Abstract: The present invention relates to a group of new plasticizers for thermoplastics such as polypropylene, copolymers of polypropylene, polyethylene, polyethylene terephthalates, polystyrene and/or other polymers or mixtures of polymers containing organic and/or inorganic fillers, where the plasticizer is used for obtaining polyvinyl chloride like aspects and improved processability as well as for obtaining a better flexibility and a lower energy consumption during the processing step.

FIG. 2
Priorities and Cross References


Field of the Invention

This invention relates to plasticizers for thermoplastics and mixtures thereof.

Background

Presently the plastics industry is facing a new challenge: the replacement of polyvinyl chloride (PVC) from almost all the compositions that are being currently used in the industry with alternate plastic compositions that do not present direct toxicity during their combustion.

One of the most important aspects of PVC replacement consists of obtaining compositions that can imitate the quality of PVCs’ softness and processability while eliminating or reducing its disadvantages of the high toxicity of the by-products that result from the combustion of PVC.

Present solutions have given unsatisfactory results in their applications.
Summary

This specification describes a novel class of plasticizers to be used as an additive for plasticizing polymer mixtures, for diminishing the amount of energy used in the processing of polymer mixtures containing inorganic and/or organic fillers, for improving the tensile strength of the resulted mixtures, for increasing the extrudability of complex mixtures and for other physicochemical effects.

The plasticizers disclosed are the reaction products of an alcohol, or alcohols, or an amine or amines or mixture thereof with an epoxidized molecule having at least 15 carbon atoms and 1 to 3 double bonds. It is further disclosed that the epoxidized molecule be an epoxidized cardanol of the formula

![Cardanol Structure]

and the number of hydrogens in the 15 carbon aliphatic chain in the meta position depends upon the number of conjugated double bonds, the number which is selected from the group consisting of 1, 2, and 3 and in the case of an alcohol, the alcohol is preferably selected from the group consisting of ethanol, isopropyl alcohol, and tert-butyl alcohol, and in the case of amines, preferably diethanol amine.

Also disclosed is that the plasticizer may be present in a composition of the plasticizer and a thermoplastic compound. It is further disclosed that the thermoplastic compound is polypropylene, a copolymer of polypropylene, high density polyethylene, a polyethylene terephthalate or polyethylene terephthalate copolymer. It is further disclosed that the plasticizer is present from about 1 to about 12 weight percent of the total composition.
It is also disclosed that the composition may further comprise a flame retardant composition or compound selected from the group consisting of a melamine, a diamine, magnesium hydroxide, aluminum hydroxide and that the flame retardant composition or compound may be present at up to 80 parts of the flame retardant composition to 20 parts of the thermoplastic polymer.

Also, disclosed is a process for the manufacture of a plasticizer compound comprising the steps of heating a mixture of an epoxidized oil and alcohol, in the presence of an acid catalyst and reacting the epoxidized oil with the alcohol until less than 10% of epoxidized oil remains, and removing the excess alcohol. However, the final solution is preferably essentially void of unreacted epoxidized cardanol, meaning the epoxide number is about 0 or is 0. It is also disclosed that the heating can be done at reflux conditions.
Brief Description of Drawings

Fig. 1 is a structural formula of epoxidized cardanol.

Fig. 2 is a structural formula of the reaction product of epoxidized cardanol with isopropyl alcohol.

Detailed Description

Disclosed in this specification is a new class of plasticizers that can be used in thermosetting, thermoplastics, thermoplastic polymer matrices, copolymers, terpolymers, coatings, paints, films, resins binders, fibers and articles formed from the same.

The thermoplastics include but are not limited to such polymers as polypropylene, copolymers of polypropylene, polyethylene, copolymers of polyethylene, high and low density polyethylene, polyethylene terephthalates and its copolymers, also known as PET, polystyrene and/or other polymers or mixtures of polymers containing organic and/or inorganic fillers.

Also disclosed in this specification are novel compounds and methods of forming different types of compounds when reacting various epoxides of cardanol with alcohols, amines, acids or other functional compounds that impart a high temperature resistance for the resulting chemical compounds.

