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Su Ref.: PCT/MY2016/050007
Nuestra Ref.: PT/5084/DAVIDH0/14/PCT
Fecha: 20 de marzo de 2017

Oficina Australiana de Patentes
PO Box 200, Woden Act 2606, Australia.
Atención: Benjamín Silva

Apreciado Señor,

RESPUESTA AL CONCEPTO ESCRITO DE LA AUTORIDAD INTERNACIONAL DE EXAMEN PRELIMINAR

SOLICITANTES: DAVID HO SUE SAN
TÍTULO: RECUPERACIÓN DE TOCOFEROLES/TOCOTRIENOLES,
GLICERINA, ESTEROLES Y ÉSTERES DE ÁCIDOS GRASOS
DE ACEITE VEGETAL DESTILADO Y PROCESO PARA LA
MISMA
SOLICITUD NO.: PCT/MY2016/050007

En el asunto de la solicitud arriba identificada para el al concepto escrito de la autoridad internacional de examen preliminar fechado el 20 de enero 2017, anexo a la presente encontrará:

1. Modificación a las reivindicaciones; y
2. Observaciones.

A) MODIFICACIONES

La reivindicación original 1 se modifica para que incluya el nivel inventivo de la solicitud. El fundamento de la modificación se puede encontrar en la página 8, línea 24-25 y en la

página 10, línea 27 de la descripción de la presente solicitud

REIVINDICACIONES

1. Un proceso para recuperar tocotrienoles/tocoferoles de destilado de aceite vegetal que comprende los pasos de:

esterificar el destilado de aceite vegetal con un alcohol en presencia de un catalizador ácido para convertir los ácidos grasos allí contenidos para producir un mezcla esterificada que comprende ésteres de ácidos grasos, reduciendo así el contenido de ácidos grasos en el aceite a menos de 3% en peso;

destilar la mezcla esterificada para separar y eliminar los ésteres de ácidos grasos allí contenidos, en la cual el paso de destilar la mezcla esterificada se lleva a cabo en dos columnas de destilación dispuestas secuencialmente, las cuales dos columnas operan a una temperatura de 120 a 180°C y una presión que varía de 0,01 a 1 kPa, en la cual una porción de la mezcla esterificada es removida a través de corriente elevada de la primera columna de destilación en tanto que las sustancias restantes alimentan la columna siguiente para separación adicional, en la cual los ácidos grasos se extraen de allí para producir una corriente de fondo que comprende tocotrienoles/tocoferoles en una cantidad de 1 a 5% en peso de la composición total:

transesterificar la mezcla destilada con un alcohol en presencia de un catalizador básico para convertir los glicéridos allí presentes en ésteres de ácidos grasos y glicerina, formando un mezcla transesterificada que comprende glicerina y ésteres de ácidos grasos;

purificar la mezcla transesterificada para extraer sustancialmente la glicerina de esta; y

destilar la mezcla purificada para extraer sustancialmente los ésteres de ácidos grasos, produciendo una composición que comprende una concentración predeterminada de tocotrienoles/tocoferoles, en la cual el paso para destilar la mezcla purificada se lleva a cabo en dos columnas de destilación dispuestas en serie, una columna que opera a una temperatura de 120 a 180°C y a una presión de 0,01 a 1 kPa, y la siguiente columna que opera a 120 a 180°C y a 0,01 a 3 kPa, en la cual la primera columna remueve los ésteres de ácidos grasos y la segunda columna remueve además los ésteres de ácidos grasos no recuperados produciendo así una corriente de residuos enriquecida con 5 a 35% en peso de tocotrienoles/tocoferoles.

2. Un proceso de acuerdo con la reivindicación 1, que además comprende un paso de lavado de la mezcla esterificada para eliminar catalizador, alcoholes sin reaccionar y otras impurezas, seguido de calentamiento de la mezcla esterificada lavada para reducir el contenido de humedad allí presente a menos de 0,07% en peso.
3. Un proceso de acuerdo con la reivindicación 1, que además comprende un paso de

lavado de la mezcla purificada para eliminar catalizador, alcoholes sin reaccionar y otras impurezas, seguido de calentamiento de la mezcla transesterificada lavada para reducir el contenido de humedad allí presente a menos de 0,07% en peso.

4. Un proceso de acuerdo con la reivindicación 1, que se caracteriza porque el paso de purificación se lleva a cabo para eliminar glicerina de la mezcla transesterificada por centrifugación o por decantación por gravedad.
5. Un proceso de acuerdo con la reivindicación 1, que además comprende un paso de recuperar esteroides de la composición.
6. Un proceso de acuerdo con las reivindicaciones 1 o 5, que se caracteriza porque los esteroides se recuperan tratando la composición con un alcohol y enfriando la mezcla así formada, produciendo así esteroides cristalizados.
7. Un proceso de acuerdo con las reivindicaciones 2 o 3, que se caracteriza porque el paso de lavado es un paso de lavado con agua a favor o en contra de la corriente.
8. Un proceso de acuerdo con las reivindicaciones 2 o 3, que se caracteriza porque el paso de lavado se lleva a cabo usando agua calentada a 40 a 80°C.
9. Un proceso de acuerdo con las reivindicaciones 2 o 3, que se caracteriza porque la mezcla lavada es calentada a una temperatura de 35 a 80°C durante 0,5 a 10 horas.
10. Un proceso de acuerdo con la reivindicación 9, que se caracteriza porque la mezcla lavada es calentada durante 0,5 a 10 horas.
11. Un proceso de acuerdo con la reivindicación 1, que además comprende un paso de calentamiento de la mezcla esterificada, o de sometimiento de la mezcla esterificada a un secador, para reducir todavía más el contenido de humedad a por debajo de 0,01% en peso.
12. Un proceso de acuerdo con la reivindicación 1, que además comprende un paso de calentamiento de la mezcla esterificada, o de sometimiento de la mezcla purificada a un secador, para reducir todavía más el contenido de humedad a por debajo de 0,01% en peso.
13. Un proceso de acuerdo con la reivindicación 1, que se caracteriza porque el catalizador básico es metóxido de sodio o metóxido de potasio.
14. Un proceso de acuerdo con cualquiera de las reivindicaciones precedentes, que se caracteriza porque el alcohol es cualquiera o una combinación de metanol, etanol, propanol y butanol.

15. Un proceso de acuerdo con la reivindicación 1, que se caracteriza porque el ácido es ácido clorhídrico, ácido acético, ácido sulfúrico o ácido cítrico.
16. Un proceso de acuerdo con la reivindicación 1, que se caracteriza porque el destilado de aceite vegetal es destilado de ácidos grasos de palma o cualquier destilado de aceite vegetal que comprende ácidos grasos en más de 10% en peso de la composición total.

(B) OBSERVACIONES

El concepto escrito del Examinador fue respetuosamente rechazado y las siguientes observaciones distinguirán la solicitud actual de los antecedentes más cercanos del estado de la técnica D1-D3 y D6.

El Examinador sostiene que la característica de “destilación usando dos columnas de destilación secuenciales” no es inventivo porque la destilación en dos etapas (o en múltiples etapas) es bien conocida del estado de la técnica. El Solicitante se permite manifestar su desacuerdo. D2 puede ser el antecedente más cercano del estado de la técnica relacionado con destilación en múltiples etapas. Sin embargo, D2 también reveló una destilación paso a paso, es decir, el proceso de destilación no es continuo. Un operador puede tener que operar el proceso de recuperación manualmente para recoger el residuo, recalibrar el estado de la columna y alimentar otra (o la misma) columna. Por el contrario, la presente configuración de columnas de destilación permite llevar a cabo un proceso continuo de múltiples etapas. Se modifica la Reivindicación 1 para aclarar aún más este punto. Por ello, el sistema que aplica dos columnas de destilación secuenciales continúa el proceso de separación de diferentes fracciones sin interrupción. Se pueden evitar la contaminación de la fracción durante la transferencia y el reacondicionamiento de columnas usadas. Otras ventajas de esta configuración se dan en el argumento previo.

En contraposición con la salida de la columna de destilación que se menciona en el antecedente citado del estado de la técnica, la solicitud actual elimina el éster metílico de ácido graso más liviano (que tiene menos de 16 átomos de carbono) en la primera columna, seguido de eliminar éster metílico de ácido graso más pesado (que tiene más de 16 átomos de carbono) en la columna siguiente. Además, la dimensión de cada columna trabaja de consuno con la presión y la temperatura que se aplican a cada columna para lograr un mejor efecto de separación en diferentes etapas.

Así mismo, no se encuentra una enseñanza relacionada con el conocimiento técnico en la aplicación de dos columnas de destilación secuenciales que obre en beneficio del proceso de separación. Aunque D6 enseña que la destilación que se puede llevar a cabo a 0,00001 mbar a 1 mbar a 50°C a 300°C, no hay conocimiento técnico explícito en llevar a cabo una destilación continua en dos etapas a una porción/fracción especificada. El Solicitante de la presente ha dedicado una cierta cantidad de esfuerzo a encontrar cuál condición le sirve a

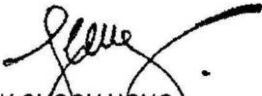
esa porción de la fracción de destilación y a hacer los ajustes correspondientes.

