining plant material; and

25 (c) contacting the remaining plant material with a hydrocarbon solvent for a sufficient time to extract the xanthophyll ester from the plant material.

The process for the present invention results in an increase in yield of the ester product when compared to the prior art processes.

30 The process of the present invention comprises contacting a plant material with a solvent which is an alcohol or a nitrile. Suitable plant materials include any material containing the xanthophyll ester and in particular the luteine ester, such as marigold flowers, berries of genus Lycium and Physalis, fruits such as oranges, peaches, papayas, prunes and mangoes. The preferred material is the
35 marigold flower. The process is also applicable to the extraction of zeaxanthin

ester. A preferred process according to the present invention is the extraction of trans-lutein from the marigold flower.

In the first step of the process, the plant material is contacted with a solvent which is selected from an alcohol or a nitrile. Suitable alcohols for use in the process are lower aliphatic alcohols, for example a C₁ to C₆ alcohol, preferably methanol, ethanol and propanol, especially methanol. Suitable nitrile solvents include acetonitrile, propionitrile and the like. The plant material is contacted with the solvent using any suitable method known in the art. The solvent is contacted with the plant material for a time sufficient to extract the non-xanthophyll compound. The amount of time necessary to do so will depend on various factors such as the amount of plant material but may suitably be from 20 minutes up to 24 hours.

In the second step of the process, the solvent is separated from the plant material. This may be carried out by any suitable method. The preferred method is by filtration. The extracted solvent is retained and distilled. The distilled solvent may then be recycled back to the plant material. The residue obtained may be discarded or may be treated further, if needed, to obtain the remaining ester contained therein.

In the third step of the process, the treated plant extract is then contacted with a hydrocarbon solvent to extract the ester product. Suitable solvents include pentane, heptane, hexane and octane, or a mixture thereof. The preferred solvent is hexane or heptane. The plant material is contacted with the hydrocarbon solvent in an amount sufficient to extract maximum amount of esters, typically from 20 minutes up to 24 hours. Suitably, this extraction procedure is carried out at a temperature of from 10 to 80°C, preferably at ambient temperature.

The resulting product comprises a mixture of cis and trans esters of the xanthophyll, with the majority being the trans-esters. The proportion of the two esters will be dependent upon the plant material. A particular advantage of the present invention is that isomerisation does not occur and the amount of cis-esters is kept to a minimum. Typically, the resulting ester product obtained by the process of the present invention comprises less than 10% cis-ester. The purity of the esters product is typically greater than 50%, preferably greater than 65%. The yield of product is typically 100%.

At the end of the third step, the resulting material may be used directly, after solvent distillation, in the nutrition industry or may be treated further to

obtain lutein which may also be used in the nutrition industry or in the animal feed industry, for example as a feed supplement for poultry. Further treatment of ester product may be carried out by any known method. Usually, the process is a saponification process.

5 A further particular advantage of the process of the present invention is that the resulting product is substantially devoid of pesticide residues. The first step of the process removes traces of residue pesticides commonly found in plants and fruits.

10 The process of the present invention may also be used to extract carotenoids from plant materials, especially vegetable materials and according to a further aspect of the present invention there is provided a process for the extraction of a carotenoid concentrate from a plant material, which process comprises

15 (a) contacting the plant material containing the carotenoid with an alcohol or a nitrile solvent for a time sufficient to extract any non-carotenoid compound from the plant material;

(b) separating the alcohol or nitrile solvent and the non-carotenoid compound dissolved therein from the remaining plant material; and

20 (c) contacting the remaining plant material with a hydrocarbon solvent for a sufficient time to extract the carotenoid from the plant material.

The carotenoids that may be extracted from the process include beta-carotene and lycopene. With regard to beta-carotene, this may be obtained from vegetable material such as carrots. With regard to lycopene, this may be obtained from tomatoes.