Cardanol is the extracted oil from the cashew nutshell. Cardanol is used to describe the decarboxylated derivatives made from the thermal decomposition of any of the naturally anacardic acids. This includes more than one compound because the composition of the side chain R varies in its degree of unsaturation. Tri unsaturated cardanol is the major component and is usually present at about 41%. The remaining cardanol is about 34% mono-unsaturated with 22% bi-unsaturated, and 2% saturated.
One of cardonal's formula is $C_6H_4(OH)-(CH_2)7-CH=CH-CH2-CH=CH-(CH_2)2-CH_3$

Epoxidized cardanol can be made in many ways. One way is with epichlorohydrine, in the presence of a base.

Epoxidized cardanol made with epichlorohydrine has the following formula:

![Epoxidized Cardanol Formula](attachment:formula_image.png)

having R in the meta position where R is a 15 carbon atom aliphatic chain having one, two or three non-conjugated double bonds and has the formula $[CH_2]_7-C_7H_10-i.CH_3$. The number of hydrogens in R will vary depending upon the number of non-conjugated double bonds.

Cardanol has several naturally occurring isomers. Therefore, the epoxidized cardanol has been characterized in the following formula and shown in Fig. 1 as

A commercial example of an epoxidized cardanol is Cardolite® NC-513 from Cardolite Corporation, Newark, NJ 07105 and has the following formula:

Compounds made in both ways can be used in the reaction to give plasticizers of the aforementioned nature.

When reacted with an alcohol, the following compounds are obtained.

In the general sense, the compound is
where $R_i$ is a 15 carbon atom aliphatic chain containing one, two or three non-conjugated double bonds and has the formula $[\text{CH}_2]^{7-\text{C7Hio-i}_{4}}\text{-CH}_3$ and $R_2$ is selected from the group consisting of alkyls and aryls, or mixtures thereof with or without one or more functional groups. The bond of the oxygen to the carbon may be any carbon of the $R_2$ species.

When reacted with isopropyl alcohol, the structure, as shown in Fig. 2, is
This compound's chemical name is 2-propanol, 1-(1-methylethoxy)-, 3-(cashew nutshell liq. oxy) derivs.

The alcohol reacted with the epoxidized cardanol can be selected from the group consisting of aliphatic or substituted alcohols with one or more functional groups in addition to the hydroxyl group, alkyl alcohols with one or more functional groups in addition to the hydroxyl group, and aryls with one or more functional groups in addition to the hydroxyl group. The alcohol can also be selected from the group consisting of methanol, ethanol, propanol isomers, and butanol isomers.

It is also known that the reaction of the epoxidized cardanol with an alcohol in the presence of an acid, in this case phosphoric acid, will create two additional species believed present in the composition. These are

![Chemical structure](image)

and
Where $R$ in both instances is a 15 carbon atom aliphatic chain containing one, two or three non-conjugated double bonds and has the formula $[\text{CH}_2]_7-\text{C}_7\text{H}_{10}-\text{i}_4-\text{CH}_3$.

The plasticizer aspect of the new molecule is based upon the discovery that when used in a polymer/filler mixture, it lowers the viscosity of the composition and increases greatly the amount of fire-retardant or any other organic/inorganic filler that can be added to the mixture, it increases the its extrudability, it increases the elongation at the breaking point, it changes the aspect of the extrudate which becomes similar to that of polyvinyl chloride (PVC).

**EXPERIMENTAL**

**Example I**

Plasticizer 1 was made introducing 1000 grams of epoxidized cardanol (Cardolite® NC-513) into a round bottom flask provided with a stirring, heating and cooling system. Approximately 1 L of ethanol was added to the flask at room temperature, approximately 25 °C. The stirring process was started, approximately 25 grams of phosphoric acid was gradually added to the flask, and the mixture heated at atmospheric reflux conditions for
3 hours. (It should be noted that methanesulphonic acid or other acidic catalysts are believed suitable as well.)

The excess of ethanol was eliminated by simple distillation. The traces of ethanol remaining in the mixture were then eliminated by applying high vacuum at a temperature of 120 °C for at least 3 hrs. At least 90% of the epoxidized cardanol should be reacted, or alternatively 10% of the epoxidized cardanol or less may remain in the solution. Preferably however, the reaction product should be preferably essentially void of unreacted epoxidized cardanol, or essentially free of unreacted epoxidized cardanol, which means that there could be some trace amounts of unreacted epoxidized cardanol. One measure is the epoxide number and an epoxide number of about 0, or 0 is considered essentially void of, essentially free of unreacted epoxidized cardanol.