En vista de lo anterior, esperamos que el Examinador reconsidere y admita ahora las reivindicaciones 1-16.

2. Sin embargo, a manera de precaución solicitamos correspondencia adicional en el evento de que el Examinador manifieste la intención de objetar la solicitud. Según esto, el Solicitante solicita respetuosamente la reconsideración y el examen a fondo de la solicitud.

3. Cualquier modificación hecha durante la objeción de la presente solicitud tiene por único objeto hacer más expedito el trámite de la solicitud. A menos que se afirme expresamente lo contrario, la cancelación o modificación de cualquier reivindicación no equivale al abandono de cualquier objeto de la solicitud.

Atentamente,
PINTAS CONSULTING GROUP



LOK CHOON HONG

Reivindicaciones

1. Un proceso para recuperar tocotrienoles/tocoferoles de destilado de aceite vegetal que comprende los pasos de:

esterificar el destilado de aceite vegetal con un alcohol en presencia de un catalizador ácido para convertir los ácidos grasos allí contenidos para producir una mezcla esterificada que comprende ésteres de ácidos grasos, reduciendo así el contenido de ácidos grasos en el aceite a menos de 3% en peso;

destilar la mezcla esterificada para separar y eliminar los ésteres de ácidos grasos allí presentes, en la cual el paso de destilar la mezcla esterificada se lleva a cabo en dos columnas de destilación dispuestas secuencialmente, las cuales dos columnas operan a una temperatura de 120 a 180°C y a una presión que varía de 0,01 a 1 kPa, en la cual una porción de la mezcla esterificada es removida a través de corriente elevada de la primera columna de destilación en tanto que las sustancias restantes alimentan la columna siguiente para separación adicional, en la cual los ácidos grasos se extraen de allí para producir una corriente de fondo que comprende tocotrienoles/tocoferoles en una cantidad de 1 a 5% en peso de la composición total;

transesterificar la mezcla destilada con un alcohol en presencia de un catalizador básico para convertir los glicéridos allí presentes en ésteres de ácidos grasos y glicerina, formando una mezcla transesterificada que comprende glicerina y ésteres de ácidos grasos;

purificar la mezcla transesterificada para extraer sustancialmente la glicerina de esta; y

destilar la mezcla purificada para extraer sustancialmente los ésteres de ácidos grasos, produciendo una composición que comprende una concentración predeterminada de tocotrienoles/tocoferoles, en la cual el paso de destilar la mezcla purificada se lleva a cabo en dos columnas de destilación dispuestas en serie, una columna que opera a una temperatura de 120 a 180°C y a una presión de 0,01 a 1 kPa, y la siguiente columna que opera a 120 a 180°C y 0,01 a 3 kPa, en la cual la primera columna remueve los ésteres de ácidos grasos y la segunda columna remueve además los ésteres de ácidos grasos no recuperados produciendo así una corriente de residuos enriquecida con 5 a 35% en peso de tocotrienoles/tocoferoles.

2. Un proceso de acuerdo con la reivindicación 1, que además comprende un paso de lavado de la mezcla esterificada para eliminar catalizador, alcoholes sin reaccionar y otras impurezas, seguido de calentamiento de la mezcla esterificada lavada para reducir el contenido de humedad allí presente a menos de 0,07% en peso.
3. Un proceso de acuerdo con la reivindicación 1, que además comprende un paso de lavado de la mezcla purificada para eliminar catalizador, alcoholes sin reaccionar y

otras impurezas, seguido de calentamiento de la mezcla transesterificada lavada para reducir el contenido de humedad allí presente a menos de 0,07% en peso.

4. Un proceso de acuerdo con la reivindicación 1, que se caracteriza porque el paso de purificación se lleva a cabo para eliminar la glicerina de la mezcla transesterificada por centrifugación o por decantación por gravedad.
5. Un proceso de acuerdo con la reivindicación 1, que además comprende un paso de recuperar esteroides de la composición.
6. Un proceso de acuerdo con las reivindicaciones 1 o 5, que se caracteriza porque los esteroides se recuperan tratando la composición con un alcohol y enfriando la mezcla así formada, produciendo así esteroides cristalizados.
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8. Un proceso de acuerdo con las reivindicaciones 2 o 3, que se caracteriza porque el paso de lavado se lleva a cabo usando agua calentada a 40 a 80°C.
9. Un proceso de acuerdo con las reivindicaciones 2 o 3, que se caracteriza porque la mezcla lavada es calentada a una temperatura de 35 a 80°C durante 0,5 a 10 horas.
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13. Un proceso de acuerdo con la reivindicación 1, que se caracteriza porque el catalizador básico es metóxido de sodio o metóxido de potasio.
14. Un proceso de acuerdo con cualquiera de las reivindicaciones precedentes, que se caracteriza porque el alcohol es cualquiera o una combinación de metanol, etanol, propanol y butanol.
15. Un proceso de acuerdo con la reivindicación 1, que se caracteriza porque el ácido es

ácido clorhídrico, ácido acético, ácido sulfúrico o ácido cítrico.

16. Un proceso de acuerdo con la reivindicación 1, que se caracteriza porque el destilado de aceite vegetal es destilado de ácidos grasos de palma o cualquier destilado de aceite vegetal que comprende ácidos grasos en más de 10% en peso de la composición total.

Reivindicaciones

1. Un proceso para recuperar tocotrienoles/tocoferoles de destilado de aceite vegetal que comprende los pasos de

esterificar el destilado de aceite vegetal con un alcohol en presencia de un catalizador ácido para convertir los ácidos grasos allí contenidos para producir una mezcla esterificada que comprende ésteres de ácidos grasos, reduciendo así el contenido de ácidos grasos en el aceite a menos de 3% en peso;

destilar la mezcla esterificada para separar y eliminar los ésteres de ácidos grasos allí presentes, en la cual el paso de destilar la mezcla esterificada se lleva a cabo en dos columnas de destilación dispuestas secuencialmente, las cuales dos columnas operan a una temperatura de 120 a 180°C y a una presión que varía de 0,01 a 1 kPa, en la cual una porción de la mezcla esterificada es removida a través de una corriente elevada de la primera columna de destilación, en tanto que las sustancias restantes alimentan la siguiente columna para separación adicional, en la cual se extraen los ácidos grasos allí presentes para producir una corriente de fondo que comprende tocotrienoles/tocoferoles en una cantidad de 1 a 5% en peso de la composición total;

transesterificar la mezcla destilada con un alcohol en presencia de un catalizador básico para convertir los glicéridos allí presentes en ésteres de ácidos grasos y glicerina, formando una mezcla transesterificada que comprende glicerina y ésteres de ácidos grasos;

purificar la mezcla transesterificada para extraer sustancialmente la glicerina de esta; y

destilar la mezcla purificada para extraer sustancialmente los ésteres de ácidos grasos, produciendo una composición que comprende una concentración predeterminada de tocotrienoles/tocoferoles, en la cual el paso de destilar la mezcla purificada se lleva a cabo en dos columnas de destilación dispuestas en serie, en donde una columna opera a una temperatura de 120 a 180°C y a una presión de 0,01 a 1 kPa, y la siguiente columna opera a 120 a 180°C y 0,01 a 3 kPa, en la cual la primera columna remueve ésteres de ácidos grasos y la segunda columna remueve además ésteres de ácidos grasos sin recuperar, produciendo así una corriente de residuos enriquecida con 5 a 35% en peso de tocotrienoles/tocoferoles.

2. Un proceso de acuerdo con la reivindicación 1, que además comprende un paso de lavado de la mezcla esterificada para eliminar catalizador, alcoholes sin reaccionar y otras impurezas, seguido de calentamiento de la mezcla esterificada lavada para reducir el contenido de humedad allí presente a menos de 0,07% en peso.
3. Un proceso de acuerdo con la reivindicación 1, que además comprende un paso de lavado de la mezcla purificada para eliminar catalizador, alcoholes sin reaccionar y otras impurezas, seguido de calentamiento de la mezcla transesterificada lavada para

reducir el contenido de humedad allí presente a menos de 0,07% en peso.