25 The present invention is now further illustrated with reference to the following examples

Example 1:

30 20 g of marigold corollas (*Tagetes Erecta*), having a lutein ester content of 51.2g/kg, as determined on an aliquot by Soxhlet extraction and subsequent spectrophotometric measurement at 445nm, were placed in a glass column. 100ml of methanol was added, at ambient temperature and allowed to continuously contact the corolla. This procedure was repeated a further three times, each cycle lasting 20 minutes. The methanol extractions were collected, distilled and the
35 combined distillate recycled back to the column. The residue was discarded. The

resulting corollas underwent a second extraction procedure by the addition of 100ml of heptane. This extraction procedure was repeated a further four times, each extraction lasting 20 minutes. The heptane fractions obtained were gathered and analysed by ultra violet (uv) spectroscopy and high performance liquid chromatography (HPLC). 1.08 g of the lutein ester was obtained with 65% purity and having a trans isomer content of 95% (as determined by HPLC after saponification).

Example 2

The procedure as detailed in Example 1 was repeated using 20 g of marigold flowers, having a lutein content of 25.6 g/kg, as determined by on an aliquot by Soxhlet extraction and subsequent spectrophotometric measurement at 445nm. 0.478 g of lutein ester was obtained with 54% purity and having a trans isomer content of 89% (as determined by HPLC after saponification).

Comparative Example 1

This example, which is included for comparative purposes only, illustrates the process of US Patent No. 6191293 for the extraction of the trans-lutein ester from the marigold flower.

20 g of marigold corollas (*Tagetes Erecta*), having a lutein ester content of 51.2g/kg, as determined on an aliquot by Soxhlet extraction and subsequent spectrophotometric measurement at 445nm, were placed in a glass column. 100 ml of heptane was added and the procedure carried out as in Example 1. This extraction procedure was repeated 5 times, each extraction lasting 20 minutes. The heptane fraction obtained were gathered and analysed by uv spectroscopy and HPLC. 0.94 g of lutein ester was obtained with 44% purity and having a trans isomer content of 95% (as determined by HPLC after saponification).

Comparative Example 2

The procedure of Comparative Example 1 was repeated using 20 g of marigold flowers having a lutein content of 25.6 g/kg, as determined by on an aliquot by Soxhlet extraction and subsequent spectrophotometric measurement at 445nm. The resulting product contained 0.47 g of lutein having 22% purity and having a trans isomer content of 88% (as determined by HPLC after saponification).

CLAIMS

1. A process for the extraction of the trans-xanthophyll esters concentrate, which process comprises
 - 5 (a) contacting plant material containing the xanthophyll esters with a solvent selected from an alcohol or a nitrile for a time sufficient to extract any non-xanthophyll compounds from the plant material;
 - (b) separating the alcohol and the non-xanthophyll compounds dissolved therein from the remaining plant material; and
 - 10 (c) contacting the remaining plant material with a hydrocarbon solvent for a sufficient time to extract the xanthophyll ester from the plant material.
2. A process as claimed in claim 1 wherein the trans-xanthophyll esters is lutein or zeaxanthin.
- 15 3. A process as claimed in claim 1 or claim 2 wherein the plant material is marigold flower, berries of genus Lycium and Physalis and fruits selected from oranges, peaches, papayas, prunes and mangoes.
4. A process as claimed in claim 1 or claim 3 wherein the solvent used in step (a) is a lower aliphatic alcohol selected from methanol, ethanol or propanol.
- 20 5. A process as claimed in claim 4 in which the alcohol is methanol.
6. A process as claimed in any one of claims 1 to 3 wherein the solvent used in step (a) is a nitrile solvent selected from acetonitrile or propionitrile.
7. A process as claimed in any one of the preceding claims in which the hydrocarbon solvent is selected from pentane, heptane, hexane, octane or a mixture thereof.
- 25 8. A process as claimed in claim 7 in which the hydrocarbon solvent is heptane or hexane.
9. A process for the extraction of the trans-luteine ester from marigold flowers as claimed in any one of the preceding claims.
- 30 10. A process for the extraction of a carotenoid concentrate from a plant material, which process comprises
 - (a) contacting the plant material containing the carotenoid with an alcohol or a nitrile solvent for a time sufficient to extract any non-carotenoid compound from the plant material;

(b) separating the alcohol or nitrile solvent and the non-carotenoid compound dissolved therein from the remaining plant material; and

(c) contacting the remaining plant material with a hydrocarbon solvent for a sufficient time to extract the carotenoid from the plant material.

- 5 11. A process as claimed in claim 10 in which the carotenoid is beta- carotene or lycopene.