The liquid, as it results with or without any neutralizing agent, was mixed in the complex plastic mixtures which may or may not contain fire retardants, organic fillers and/or inorganic fillers. The epoxidic ring can be attacked by the nucleophile in either location, nevertheless for simplicity, we have shown only the major compounds formed.

A variant of this reaction implies a reverse order addition. 25 grams of acid catalyst, either methanesulphonic acid, phosphoric acid or other acids, is added to one liter of ethanol. Then, after increasing the temperature of this solution to about 40 °C, one kilogram of the epoxidized cardanol is gradually added.

The resulting compound can be described as the reaction product of epoxidized cardanol and ethanol. As shown in TABLE II, it is believed that other alcohols work as well, therefore this class of plasticizers can be called the reaction product of epoxidized cardanol with at least one alcohol.

This material can be compounded with a polyolefinic compound with or without the presence of a thermoplastic polyolefin (such as Engage® (The DOW Chemical
Company, USA or Kraton® (KRATON Polymers U.S. LLC) polymers), in the preferred range of 0.5 - 12 percent by weight of the composition.

The plasticizing property is demonstrated in Table I - Properties of Plasticizer 1 in Polypropylene. The properties indicated were measured on a mixture of polypropylene (Marlex® ALN-070 Chevron Phillips Chemical Company, LLC USA, which is an impact copolymer of high strength and already lubricated) with Plasticizer 1, at the levels indicated.

<table>
<thead>
<tr>
<th>Plasticizer (wt %)</th>
<th>Control</th>
<th>Exp 1</th>
<th>Exp 2</th>
<th>Exp 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Polypropylene (wt %)</td>
<td>100</td>
<td>95</td>
<td>93</td>
<td>91</td>
</tr>
<tr>
<td>Melt Flow (g/10min @ 230 °C)</td>
<td>6.64</td>
<td>7.96</td>
<td>10.42</td>
<td>10.84</td>
</tr>
<tr>
<td>Hardness (Shore A)</td>
<td>96.3</td>
<td>94.6</td>
<td>95.2</td>
<td>94.8</td>
</tr>
<tr>
<td>Tensil (psi)</td>
<td>2881</td>
<td>1791</td>
<td>2400</td>
<td>2421</td>
</tr>
<tr>
<td>Elongation (%)</td>
<td>17.6</td>
<td>179.12</td>
<td>406.93</td>
<td>403.95</td>
</tr>
</tbody>
</table>

Plasticizer 1 also enabled loading of optional flame retardants at levels previously not achievable. For example, Plasticizer 1 was used at 7 wt % of the composition and the loading of 65% Magnesium hydroxide was achieved in high density polyethylene. This is approximately 20% higher than the loading without the plasticizer. The composition can also be halogen free.

An optional flame retardant powder mixture can be added and is described as a nitrogenous phosphate or sulfonate formed in the presence of a char catalyst. The nitrogenous phosphate or sulfonate component may be formed using any of the conventional techniques with the char catalyst being introduced at least in part prior to the formation of the final product. For example, the char catalyst may be introduced either in total or in part with one or more of the nitrogen containing compounds and then have the phosphorus or sulfur containing compound reacted in. These nitrogenous phosphate or sulfonate components may also be formed by introducing parts of one or more of the reactants in steps. For example, a portion of the total amount of the nitrogen
containing reactant may be added to a portion of the activator followed by a portion of
the phosphorus or sulfur containing reactant and then repeated in any order and in as
many steps as desired to obtain the final activated flame retardant.