4. Un proceso de acuerdo con la reivindicación 1, que se caracteriza porque el paso de purificación se lleva a cabo para eliminar glicerina de la mezcla transesterificada por centrifugación o por decantación por gravedad.
5. Un proceso de acuerdo con la reivindicación 1, que además comprende un paso de recuperar esteroides de la composición.
6. Un proceso de acuerdo con las reivindicaciones 1 o 5, que se caracteriza porque los esteroides se recuperan tratando la composición con un alcohol y enfriando la mezcla así formada, produciendo así esteroides cristalizados.
7. Un proceso de acuerdo con las reivindicaciones 2 o 3, que se caracteriza porque el paso de lavado es un paso de lavado con agua a favor o en contra de la corriente.
8. Un proceso de acuerdo con las reivindicaciones 2 o 3, que se caracteriza porque el paso de lavado se lleva a cabo usando agua calentada a 40 a 80°C.
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11. Un proceso de acuerdo con la reivindicación 1, que además comprende un paso de calentamiento de la mezcla esterificada, o de sometimiento de la mezcla esterificada a un secador, para reducir todavía más el contenido de humedad a por debajo de 0,01% en peso.
12. Un proceso de acuerdo con la reivindicación 1, que además comprende un paso de calentamiento de la mezcla purificada, o de sometimiento de la mezcla purificada a un secador, para reducir todavía más el contenido de humedad a por debajo de 0,01% en peso.
13. Un proceso de acuerdo con la reivindicación 1, que se caracteriza porque el catalizador básico es metóxido de sodio o metóxido de potasio.
14. Un proceso de acuerdo con cualquiera de las reivindicaciones precedentes, que se caracteriza porque el alcohol es cualquiera o una combinación de metanol, etanol, propanol y butanol.
15. Un proceso de acuerdo con la reivindicación 1, que se caracteriza porque el ácido es ácido clorhídrico, ácido acético, ácido sulfúrico o ácido cítrico.

16. Un proceso de acuerdo con la reivindicación 1, que se caracteriza porque el destilado de aceite vegetal es destilado de ácidos grasos de palma o cualquier destilado de aceite vegetal que comprende ácidos grasos en más de 10% en peso de la composición total.

PATENT COOPERATION TREATY

From the:
INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

To:

LOK, Choon Hong
IP Pintas Group Sdn Bhd, No. 19,
Jalan SS 1/36, Petaling Jaya 47300
Selangor
Malaysia

PCT

NOTIFICATION OF TRANSMITTAL OF
INTERNATIONAL PRELIMINARY
REPORT ON PATENTABILITY
(Chapter II of the Patent Cooperation Treaty)

(PCT Rule 71.1)

Date of mailing
(day/month/year) 19 April 2017

Applicant's or agent's file reference
PT/5084/DAVIDHO/14/PCT

IMPORTANT NOTIFICATION

International application No.
PCT/MY2016/050007

International filing date (day/month/year)
04 February 2016

Priority date (day/month/year)
06 February 2015

Applicant
HO, David Sue San

1. The applicant is hereby notified that this International Preliminary Examining Authority transmits herewith the international preliminary report on patentability and its annexes, if any, established on the international application.
2. A copy of the report and its annexes, if any, is being transmitted to the International Bureau for communication to all the elected Offices.
3. Where required by any of the elected Offices, the International Bureau will prepare an English translation of the report (but not of any annexes) and will transmit such translations to those Offices.
4. **REMINDER**

The applicant must enter the national phase before each elected Office by performing certain acts (filing translations and paying national fees) within 30 months from the priority date (or later in some Offices) (Article 39(1)) (see also the reminder sent by the International Bureau with Form PCT/IB/301).

Where a translation of the international application must be furnished to an elected Office, that translation must contain a translation of any annexes to the international preliminary report on patentability. It is the applicant's responsibility to prepare and furnish such translation directly to each elected Office concerned.

For further details on the applicable time limits and requirements of the elected Offices, see Volume II of the *PCT Applicant's Guide*.

The applicant's attention is drawn to Article 33(5), which provides that the criteria of novelty, inventive step and industrial applicability described in Article 33(2) to (4) merely serve the purposes of international preliminary examination and that "any Contracting State may apply additional or different criteria for the purposes of deciding whether, in that State, the claimed invention is patentable or not" (see also Article 27(5)). Such additional criteria may relate, for example, to exemptions from patentability, requirements for enabling disclosure, clarity and support for the claims.

Name and mailing address of the IPEA/AU

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PATENT COOPERATION TREATY
PCT
INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY
(Chapter II of the Patent Cooperation Treaty)
(PCT Article 36 and Rule 70)

Applicant's or agent's file reference PT/5084/DAVIDHO/14/PCT	FOR FURTHER ACTION See Form PCT/IPEA/416	
International application No. PCT/MY2016/050007	International filing date (<i>day/month/year</i>) 04 February 2016	Priority date (<i>day/month/year</i>) 06 February 2015
International Patent Classification (IPC) or national classification and IPC See Supplemental Box		
Applicant HO, David Sue San		
1. This report is the international preliminary examination report, established by this International Preliminary Examining Authority under Article 35 and transmitted to the applicant according to Article 36. 2. This REPORT consists of a total of 5 sheets, including this cover sheet. 3. This report is also accompanied by ANNEXES, comprising: a. ☒ (<i>sent to the applicant and to the International Bureau</i>) a total of 10 sheets, as follows: ☒ sheets of the description, claims and/or drawings which have been amended and/or sheets containing rectifications authorized by this Authority, unless those sheets were superseded or cancelled, and any accompanying letters (see Rules 46.5, 66.8, 70.16, 91.2, and Section 607 of the Administrative Instructions). ☐ sheets containing rectifications not taken into account by this Authority because they were not available at the time when this Authority began to draw up this report, and any accompanying letters (Rules 66.4<i>bis</i>, 70.2(e), 70.16 and 91.2). ☐ Superseded sheets and any accompanying letters, where this Authority either considers that the superseding sheets contain an amendment that goes beyond the disclosure in the international application as filed, or the superseding sheets were not accompanied by a letter indicating the basis for the amendments in the application filed, as indicated in item 4 of Box No. I and the Supplemental Box (see Rule 70.16(b)). b. ☐ (<i>sent to the International Bureau only</i>) a total of (indicate type and number of electronic carrier(s)) , containing a sequence listing, in electronic form only, as indicated in the Supplemental Box Relating to Sequence Listing (see paragraph 3<i>bis</i> of Annex C of the Administrative Instructions). 4. This report contains indications relating to the following items: ☒ Box No. I Basis of the report ☐ Box No. II Priority ☐ Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability ☐ Box No. IV Lack of unity of invention ☒ Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step and industrial applicability; citations and explanations supporting such statement ☐ Box No. VI Certain documents cited ☐ Box No. VII Certain defects in the international application ☐ Box No. VIII Certain observations on the international application		
Date of submission of the demand 06 December 2016	Date of completion of this report 19 April 2017	
Name and mailing address of the IPEA/AU AUSTRALIAN PATENT OFFICE PO BOX 200, WODEN ACT 2606, AUSTRALIA Email address: pct@ipaaustralia.gov.au	Authorised officer Benjamin Silva AUSTRALIAN PATENT OFFICE (ISO 9001 Quality Certified Service) Telephone No. +61 3 9935 9611	

Box No. I

Basis of the report

1. With regard to the **language**, this report has been established on the basis of:
- ☒ The international application in the language in which it was filed:
- ☐ A translation of the international application into , which is the language of a translation furnished for the purposes of :
- ☐ international search (under Rules 12.3(a) and 23.1 (b)).
- ☐ publication of the international application (under Rule 12.4(a)).
- ☐ international preliminary examination (Rules 55.2(a) and/or 55.3(a) and (b)).
2. With regard to the **elements** of the international application, this report is based on (*replacement sheets which have been furnished to the receiving office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report*):
- ☐ the international application as originally filed/furnished, or
- ☒ the description: pages **1-12** , as originally filed/furnished
pages , received by this Authority on with the letter of
- ☒ the claims: pages , as originally filed/furnished
pages , as amended (together with any statement) under Article 19,
pages **13-15** , received by this Authority on **20 March 2017** with the letter of **20 March 2017**
- ☐ the drawings: pages , as originally filed/furnished
pages , received by this Authority on with the letter of
- ☐ a sequence listing - see Supplemental Box Relating to Sequence Listing.
3. ☐ The amendments have resulted in the cancellation of:
- ☐ the description, pages
- ☐ the claims, Nos.
- ☐ the drawings, sheets/figs
- ☐ the sequence listing (specify):
4. ☐ This report has been established as if (some of) the amendments annexed to this report and listed below had not been made, since either they are considered to go beyond the disclosure as filed, or they were not accompanied by a letter indicating the basis for the amendments in the application as filed, as indicated in the Supplemental Box (Rules 70.2(c) and (*c-bis*)):
- ☐ the description, pages
- ☐ the claims, Nos.
- ☐ the drawings, sheets/figs
- ☐ the sequence listing (specify):
5. ☐ This report has been established:
- ☐ taking into account the **rectification of an obvious mistake** authorized by or notified to this Authority under Rule 91 (Rules 66.1(d-*bis*) and 70.2(e)).
- ☐ without taking into account the **rectification of an obvious mistake** authorized by or notified to this Authority under Rule 91 (Rules 66.4*bis* and 70.2(e)).
6. With regard to top-up searches (Rules 66.1*ter* and 70.2(f)):
- ☒ A top-up search was carried out by this Authority on 17 January 2017.
- ☒ Additional relevant documents have been discovered during the top-up search.
- ☐ No top-up search was carried out by this Authority because it would serve no useful purpose.
7. ☐ Supplementary international search report(s) from Authority(ies)
has/have been received and taken into account in establishing this report (Rule 45*bis*.8(b) and (c)).

* If item 4 applies, some or all of those sheets may be marked "superseded."

Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step and industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Claims 1-16	YES
	Claims NONE	NO
Inventive step (IS)	Claims 1-16	YES
	Claims NONE	NO
Industrial applicability (IA)	Claims 1-16	YES
	Claims NONE	NO

Note: Details of earlier searches conducted following the filing of a demand can be found on Patentscope.

The present claims are directed to a process of recovering tocotrienols/tocopherols from a vegetable oil distillate (VOD), comprising the steps of esterification of the VOD with an alcohol in the presence of an acid catalyst; distilling the esterified VOD in two distillation columns arranged sequentially for separating and removing the fatty acid esters; transesterifying the distilled mixture with an alcohol in the presence of a basic catalyst to form a transesterified mixture of glycerin and fatty acid esters; purifying the transesterified mixture to remove glycerine; and distilling the purified mixture in two distillation columns arranged sequentially to remove fatty acid esters, producing a composition comprising 5-35% tocotrienols/tocopherols.

2. CITATIONS AND EXPLANATIONS:**CITATIONS**

D1: US 2008/0015367 A1 (DOBBINS et al.) 17 January 2008

D2: US 7544822 B2 (HO) 09 June 2009

D3: ARANDA, D. A. G. et al., "Acid-Catalyzed Homogeneous Esterification Reaction for Biodiesel Production from Palm Fatty Acids", Catalysis Letters, 2008, vol. 122, pages 20-25

D4: US 5190618 A (TOP et al.) 02 March 1993

D5: US 5627289 A (JEROMIN et al.) 06 May 1997

D6: US 2011/0220483 A1 (MARGNAT et al.) 15 September 2011

NOVELTY (N)

D1 is directed to a process for recovering phytosterols and tocopherols from deodoriser distillates, including palm oil distillate (D1: Abstract; paragraph 0039), and discloses a process comprising the steps of: (i) esterifying the deodoriser distillate with methanol and a catalytic amount of sulphuric acid; (ii) washing the esterified mixture with water and allowing to separate at higher temperatures; (iii) removing the FAMES, containing less than 4% FFA, from the esterified mixture by distillation; (iv) transesterification of the distilled mixture with methanol and KOH, a step described as a saponification step in D1 because the phytosterols/tocopherols are saponified during this step; and (v) purifying the transesterified mixture by washing with water (D1: Fig. 2A & 2B; paragraphs 0034 & 0051; Example 3; claim 7).

D2 is directed to a process for the extraction and purification of tocopherols, tocotrienols and/or sterols from vegetable oils, said process comprising the steps of: (a) trans-esterification of oil containing tocotrienols/tocopherols in the presence of a monohydric alcohol, and base or acid to form an ester-rich layer and a glycerol-rich layer; (b) separating the ester-rich layer from the glycerol-rich layer by gravitational settling, decantation or separation by centrifugal forces; (c) washing and drying the ester-rich layer; (d) distillation of the resultant dried ester-rich layer to yield a concentrated mixture of tocotrienols/tocopherols, carotenoids; (e) Further trans-esterification of the mixture obtained in (d) in the presence of a monohydric alcohol, and base or acid to convert glycerides in the oil to form an ester-super-rich layer and a glycerol-rich layer; and (f) repeating the above trans-esterification reactions and step-wise molecular distillations to achieve the desired concentration of tocotrienols/tocopherols, carotenoids and sterols (D2: column 2, line 45 to column 3, line 7; claim 1).

D3 is directed to processes for producing biodiesel by the esterification of palm fatty acids, and discloses the following: (a) alkali catalysed transesterification is much faster – about 4000 times – than acid-catalysed transesterification; (b) esterification of FFA with low molecular weight alcohol can be used as a pretreatment for basic transesterification (D3: Section 1, paragraph 2).

D4 is directed to a process for producing high concentration tocopherols/tocotrienols from palm fatty acid distillates (PFADs), said process comprising converting free fatty acids and glycerides in PFAD into fatty acid esters using an alcohol; separating the tocopherol/tocotrienols from said fatty acid esters and other impurities (D4: Abstract; column 1, line 39 to column 2, line 10; Figure on second page).

D5 discloses a process for simultaneously recovering tocopherol and sterol from a mixture containing tocopherol, fats and/or fat derivatives, including a steamer distillate of natural oils and fats, said process comprising the steps of: free fatty acids present in the mixture are esterified with a lower alcohol, preferably methanol; the mixture is subsequently transesterified with the lower alcohol in the presence of a basic catalyst; the excess lower alcohol is distilled off; the catalyst and the glycerol present are removed, for example by washing; the fatty acid alkyl ester is distilled off from the mixture; and, tocopherol and sterol are separated by methods known in the art (D5: Abstract).

D6 is directed to a method of extracting sterols, vitamin E and squalene from deodourisation distillates (DD) of vegetable oils (D6: Abstract), and discloses a process comprising the steps of: (i) esterification of the DD with alcohol in the presence of an acid catalyst (D6: paragraphs 0050 & 0067); (ii) transesterification with an alcohol in the presence of a basic catalyst (D6: paragraphs 0055 & 0072); (iii) three steps of staged distillation (D6: paragraphs 0073-0079); and (iv) crystallisation of sterols (D6: paragraph 0080).

None of the documents D1-D6 discloses the specific method of distillation used in the process of claim 1. Therefore independent claim 1 is considered new. Appended claims 2-16 include further features in addition to those defined in claim 1. Therefore the subject matter of claims 2-16 is also new and meets the requirements of Article 33(2) of the PCT.

INVENTIVE STEP (IS)

D1-D2 & D6 are considered to represent the closest prior art. Their disclosure was discussed in detail under Novelty above. The specific method of distillation used in the process of claim 1 is not obvious to a skilled person in the art from the cited documents, when taken individually or in any combination.

Independent claim 1 therefore involves an inventive step. Appended claims 2-16 include further features in addition to those defined in claim 1. Therefore the subject matter of claims 2-16 also involves an inventive step, and meets the requirements of Article 33(3) of the PCT.

INDUSTRIAL APPLICABILITY (IA)

The invention defined in claims 1-16 is considered to meet the requirements of Industrial Applicability under Article 33(4) of the PCT because it can be made by, or used in, industry.

INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY

International Application No.

Supplemental Box – IPC marks**PCT/MY2016/050007*****C11B 7/00 (2006.01)******C07D 311/72 (2006.01)******C11B 3/12 (2006.01)******C11B 3/02 (2006.01)******C11C 1/10 (2006.01)***

Your Ref : PCT/MY2016/050007
Our Ref : PT/5084/DAVIDHO/14/PCT
Date : 20 March 2017

Australian Patent Office
PO Box 200, Woden Act 2606,
Australia.
Attn: Benjamin Silva

Dear Sir/Madam,

RESPONSE TO WRITTEN OPINION OF THE INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

APPLICANTS : DAVID HO SUE SAN
TITLE : RECOVERY OF TOCOPHEROLS/TOCOTRIENOLS, GLYCERINE, STEROLS AND
FATTY ACID ESTERS FROM VEGETABLE OIL DISTILLATE AND THE PROCESS
THEREOF
APPLICATION NO. : PCT/MY2016/050007

In the matter of the above-identified application for the Written Opinion of International Preliminary Examining Authority dated 20 January 2017, attached are:

1. Amendment to the claims; and
2. Remarks.

A) AMENDMENT(S)

Original claim 1 is amended to include inventive feature of the application. Basis of amendment can be found in page 8, line 24-25 and page 10, line 27 of the description of the present application

CLAIMS

1. A process for recovering tocotrienols/tocopherols from vegetable oil distillate comprising the steps of:
esterifying the vegetable oil distillate with an alcohol in the presence of an acid catalyst to convert the fatty acids contained therein to produce an esterified mixture comprising fatty acid esters, thereby reducing fatty acid content in the oil to less than 3% by weight;
distilling the esterified mixture for separating and removing the fatty acid esters therefrom, in which the step of distilling the esterified mixture is conducted in two distillation columns arranged sequentially, both columns operating at a temperature of 120 to 180°C and a pressure ranging from 0.01 to 1 kPa, in which a portion of the esterified mixture is removed via overhead stream from the first distillation column whereas the remaining substances are fed into the subsequent column for further separation, in which fatty acids are removed therein to produce a bottom stream comprising tocotrienols/tocopherols in an amount of 1 to 5% by weight of total composition;

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transesterifying the distilled mixture with an alcohol in the presence of a basic catalyst for converting glycerides present therein to fatty acid esters and glycerine, forming a transesterified mixture comprising glycerine and fatty acid esters;

purifying the transesterified mixture to substantially remove glycerine therefrom; and

distilling the purified mixture to substantially remove fatty acid esters, producing a composition comprising a predetermined concentration of tocotrienols/tocopherols, in which the step for distilling the purified mixture is conducted in two distillation columns arranged in series, one column operating at a temperature of 120 to 180°C and at a pressure of 0.01 to 1 kPa, and the subsequent column operating at 120 to 180°C and 0.01 to 3 kPa., in which the first column removes fatty acid esters and the second column further removes unrecovered fatty acid esters thereby producing a residue stream enriched with 5 to 35% by weight of tocotrienols/tocopherols.