The selection of the nitrogen containing reactant and the phosphorus or sulfur containing
reactant used can depend upon the application. Suitable reactants include those known
for use in forming nitrogenous phosphate or sulfonate flame retardants, for example,
ammonium phosphate, ammonium pyrophosphate, ammonium polyphosphate, ethylene-
diamine phosphate, piperazine phosphate, piperazine-pyrophosphate, melamine
phosphate, dimelamine phosphate, melamine pyrophosphate, guanidine phosphate,
dicyanodiamide phosphate urea phosphate, ammonium sulfonate, ammonium
polysulfonate, ethylenediamine sulfonate, dimelamine sulfonate, guanidine sulfonate, and
dicyanodiamide sulfonate. For example, suitable nitrogen containing reactants include
ammonium, alkenylenamines (including diamines), triazine, melamine, melam, melem,
melon, ammeline, ammelide, 2-ureido melamine, acetoguanamine, benzoguanamine,
guanidine, dicyanodiamide, diamine phenyltriazine or mixtures thereof. Preferred
nitrogen containing reactants include melamine, ammonium, and ethylene diamine.
Examples of suitable phosphorus or sulfur containing reactants include phosphoric acid
and sulfonic acid.

The char catalyst is a component that often enhances the performance of the nitrogenous
phosphate and/or sulfonate component and preferably is a char forming catalyst or a
phase transfer agent or a combination of both. The char catalyst can be present in the
flame retardant in any amount that provides the acceptable enhanced flame retardancy,
for example up to 5, 8 or 10 wt % and as little as 0.01, 0.1 and 0.2 wt %. It is preferred to
use both a char forming catalyst and a phase transfer catalyst together these may be
present in the flame retardant in an amount between 0.1, preferably 0.3, and 3.0,
preferably 2.5 wt %, based on the total weight of the flame retardant.

Although not wishing to be bound by any particular theory, it is believed that the char
forming catalyst may act, at the time of decomposition of part of the system, to "grab
onto" or react with decomposing molecules, thereby minimizing the production of low molecular weight components that can burn. This it is believed generally enables the development of char rather than burn. Consequently, exemplary char forming catalysts include multi-cyclic compounds having at least one reactive group in each of at least two rings. Typically, the rings are joined together by atoms common to both rings. These may include spiro-compounds. For example, the spiro compounds can define at least two heterocyclic ring structures, e.g., that each include oxygen, joined by at least one carbon atom common to both rings.

Preferred char forming catalyst include spiro-compounds represented by the following formula 1:

$$R_2-R_i-(R_o)m-R_i-R_2$$

where m represents a number between 1 and 8, for example less than 3 or less than 2; R_o -independently represent a di- tri-, or quad-valent radical including two independently substituted or unsubstituted, saturated or unsaturated heterocyclic ring structures joined by at least one common carbon atom and preferably no more than two, for example one, carbon atoms common to the heterocyclic ring structures; R_i —independently represents a bond; or a substituted or unsubstituted, saturated or unsaturated hydrocarbyl or heterocarbyl linking group, preferably a C_i-C_j alkyl linking group, for example a C_3 alkyl; and R_2 —independently represents a terminal group, preferably a terminal amine for example a primary amine.

Exemplary compounds include those in which the heterocyclic ring structure comprises at least two heteroatoms in at least two heterocyclic ring structures, and/or R_o independently represents a divalent radical, preferably with at least one, for example, two (including two adjacent), heterocyclic ring structures being 6-member. These compounds may also include those in which the heteroatom in the heterocyclic rings is predominately oxygen.

The preferred char catalyst includes those having a molecular weight of at least 180, preferably at least 200 and/or a flash point of greater than 200 °C.
Some preferred embodiments include one or more tetraoxaspiro materials, such as derivatives of a tetraoxaspiro undecane (e.g., amine derivatives), such as one or more 2,4,8,10-tetraoxa-spiro[5.5]undecane compounds and/or one or more 1,5,7,11-tetraoxa-spiro[5.5]undecane compounds.

The char forming catalyst component may also include adducts, for example, amine adducts, nitrile adducts (including 2-propenenitrile or acrylonitrile) and/or oxirane adducts (including butoxymethyl oxirane). 2,4,8,10-tetraoxa-spiro[5.5]undecane-3,9-dipropanamine with an amine or nitrile adduct, such as acrylonitrile is a specific example.