2. A process according to claim 1 further comprising a step of washing the esterified mixture to remove catalyst, unreacted alcohols and other impurities, followed by heating the washed esterified mixture to reduce moisture content therein to less than 0.07 wt%.
3. A process according to claim 1 further comprising a step of washing the purified mixture to remove catalyst, unreacted alcohols and other impurities, followed by heating the washed transesterified mixture to reduce moisture content therein to less than 0.07 wt%.
4. A process according to claim 1, wherein the purifying step is performed to remove glycerine from the transesterified mixture by centrifugation or by gravitational settling.
5. A process according to claim 1 further comprising a step of recovering sterols from the composition.
6. A process according to claim 1 or 5, wherein the sterols are recovered by treating the composition with an alcohol and chilling the mixture formed thereof, thereby producing crystallised sterols.
7. A process according to claim 2 or 3, wherein the washing step is a direct-current or a counter-current water washing step.
8. A process according to claim 2 or 3, wherein the washing step is performed using water heated to 40 to 80°C.
9. A process according to claim 2 or 3, wherein the washed mixture is heated to a temperature of 35 to 80°C for 0.5 to 10 hours.
10. A process according to claim 9, wherein the washed mixture is heated for 0.5 to 10 hours.
11. A process according to claim 1 further comprising a step of heating the esterified mixture, or subjecting the esterified mixture to a dryer, to further reduce the moisture content to below 0.01% by weight.

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12. A process according to claim 1 further comprising a step of heating the purified mixture, or subjecting the purified mixture to a dryer, to further reduce the moisture content to below 0.01% by weight.
13. A process according to claim 1, wherein the basic catalyst is sodium methoxide or potassium methoxide.
14. A process according to any one of the preceding claims, wherein the alcohol is any one or a combination of methanol, ethanol, propanol and butanol.
15. A process according to claim 1, wherein the acid is hydrochloric acid, acetic acid, sulphuric acid or citric acid.
16. A process according to claim 1, wherein the vegetable oil distillate is palm fatty acid distillate or any vegetable oil distillate comprising fatty acids of more than 10% by weight of total composition.

(B) REMARKS

The Examiner's written opinion was respectfully traversed and remarks below shall distinguish the current application from the closest prior art D1-D3 and D6.

The examiner found that the feature of 'distillation using two sequential distillation columns' is not inventive because two stage (or multi-stage) distillation is well known in the art. The applicant begs to differ. D2 may be the closest prior art related to multi-stage distillation. However, D2 also disclosed a **step-wise** distillation, i.e. the distillation process is **not continuous**. An operator may have to operate the recovery process manually so as to collect the residue, recalibrate the column condition and feed it into another (or the same) column. On the contrary, the present distillation column arrangement enables a continuous multi-stage process to be carried out. Amendment is made to Claim 1 to further clarify this point. Hence, the system applying two sequential distillation columns continues separation process of different fractions without interruption. Contamination of fraction during transfer and re-conditioning of used columns can be avoided. Further advantages of such arrangement are provided in previous argument.

In contrast to the output of distillation column as stated in the cited prior art, the current application removes lighter fatty acid methyl ester (having less than 16 carbon atoms) in the first column, followed by removing heavier fatty acid methyl ester (having more than 16 carbon atoms) at the subsequent column. Further, the dimension of each columns works in conjunction with pressure and temperature applied in each column to achieve a better separation effect in

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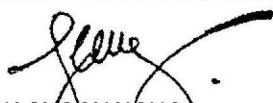
different stages.

Also, there is no found teaching related to the know-how in applying two sequential distillation columns which benefits separation process. Even though D6 teaches that distillation with can be carried out at 0.00001 mbar to 1 mbar at 50 °C to 300 °C, there is no explicit know-how in conducting a continuous two-stage distillation to specified portion/fraction. Applicant of the present application has to spend certain amount of effort in finding out what condition suits that portion of distillation fraction and makes adjustment accordingly.

In view of the above, we hope that the Examiner will reconsider and put the current claims 1-16 in allowable state.

2. Purely as a matter of precaution, however we request further corresponding in the event the Examiner forms the intention to object the application. Accordingly, the applicant respectfully requests reconsideration and further examination of the application.
3. Any amendments made during the objection of this application are intended solely to expedite prosecution of the application. Unless expressly stated otherwise, the cancellation or amendment of any claim does not amount to abandonment of any subject matter in the application.

Yours faithfully,
PINTAS CONSULTING GROUP


LOK CHOON HONG

Claims

1. A process for recovering tocotrienols/tocopherols from vegetable oil distillate comprising the steps of
- 5 esterifying the vegetable oil distillate with an alcohol in the presence of an acid catalyst to convert the fatty acids contained therein to produce an esterified mixture comprising fatty acid esters, thereby reducing fatty acid content in the oil to less than 3% by weight;
- distilling the esterified mixture for separating and removing the fatty acid esters
- 10 therefrom, in which the step of distilling the esterified mixture is conducted in two distillation columns arranged sequentially, both columns operating at a temperature of 120 to 180°C and a pressure ranging from 0.01 to 1 kPa, in which a portion of the esterified mixture is removed via overhead stream from the first distillation column whereas the remaining substances are fed into the subsequent column for further
- 15 separation, in which fatty acids are removed therein to produce a bottom stream comprising tocotrienols/tocopherols in an amount of 1 to 5% by weight of total composition;
- transesterifying the distilled mixture with an alcohol in the presence of a basic catalyst for converting glycerides present therein to fatty acid esters and glycerine, forming a
- 20 transesterified mixture comprising glycerine and fatty acid esters;
- purifying the transesterified mixture to substantially remove glycerine therefrom; and
- distilling the purified mixture to substantially remove fatty acid esters, producing a composition comprising a predetermined concentration of tocotrienols/tocopherols, in which the step for distilling the purified mixture is conducted in two distillation
- 25 columns arranged in series, one column operating at a temperature of 120 to 180°C and at a pressure of 0.01 to 1 kPa, and the subsequent column operating at 120 to 180°C and 0.01 to 3 kPa., in which the first column removes fatty acid esters and the second column further removes unrecovered fatty acid esters thereby producing a residue stream enriched with 5 to 35% by weight of tocotrienols/tocopherols.

2. A process according to claim 1 further comprising a step of washing the esterified mixture to remove catalyst, unreacted alcohols and other impurities, followed by heating the washed esterified mixture to reduce moisture content therein to less than
5 0.07 wt%.

3. A process according to claim 1 further comprising a step of washing the purified mixture to remove catalyst, unreacted alcohols and other impurities, followed by heating the washed transesterified mixture to reduce moisture content therein to less
10 than 0.07 wt%.

4. A process according to claim 1, wherein the purifying step is performed to remove glycerine from the transesterified mixture by centrifugation or by gravitational settling.
15

5. A process according to claim 1 further comprising a step of recovering sterols from the composition.

6. A process according to claim 1 or 5, wherein the sterols are recovered by treating
20 the composition with an alcohol and chilling the mixture formed thereof, thereby producing crystallised sterols.

7. A process according to claim 2 or 3, wherein the washing step is a direct-current or a counter-current water washing step.
25

8. A process according to claim 2 or 3, wherein the washing step is performed using water heated to 40 to 80°C.

9. A process according to claim 2 or 3, wherein the washed mixture is heated to a

temperature of 35 to 80°C for 0.5 to 10 hours.

10. A process according to claim 9, wherein the washed mixture is heated for 0.5 to 10 hours.

5

11. A process according to claim 1 further comprising a step of heating the esterified mixture, or subjecting the esterified mixture to a dryer, to further reduce the moisture content to below 0.01% by weight.

10 12. A process according to claim 1 further comprising a step of heating the purified mixture, or subjecting the purified mixture to a dryer, to further reduce the moisture content to below 0.01% by weight.

15 13. A process according to claim 1, wherein the basic catalyst is sodium methoxide or potassium methoxide.

14. A process according to any one of the preceding claims, wherein the alcohol is any one or a combination of methanol, ethanol, propanol and butanol.

20 15. A process according to claim 1, wherein the acid is hydrochloric acid, acetic acid, sulphuric acid or citric acid.

25 16. A process according to claim 1, wherein the vegetable oil distillate is palm fatty acid distillate or any vegetable oil distillate comprising fatty acids of more than 10% by weight of total composition.

Claims

1. A process for recovering tocotrienols/tocopherols from vegetable oil distillate comprising the steps of
 - 5 esterifying the vegetable oil distillate with an alcohol in the presence of an acid catalyst to convert the fatty acids contained therein to produce an esterified mixture comprising fatty acid esters, thereby reducing fatty acid content in the oil to less than 3% by weight;
distilling the esterified mixture for separating and removing the fatty acid esters
10 therefrom, in which the step of distilling the esterified mixture is conducted in two distillation columns arranged sequentially, both columns operating at a temperature of 120 to 180°C and a pressure ranging from 0.01 to 1 kPa, in which a portion of the esterified mixture is removed via overhead stream from the first distillation column whereas the remaining substances are fed into the subsequent column for further
15 separation, in which fatty acids are removed therein to produce a bottom stream comprising tocotrienols/tocopherols in an amount of 1 to 5% by weight of total composition;
transesterifying the distilled mixture with an alcohol in the presence of a basic catalyst for converting glycerides present therein to fatty acid esters and glycerine, forming a
20 transesterified mixture comprising glycerine and fatty acid esters;
purifying the transesterified mixture to substantially remove glycerine therefrom; and
distilling the purified mixture to substantially remove fatty acid esters, producing a composition comprising a predetermined concentration of tocotrienols/tocopherols, in which the step for distilling the purified mixture is conducted in two distillation
25 columns arranged in series, one column operating at a temperature of 120 to 180°C and at a pressure of 0.01 to 1 kPa, and the subsequent column operating at 120 to 180°C and 0.01 to 3 kPa, in which the first column removes fatty acid esters and the second column further removes unrecovered fatty acid esters thereby producing a residue stream enriched with 5 to 35% by weight of tocotrienols/tocopherols.

2. A process according to claim 1 further comprising a step of washing the esterified mixture to remove catalyst, unreacted alcohols and other impurities, followed by heating the washed esterified mixture to reduce moisture content therein to less than
5 0.07 wt%.

3. A process according to claim 1 further comprising a step of washing the purified mixture to remove catalyst, unreacted alcohols and other impurities, followed by heating the washed transesterified mixture to reduce moisture content therein to less
10 than 0.07 wt%.

4. A process according to claim 1, wherein the purifying step is performed to remove glycerine from the transesterified mixture by centrifugation or by gravitational settling.
15

5. A process according to claim 1 further comprising a step of recovering sterols from the composition.

6. A process according to claim 1 or 5, wherein the sterols are recovered by treating
20 the composition with an alcohol and chilling the mixture formed thereof, thereby producing crystallised sterols.

7. A process according to claim 2 or 3, wherein the washing step is a direct-current or a counter-current water washing step.
25

8. A process according to claim 2 or 3, wherein the washing step is performed using water heated to 40 to 80°C.

9. A process according to claim 2 or 3, wherein the washed mixture is heated to a

temperature of 35 to 80°C for 0.5 to 10 hours.

10. A process according to claim 9, wherein the washed mixture is heated for 0.5 to 10 hours.

5

11. A process according to claim 1 further comprising a step of heating the esterified mixture, or subjecting the esterified mixture to a dryer, to further reduce the moisture content to below 0.01% by weight.

10 12. A process according to claim 1 further comprising a step of heating the purified mixture, or subjecting the purified mixture to a dryer, to further reduce the moisture content to below 0.01% by weight.

15 13. A process according to claim 1, wherein the basic catalyst is sodium methoxide or potassium methoxide.

14. A process according to any one of the preceding claims, wherein the alcohol is any one or a combination of methanol, ethanol, propanol and butanol.

20 15. A process according to claim 1, wherein the acid is hydrochloric acid, acetic acid, sulphuric acid or citric acid.

25 16. A process according to claim 1, wherein the vegetable oil distillate is palm fatty acid distillate or any vegetable oil distillate comprising fatty acids of more than 10% by weight of total composition.

**RECOVERY OF TOCOPHEROLS/TOCOTRIENOLS, GLYCERINE,
STEROLS AND FATTY ACID ESTERS FROM VEGETABLE OIL
DISTILLATE AND THE PROCESS THEREOF**

5 Field Of Invention

The present invention relates to recovery of valuable constituents from vegetable oil distillates from refineries. More particularly, the invention pertains to a multistep process utilizing esterification, distillation and transesterification to recover high
10 quality tocotrienols/tocopherols, sterols and optionally glycerine, as well as free fatty acids in form of ester, from palm oil distillate.

Background Of The Invention

15 Over the years, crude vegetable oils such as palm, palm kernel, rice bran, soy bean, sunflower, canola, rapeseed, cottonseed, safflower and corn oil have been identified as containing high levels of constituents beneficial to human health. These beneficial constituents may include but not limited to tocopherols, tocotrienols, carotenoids and steroids.

20

Tocopherols and tocotrienols are valuable constituents of vegetable oils because of their abilities to act as antioxidants and to provide protection against cell damage in the brain, tumors and various types of cancers, as well as to assist in the rehabilitation of damaged cells. Unique molecular structure of the tocotrienols further imparts
25 hypocholesterolemic characteristics, thereby helping to maintain a healthy cardiovascular system. Specifically, tocotrienols can help lower blood cholesterol level through cleansing of the arteries of accumulated cholesterol.

Carotenoids are natural pigments synthesized by plants imparting yellow, orange or

red colour. Of all the carotenoids, alpha-carotene, beta-carotene and beta-cryptoxanthin are precursors to vitamin A (or retinol), or interchangeably known as provitamin A, to serve as a source of vitamin A. Other carotenoids such as lutein, lycopene and zeaxanthin cannot be converted to vitamin A but they are still of special
5 interest because they are good antioxidants.

Plant based sterols, on the other hand, are recognized for their abilities to block absorption of cholesterol and reduce blood cholesterol level. Because the plant sterols are nearly identical to the cholesterol, they compete with each other for
10 absorption in the small intestines. However, plant sterols are poorly absorbed by humans and they appear to block the absorption of the dietary cholesterol, thereby reducing the blood cholesterol level, as well as the risk of coronary heart disease. Some research studies have also demonstrated that the plant sterols possess anti-cancer, anti-inflammatory, anti-atherogenic and antioxidant characteristics.

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In view of the foregoing, it is desired to extract or recover these beneficial constituents from the crude vegetable oils. Often, during recovery of these constituents, a large fraction of tocotrienols and tocopherols is lost to various distillate streams. These distillate streams typically comprise tocotrienols/tocopherols in an
20 amount of more than 0.10% by weight (or 1000 ppm). However, these streams also comprise high free fatty acid contents which can be more than 10% by weight. Not only free fatty acids, other components such as sterol esters, monoglycerides, diglycerides, triglycerides, glycerine, etc. may also present in the distillate streams. Therefore, in order to obtain a composition concentrated in tocotrienols/tocopherols, it
25 is necessary to remove these undesirable impurities from the oil distillates.

Numerous recovery techniques have been presented in the past, such as solvent extraction, solvent fractionation, ion exchange resin treatment and chromatography method. However, these techniques have several drawbacks, including low yield,

substantial degradation of constituents throughout the process, use of carcinogenic organic solvent, etc. Accordingly, there exists a need for an improved method for recovery of tocotrienols/tocopherols.

5 The present invention provides an improved process for isolating tocotrienols/tocopherols from palm oil distillates, and simultaneously recovering glycerine and free fatty acids in form of esters. In order to further improve quality of the recovered components, acid esterification is utilized as a pretreatment step in the present invention, and then followed by transesterification to remove fatty acids
10 present therein, prior to recovery of tocotrienols/tocopherols.

Summary Of Invention

One of the objects of the invention is to provide a process for isolating tocotrienols,
15 tocopherols and sterols from palm oil distillates, and simultaneously recovering glycerine and free fatty acids in form of esters.

Another object of the invention is to provide a process utilizing acid esterification as a pretreatment step for reducing fatty acid content in the crude oils before being
20 processed further by transesterification, thereby improving quality of the components recovered or isolated from the oils.

Still another object of the invention is to provide a process for isolation and recovery of tocotrienols/tocopherols and optionally other components, such as glycerine and
25 sterols, without being degraded or modified throughout the process.

Yet another object of the invention is to introduce multi-stage distillation as a post-processing step so as to further increase purity of the desirable composition.