The phase transfer agent utilized can be or can include tetrahydrocarbyl ammonium salts, for example, tetramethyl, tetraethyl, tetrapropyl, tetralkyl, and/or aryltrialkyl ammonium salt in which the salt is a bromide, chloride, hydroxide and/or hydrogen sulfate ammonium salt. Preferably, the phase transfer catalyst includes phosphate esters, tetraethylammonium bromide, tetraethylammonium hydroxide, tetrabutylammonium bromide, tetrabutyl ammonium hydroxide, tetrabutyl ammonium hydrogen sulfate and/or benzyltriethyl ammonium chloride.

An exemplary retardant may be prepared as follows.

To 900 grams of water, while under agitation, add 180 grams of ethylene diamine, 60 grams of melamine and 0.5 wt percent by finished recovered product of 2,4,8,10-tetraoxa-spiro[5.5]undecane-3,9-dipropanamine adduct with 2-propenenitrile (acrylonitrile), which is commercially available from Ajinomoto, Inc. under the tradename YSE-CURE B-001. The mixture is warmed to 77 °C and, while maintaining temperature, phosphoric acid is slowly added until a pH of 7.0 is reached. The mixture is cooled, filtered, dried and ground to recover the flame retardant.

One of ordinary skill can easily modify the above technique to achieve a preferred ratio of 80:20 ethylene diamine phosphate to melamine phosphate.
This flame retardant can be present in amounts ranging from about 15 to about 85% by weight of the total composition. The char forming catalyst will therefore be present in the insulative wall in amounts ranging from about 0.15 weight percent to about 8.5 weight percent.

5

The compound described above can be described as the reaction product of epoxidized cardanol and ethanol. Other alcohols will work as well, as described in the following table:

<table>
<thead>
<tr>
<th>Plasticizer ID</th>
<th>Starting Material</th>
<th>Catalyst</th>
<th>Reactant</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ex – 2</td>
<td>NC-513® (370 g)</td>
<td>Phosphoric Acid (20.7 g)</td>
<td>t-BuOH (200 g)</td>
</tr>
<tr>
<td>Ex – 2</td>
<td>NC-513® (800 g)</td>
<td>Phosphoric Acid (0.8 g)</td>
<td>EtOH (800 mL)</td>
</tr>
<tr>
<td>Ex – 3</td>
<td>NC-513 (400 g)</td>
<td>Methanesulfonic Acid (10mL)</td>
<td>EtOH (800 mL)</td>
</tr>
<tr>
<td>Ex – 4</td>
<td>NC-513® (200 g)</td>
<td>Phosphoric Acid (6.25 g)</td>
<td>EtOH (200 g)</td>
</tr>
<tr>
<td>Ex – 5</td>
<td>NC-513® (300 g)</td>
<td>p-toluenesulfonic acid (3g)</td>
<td>EtOH (300 g)</td>
</tr>
<tr>
<td>Ex – 6</td>
<td>NC-513® (300 g)</td>
<td>None</td>
<td>Diethanol amine (200 g)</td>
</tr>
<tr>
<td>Ex – 7</td>
<td>NC-513® (600 g)</td>
<td>Phosphoric acid (19 g)</td>
<td>IPA (400 g)</td>
</tr>
<tr>
<td>Ex – 8</td>
<td>NC-513® (600 g)</td>
<td>p-toluenesulfonic acid (9g)</td>
<td>IPA (400 g)</td>
</tr>
<tr>
<td>Ex – 9</td>
<td>NC-513® (600 g)</td>
<td>p-toluenesulfonic acid (18g)</td>
<td>IPA (400 g)</td>
</tr>
</tbody>
</table>

IPA is isopropyl alcohol

Also disclosed is the reaction of the epoxidized cardanol with compounds having at least one amine group under a similar processing conditions. The preferred is diethanol amine as disclosed in experiment 6.
The following procedure was used to prepare the plasticizers:

The materials described in Table II were introduced into a 5 liter round bottom flask with stirring, heating, a thermometer and water condenser. The contents were kept under atmospheric reflux was for 4 hours. The final epoxy number in all cases was zero at the end of all reactions. There was no separation of the composition; the entire composition was used as the plasticizer.
CLAIMS

We claim

5. A plasticizer composition comprising the reaction product of a first compound which is an alcohol or an amine with second compound which is an epoxidized molecule having at least 15 carbon atoms and 1 to 3 double bonds in the epoxidized molecule.