At least one of the preceding objects is met, in whole or in part, by the invention, in which one of the embodiments of the invention describes a process for recovering tocotrienols/tocopherols from vegetable oil distillate comprising the steps of esterifying the vegetable oil distillate with an alcohol in the presence of an acid catalyst to convert the fatty acids contained therein to produce an esterified mixture comprising fatty acid esters, thereby reducing fatty acid content in the oil to less than 3% by weight; distilling the esterified mixture for separating and removing the fatty acid esters therefrom; transesterifying the distilled mixture with an alcohol in the presence of a basic catalyst for converting glycerides present therein to fatty acid esters and glycerine, forming a transesterified mixture comprising glycerine and fatty acid esters; purifying the transesterified mixture to substantially remove glycerine therefrom; and distilling the purified mixture to substantially remove fatty acid esters, producing a composition comprising a predetermined concentration of tocotrienols/tocopherols.

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In a preferred embodiment of the invention, the vegetable oil distillate is palm fatty acid distillate or any vegetable oil distillate comprising fatty acids of more than 10% by weight of total composition.

According to the previous embodiment, the vegetable oil distillate is subjected to esterification, distillation, transesterification and purification before being subjected to a further distillation. Preferably, the esterification is conducted in presence of an acid catalyst which can be hydrochloric acid, acetic acid, sulphuric acid or citric acid, whilst the transesterification is performed in presence of a basic catalyst such as sodium methoxide or potassium methoxide.

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The purifying step mentioned in the foregoing refers to centrifugation or gravitational settling so as to remove glycerine from the transesterified mixture.

In another preferred embodiment of the invention, the process further comprises a step of washing the esterified mixture, or the purified mixture, to remove catalyst, unreacted alcohols and other impurities, followed by heating the washed mixture to reduce moisture content therein to less than 0.07 wt%.

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Still another preferred embodiment of the invention is a process for recovering tocotrienols/tocopherols from vegetable oil distillate, further comprising a step of recovering sterols from the composition. The sterols are recovered in crystallised form, as a result of treating the composition with an alcohol and chilling the mixture

10 formed thereof.

Preferably, the alcohol used in esterification, transesterification or sterol recovery step is any one or a combination of methanol, ethanol, propanol and butanol.

15 One skilled in the art will readily appreciate that the invention is well adapted to carry out the aspects and obtain the ends and advantages mentioned, as well as those inherent therein. The embodiments described herein are not intended as limitations on the scope of the invention.

20 **Detailed Description Of The Invention**

Hereinafter, the invention shall be described according to the preferred embodiments of the present invention and by referring to the accompanying description and drawings. However, it is to be understood that limiting the description to the preferred

25 embodiments of the invention and to the drawings is merely to facilitate discussion of the present invention and it is envisioned that those skilled in the art may devise various modifications without departing from the scope of the appended claim.

The term “tocotrienol/tocopherol” or “tocopherol/tocotrienol” used herein throughout

the description shall refer to any one or a combination of tocotrienols and tocopherols.

The present invention relates to a multistep process involving esterification, distillation, transesterification and further distillation for recovery of
5 tocotrienols/tocopherols from vegetable oil distillates from refineries. Optionally, sterols and glycerine, as well as free fatty acids in form of ester, are recovered from the process provided herein.

It should be appreciated that the vegetable oil distillate referred herein preferably has
10 a composition comprising tocotrienols/tocopherols, sterol esters, fatty acids, monoglycerides, diglycerides and triglycerides, wherein tocotrienols/tocopherols are present in an amount of 0.1 to 5% by weight of total composition, whilst the fatty acids are present in an amount of more than 10% by weight of total composition.

15 The process for recovering tocotrienols/tocopherols from vegetable oil distillate, according to its preferred embodiment, comprises the steps of esterifying the vegetable oil distillate with an alcohol in the presence of an acid catalyst to convert the fatty acids contained therein to produce an esterified mixture comprising fatty acid esters, thereby reducing fatty acid content in the oil to less than 3% by weight;
20 distilling the esterified mixture for separating and removing the fatty acid esters therefrom; transesterifying the distilled mixture with an alcohol in the presence of a basic catalyst for converting glycerides present therein to fatty acid esters and glycerine, forming a transesterified mixture comprising glycerine and fatty acid esters; purifying the transesterified mixture to substantially remove glycerine
25 therefrom; and distilling the purified mixture to substantially remove fatty acid esters, producing a composition comprising a predetermined concentration of tocotrienols/tocopherols.

As disclosed in the preferred embodiment of the invention, the vegetable oil distillate

comprising tocotrienols/tocopherols is esterified in the presence of an alcohol and an acid catalyst to convert the fatty acids present in the oil to fatty acid alkyl esters, thus forming an esterified mixture comprising glycerides, fatty acid alkyl esters, sterol esters and tocopherols/tocotrienols, thereby reducing the content of the free fatty acids to less than 1%. Preferably, the alcohol used in the esterification is a lower alkyl alcohol such as methanol, ethanol, iso-propanol and butanol, whilst hydrochloric acid, phosphoric acid, citric acid or other suitable acid can be used as the acid catalyst. It should be appreciably noted that the ratio of the acid catalyst to the lower alkyl alcohol in the esterification solution (i.e. the mixture of the acid catalyst and alcohol) may range from 0.005-5:1.

In the preferred embodiment, the acidic esterification is conducted at a temperature of 5 to 90°C for 0.5 to 6 hours, with or without agitation, but preferably with agitation.

Preferably, after the acid esterification step, the entire esterified mixture is subjected to a heating step, preferably a vacuum evaporation or using the art of equipment of a falling film evaporator, so as to substantially remove the excessive methanol from the mixture. The evaporation may be performed for a period ranging from 1 to 6 hours, at a temperature ranging from 30 to 60°C. The ratio of the methanol being removed to the esterified mixture (i.e. the mixture of the acid catalyst, alcohol and oil distillate) may range from 0.05-0.166:1.

Subsequently, the esterified mixture as obtained is subjected to either direct-current or counter-current water washing, so as to substantially remove the excessive acid catalyst and possibly, unreacted alcohol and other impurities such as sterol esters. Hot or cold water can be used in this step but preferably, water heated to a temperature ranging from 40 to 80°C is used. In order to ensure optimum removal of the catalyst, alcohol and other impurities from the esterified mixture, the washing step is performed repeatedly until the pH of the esterified mixture achieves a range of 6 to 8.

Resulting from the washing step, the moisture content in the esterified mixture may be increased. It is, therefore, of particular interest to reduce the moisture content therein to less than 0.07% by weight prior to subjecting to further processing. Preferably, the moisture in the washed esterified mixture is reduced by drying, whereby the washed mixture is heated to a temperature of 35 to 80°C for 0.5 to 10 hours under atmospheric pressure to evaporate the moisture present therein.

Subsequently, the dried esterified mixture is subjected to distillation, in which the distillation is preferably conducted in a pressure lower than atmospheric pressure (also known as “vacuum distillation”). The distillation may involve one or more than one distillation column to produce a first fraction enriched with fatty acid alkyl esters and a second fraction enriched with tocotrienols/tocopherols. In the preferred embodiment of the invention, the distillation is performed in two distillation columns arranged sequentially, both columns operating at a temperature of 120 to 180°C and a pressure ranging from 0.01 to 1 kPa. Substantial proportion of fatty acid alkyl esters, unreacted fatty acids and other low boiling point substances is separated from the esterified mixture as an overhead stream from the first distillation column. The remaining substances then leave the first distillation column as a residue stream which will be fed into the subsequent column for further separation of fatty acid alkyl esters therefrom. From the subsequent distillation column, fatty acid alkyl esters and possibly, fatty acids are substantially separated to produce a bottom stream, or referred interchangeably to as the “distilled esterified mixture” mentioned in the preceding description, in which the bottom stream comprises tocotrienols/tocopherols in an amount of 1 to 5% by weight of total composition.

In a more preferable embodiment, prior to the distillation, the esterified mixture is subjected to a column dryer to further reduce the moisture content to below 0.01% by weight.

In accordance with the preferred embodiment of the invention, the distilled esterified mixture is subjected to transesterification in the presence of an alcohol and a basic catalyst to convert the glycerides therein to fatty acid esters and glycerine, forming a transesterified mixture comprising glycerine, fatty acid esters, tocotrienols/tocopherols and other impurities such as sterol esters and glycerides. The alcohol used in the transesterification is preferably a lower alkyl alcohol such as methanol, ethanol, iso-propanol and butanol, whilst the basic catalyst can be sodium methoxide or potassium methoxide. Those skilled in the art should appreciate the fact that the ratio of the distilled esterified mixture to the transesterification solution (i.e. the mixture of basic catalyst and alcohol) may range from 0.5-10:1.

Preferably, the transesterification is conducted at a temperature of 5 to 90°C for 0.5 to 6 hours, with or without agitation, but preferably with agitation. More preferably, prior to transesterification, it is desirable to remove moisture contained therein by heating such that the moisture is evaporated, thereby reducing the moisture content of the first fraction to a level of less than 0.20% by weight.