2. The plasticizer composition of claim 1, wherein the epoxidized molecule is an epoxidized cardanol.

3. The plasticizer composition of claim 1, wherein the first compound is ethanol.

4. The plasticizer composition of claim 1, wherein the first compound is isopropyl alcohol.

5. The plasticizer composition of claim 1, wherein the alcohol is selected from the group consisting of ethanol, isopropyl alcohol, tert-butyl alcohol, and the amine is diethanol amine.

6. The plasticizer composition of claim 1, wherein the epoxidized molecule has a 15 carbon aliphatic chain in the meta position and is of the formula

\[
\text{O} \quad \text{O-CH}_2\text{-CH-CH}_2
\]

\[
\text{[CH}_2\text{]}_{7}\text{C}_7\text{H}_{10-14}\text{-CH}_3
\]
wherein the number of hydrogens in the 15 carbon aliphatic chain in the meta position depends upon the number of conjugated double bonds, the number of which are selected from the group consisting of 1, 2, and 3.

7. The plasticizer composition of claim 6, wherein the first compound is ethanol.

8. The plasticizer composition claim 6, wherein the first compound is isopropyl alcohol.

9. The plasticizer composition of claim 6, wherein the alcohol is selected from the group consisting of ethanol, isopropyl alcohol, tert-butyl alcohol, and the amine is diethanol amine.

10. The plasticizer composition of claim 1 wherein the plasticizer composition has been mixed with a thermoplastic compound.

11. The composition of claim 10, wherein the thermoplastic compound is polypropylene or a copolymer of polypropylene.

12. The composition according to claim 10, wherein the plasticizer is present from about 0.5 to about 12 weight percent of the total composition.

14. The composition according to claim 12, wherein the composition further comprises a flame retardant composition.

15. A plasticizer composition comprising a compound of the formula:
where $R_i$ is a 15 carbon atom aliphatic chain containing one, two or three non-conjugated double bonds and has the formula $-\left[\text{CH}_2\right]_7\text{-C}_7\text{H}_{10}\text{-i}_4\text{-CH}_3$ and $R_2$ is selected from the group consisting alkyls, and aryls optionally having one or more functional groups, and the oxygen is bonded to any carbon of $R_2$.

17. The plasticizer of claim 16, wherein $R_2$ is an alkyl without a functional group.

18. The plasticizer of claim 17, wherein $R_2$ has the formula $-\text{C}_n\text{H}_m$, where $n$ is an integer ranging from 1 to 10 and $m$ is an integer ranging from 3 to 21 and the bond of the oxygen to the carbon of $R_2$ may be any carbon of $R_2$.

19. A plasticizer of claim 16, wherein $R_2$ is an alcohol selected from the group consisting of ethanol, the isomers of propanol and the isomers of butanol without the OH group.

20. A plasticizer of claim 16 having the formula of:
21. The plasticizer of claim 15; wherein the plasticizer has been mixed with a thermoplastic and a flame retardant composition and the flame retardant composition comprises a compound selected from the group consisting of a melamine, a diamine, magnesium hydroxide and aluminum hydroxide.

22. The plasticizer of claim 16; wherein the plasticizer has been mixed with a thermoplastic and a flame retardant composition and the flame retardant composition comprises at least one compound selected from the group consisting of a melamine, a diamine, magnesium hydroxide and aluminum hydroxide.

23. The plasticizer of claim 17; wherein the plasticizer has been mixed with a thermoplastic and a flame retardant composition and the flame retardant composition comprises at least one compound selected from the group consisting of a melamine, a diamine, magnesium hydroxide and aluminum hydroxide.

24. The plasticizer of claim 18; wherein the plasticizer has been mixed with a thermoplastic and a flame retardant composition and the flame retardant composition comprises at least one compound selected from the group consisting of a melamine, a diamine, magnesium hydroxide and aluminum hydroxide.
25. The plasticizer of claim 19; wherein the plasticizer has been mixed with a thermoplastic and a flame retardant composition and the flame retardant composition comprises at least one compound selected from the group consisting of a melamine, a diamine, magnesium hydroxide and aluminum hydroxide.