Further in accordance with the preferred embodiment of the invention, it is desirable to purify the transesterified mixture by separating the glycerine therefrom. It can be achieved by subjecting the transesterified mixture to centrifugation or gravitational settling to produce an ester-rich portion (or referred to as "purified mixture" hereinafter) and a glycerine-rich portion comprising 20 to 40% by weight of glycerine.

The glycerine-rich portion is neutralized using an acid such as hydrochloric acid, acetic acid or sulphuric acid to convert the excessive basic catalyst present therein to salts which can be easily separated from the glycerine, thereby producing 75 to 90% by weight of glycerine.

The purified mixture obtained from the preceding step is preferred to be subjected to direct-current or counter-current water washing for removal of the excessive catalyst and possibly, unreacted alcohol and other impurities such as sterols. Hot or cold water
5 can be used in this step but preferably, water heated to a temperature ranging from 40 to 80°C is used. Like the preceding washing step, it is preferred to wash the ester-rich portion repeatedly until the pH of the ester-rich portion falls to a range of 6 to 8.

After washing, the moisture content in the purified mixture may be increased and
10 thus, it is desirable to reduce the moisture content therein to less than 0.07% by weight, prior to further processing. It can be achieved by heating the purified mixture to a temperature of 35 to 80°C for 0.5 to 10 hours, substantially removing the moisture present therein.

15 Subsequent to the moisture reducing or removal step, the resulting purified mixture is distilled for recovery of tocotrienols/tocopherols. It is worth noting that the distillation herein is similar to that depicted in the foregoing. Specifically, the step for distilling the purified mixture refers to vacuum distillation conducted in two distillation columns arranged in series, the first column operating at a temperature of 120 to
20 180°C and at a pressure of 0.01 to 1 kPa, and the subsequent column operating at 120 to 180°C and 0.01 to 3 kPa. From the first column, fatty acid esters as well as other low boiling point components are primarily recovered as distillate, whilst the components remained unrecovered are discharged as residue stream which is to be fed into the next column for further separation of fatty acid esters therefrom. Substantial
25 portion of fatty acid esters is, again, removed from the next column as distillate. At the same time, a residue stream enriched with tocotrienols/tocopherols is produced, in which the tocotrienols/tocopherols are present in an amount of 5 to 35% by weight. It should be noted that the residue stream is referred interchangeably to as “distilled transesterified mixture”.

In a more preferable embodiment, prior to distillation, the purified mixture is heated, or subjected to a column dryer, to further reduce the moisture content to below 0.01% by weight.

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However, those skilled in the art may appreciate the fact that for producing a higher purity stream, or a stream comprising tocotrienols/tocopherols present in a desirable weight percentage, the distilled transesterified mixture can be subjected to another step or more steps of distillation. Alternatively, the distilled transesterified mixture
10 can be recycled to the distillation columns.

Another preferred embodiment of the invention may disclose that the distillate streams obtained in distillation step, comprising predominantly fatty acid esters, can be subjected to further processing or sold directly to fulfill the global market
15 requirement.

Still further in accordance with the preferred embodiment, the distilled transesterified mixture enriched with tocotrienols/tocopherols, is subjected to further processing. Preferably, the distilled transesterified mixture is added with a lower alkyl alcohol,
20 such as any one or a combination of methanol, ethanol, propanol and butanol, to separate sterols in form of crystal. The resulting mixture is continuously agitated and chilled at a temperature ranging from -30 to 0°C for 12 hours to 3 days, consequently forming crystallised sterols. Subsequently, the mixture is subjected to a solid-liquid separation process for removal of the crystallized sterols. Preferably, the mixture is
25 centrifuged, producing a top layer and a bottom layer.

The bottom layer comprising the sterol crystals are then further processed to evaporate the solvent contained therein through heating or by other drying means.

It should be appreciated that the top layer as obtained from the centrifugation comprises tocotrienols/tocopherols in an amount of 30 to 90% by weight. The top layer may be further heated to evaporate and remove portion of the alcohol present therein, forming a composition having a desirable concentration of
5 tocotrienols/tocopherols. Those skilled in the art may also appreciate that the composition may also contain other compounds such as squalene, sterols and CoQ10.

The disclosure includes as contained in the appended claims, as well as that of the foregoing description. Although this invention has been described in its preferred
10 form with a degree of particularity, it is understood that the disclosure of the preferred form has been made only by way of example and that numerous changes in the details of construction and the combination and arrangements of parts may be resorted to without departing from the scope of the invention.

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Claims

1. A process for recovering tocotrienols/tocopherols from vegetable oil distillate comprising the steps of
 - 5 esterifying the vegetable oil distillate with an alcohol in the presence of an acid catalyst to convert the fatty acids contained therein to produce an esterified mixture comprising fatty acid esters, thereby reducing fatty acid content in the oil to less than 3% by weight;
 - distilling the esterified mixture for separating and removing the fatty acid esters
 - 10 therefrom;
 - transesterifying the distilled mixture with an alcohol in the presence of a basic catalyst for converting glycerides present therein to fatty acid esters and glycerine, forming a transesterified mixture comprising glycerine and fatty acid esters;
 - purifying the transesterified mixture to substantially remove glycerine therefrom; and
 - 15 distilling the purified mixture to substantially remove fatty acid esters, producing a composition comprising a predetermined concentration of tocotrienols/tocopherols.
2. A process according to claim 1 further comprising a step of washing the esterified mixture to remove catalyst, unreacted alcohols and other impurities, followed by
 - 20 heating the washed esterified mixture to reduce moisture content therein to less than 0.07 wt%.
3. A process according to claim 1 further comprising a step of washing the purified mixture to remove catalyst, unreacted alcohols and other impurities, followed by
 - 25 heating the washed transesterified mixture to reduce moisture content therein to less than 0.07 wt%.
4. A process according to claim 1, wherein the purifying step is performed to remove glycerine from the transesterified mixture by centrifugation or by gravitational

settling.

5. A process according to claim 1 further comprising a step of recovering sterols from the composition.

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6. A process according to claim 1 or 5, wherein the sterols are recovered by treating the composition with an alcohol and chilling the mixture formed thereof, thereby producing crystallised sterols.

10 7. A process according to claim 2 or 3, wherein the washing step is a direct-current or a counter-current water washing step.

8. A process according to claim 2 or 3, wherein the washing step is performed using water heated to 40 to 80°C.

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9. A process according to claim 2 or 3, wherein the washed mixture is heated to a temperature of 35 to 80°C for 0.5 to 10 hours.

10. A process according to claim 9, wherein the washed mixture is heated for 0.5 to 10 hours.

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11. A process according to claim 1, wherein the step for distilling the esterified mixture is conducted in two distillation columns arranged sequentially, both columns operating at a temperature of 120 to 180°C and a pressure ranging from 0.01 to 1 kPa.

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12. A process according to claim 1, wherein the step for distilling the purified mixture is conducted in two distillation columns arranged in series, one column operating at a temperature of 120 to 180°C and at a pressure of 0.01 to 1 kPa, and the subsequent column operating at 120 to 180°C and 0.01 to 3 kPa.

13. A process according to claim 1 further comprising a step of heating the esterified mixture, or subjecting the esterified mixture to a dryer, to further reduce the moisture content to below 0.01% by weight.

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14. A process according to claim 1 further comprising a step of heating the purified mixture, or subjecting the purified mixture to a dryer, to further reduce the moisture content to below 0.01% by weight.

10 15. A process according to claim 1, wherein the basic catalyst is sodium methoxide or potassium methoxide.

16. A process according to any one of the preceding claims, wherein the alcohol is any one or a combination of methanol, ethanol, propanol and butanol.

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17. A process according to claim 1, wherein the acid is hydrochloric acid, acetic acid, sulphuric acid or citric acid.

18. A process according to claim 1, wherein the vegetable oil distillate is palm fatty
20 acid distillate or any vegetable oil distillate comprising fatty acids of more than 10% by weight of total composition.

25

Abstract**Recovery Of Tocopherols/Tocotrienols, Glycerine, Sterols And Fatty Acid Esters
From Vegetable Oil Distillate And The Process Thereof**

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A process for recovering tocotrienols/tocopherols from vegetable oil distillate comprising the steps of esterifying the vegetable oil distillate with an alcohol in the presence of an acid catalyst to convert the fatty acids contained therein to produce an esterified mixture comprising fatty acid esters, thereby reducing fatty acid content in
10 the oil to less than 3% by weight; distilling the esterified mixture for separating and removing the fatty acid esters therefrom; transesterifying the distilled mixture with an alcohol in the presence of a basic catalyst for converting glycerides present therein to fatty acid esters and glycerine, forming a transesterified mixture comprising glycerine and fatty acid esters; purifying the transesterified mixture to substantially remove
15 glycerine therefrom; and distilling the purified mixture to substantially remove fatty acid esters, producing a composition comprising a predetermined concentration of tocotrienols/tocopherols.

No illustrations for abstract.

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