26. The plasticizer of claim 20; wherein the plasticizer has been mixed with a thermoplastic and a flame retardant composition and the flame retardant composition comprises at least one compound selected from the group consisting of a melamine, a diamine, magnesium hydroxide and aluminum hydroxide.

27. A process for the manufacture of a plasticizer compound comprising the steps of heating a mixture of an epoxidized oil and a first compound selected from the group consisting of alcohols and amines, in the presence of an acid catalyst and reacting the epoxidized oil with the first compound until at least less than 10% of epoxidized oil remains, and removing the excess alcohol.

28. The process of claim 27, wherein the epoxidized oil is epoxidized cardanol.

29. The process of claim 28, wherein the epoxidized cardanol comprises at least one compound having a 15 carbon aliphatic chain in the meta position with the compound having the formula

\[
\begin{align*}
\text{O} & \quad \text{CH}_2\text{CH} \quad \text{CH}_2 \\
\text{O} & \quad \text{CH}_2 \quad \text{CH} \quad \text{CH}_2 \\
& \quad \text{[CH}_2\text{]}_{7} \text{C}_7\text{H}_{10-14} \quad \text{CH}_3
\end{align*}
\]
and the number of hydrogens in the 15 carbon aliphatic chain in the meta position
depends upon the number of conjugated double bonds, the number of which are selected
from the group consisting of 1, 2, and 3.

30. The process of claim 29, wherein the alcohol is selected from the group consisting of
alkyls and aryls optionally having one or more functional groups in additional to having
at least one hydroxyl group.

31. The process of claim 27, wherein the alcohol is selected from the group consisting of
ethanol, the isomers of propanol and the isomers of butanol.

33. The process of claim 28, wherein the heating is done at reflux conditions.

34. The process of claim 29, wherein the heating is done at reflux conditions.

35. The process of claim 30, wherein the heating is done at reflux conditions.

36. The process of claim 31, wherein the heating is done at reflux conditions.
FIG. 1
FIG. 2
**A. CLASSIFICATION OF SUBJECT MATTER**

INV. C07C43/23  C07C41/03  C08K5/06  C07C43/166

According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)

C07C  C08K

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, COMPENDEX, CHEM ABS Data

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

<table>
<thead>
<tr>
<th>Category</th>
<th>Citation of document, with indication, where appropriate, of the relevant passages</th>
<th>Relevant to claim</th>
</tr>
</thead>
<tbody>
<tr>
<td>X</td>
<td>US 2 434 797 A (THE HARVEL CORP.) 20 January 1948 (1948-01-20) column 2, line 51 - line 54 examples 1,2</td>
<td>1-10, 15-19</td>
</tr>
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<td>X</td>
<td>US 7 084 103 Bl (SPRINGSTED PERRY [US] ET AL) 1 August 2006 (2006-08-01) column 3, line 5 - line 40; compound I column 3, line 46 - line 48 claim 1</td>
<td>1-9, 15-19</td>
</tr>
<tr>
<td>X</td>
<td>US 3 074 900 A (DAVID WASSERMAN) 22 January 1963 (1963-01-22) column 2, line 13 - line 24 examples claims 1,2</td>
<td>1-10, 12, 15-19</td>
</tr>
</tbody>
</table>

Further documents are listed in the continuation of Box C

See patent family annex

* Special categories of cited documents
  *A* document defining the general state of the art which is not considered to be of particular relevance
  *E* earlier document but published on or after the international filing date
  *L* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
  *O* document referring to an oral disclosure, use, exhibition or other means
  *P* document published prior to the international filing date but later than the priority date claimed

IT' later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

IX' document of particular relevance, the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

*Y* document of particular relevance, the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

*S* document member of the same patent family

Date of the actual completion of the international search

10 March 2010

Date of mailing of the international search report

18/03/2010

Name and mailing address of the ISA/

European Patent Office, P B 5818 Patentlaan 2
NL - 2280 HV Rijswijk
Tel: (+31-70) 340-2040,
Fax: (+31-70) 340-3016

Authorized officer

Russel l, Graham
<table>
<thead>
<tr>
<th>Category</th>
<th>Citation of document, with indication, where appropriate, of the relevant passages</th>
<th>Relevant to claim No</th>
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