HEPTANONIC ACID AND FURANONE COMPOUNDS AND COMPOSITIONS

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ABSTRACT

A natural inexpensive scyethyl monoterpenic ketone (dihydroagrolone) of formula (4), isolated from the oil of Tagetes sp., was smoothly oxidized with meta-periodate/potassium permanganate into 2,6-dimethyl-4-oxo-heptanoic acid of formula (3), the reduction of 3 with metal hydride such as sodium borohydride or lithium aluminium hydride provided 4-hydroxyacid of formula (2) which on without isolation undergone lactonisation in acidic medium furished two chiral centered 5-isobutyl-3-methyl-4,5-di-dihydro-2(3H)-furane of formula (1) as an analogue of whisky lactone 5-butyl-4-methyl-4,5-dihydro-2(3H)-furane of formula (1a) responsible for high quality of alcoholic beverage (whisky, wine, brandy and scotch), in addition, coconut flavoured 5-butyl-4-methyl-4,5-dihydro-2(3H)-furane of formula (1) is also as an analogue of coconut aldehyde (γ-nonolactone, F.E.M.A. No. 2751) of formula (1b) which is responsible for flavouring a wide range of food stuffs including baked goods and confectionery.
$^1$H NMR (300 MHz) spectra of 2,6-dimethyl-4-oxo-heptanoic acid in CDCl$_3$

FIG. 1
FIG. 2

C-NMR (75.4 MHz) spectra of 2,6-dimethyl-4-oxo-heptanoic acid in CDCl3
$^1$H NMR (300 MHz) spectra of 5-isobutyl-3-methyl-4,5-dihydro-2(3H)-furanone

FIG. 3
C-NMR (75.4 MHz) spectra of 5-isobutyl-3-methyl-4,5-dihydro-2 (3H)-furanone in CDCl3.

FIG. 4
GC of 5-isobutyl-3-methyl-4,5-dihydro-2(3H)-furanone

FIG. 5
Mass spectra of 5-isobutyl-3-methyl-4,5-dihydro-2(3H) furanone.

FIG. 6
HEPTANONIC ACID AND FURANONE COMPOUNDS AND COMPOSITIONS

This invention relates to a novel method for converting dihydroacetone, a bifunctional acyclic monoterpenic ketone, isolated from plants of Tagetes species into coconut flavoured 5-isobutyl-3-methyl-4,5-dihydro-2(3H)-furanone with two chiral centers of formula (1). Interestingly, the compound (1) is found as a novel analogue of whisky lactone (also named as Quercus lactone or oak lactone) of formula (1a), which is contained, for example, in high quality alcoholic beverages (Napoleonic whisky, Jamaica and cognac etc.), as fragrance ingredient, in insect sex pheromones and several natural products containing this structural moiety. Moreover, it is found in coconut flavoured compound (1) is also an analogue of coconut aldehyde (γ-nonanalactone or aldehydic C-18, FEMA No. 2751) of formula (1b) which is responsible for flavouring a wide range of foodstuffs including baked goods and confectionery.

(1) No. of chiral centers=2; (1a) No. of chiral centres=2; (1b) No. of chiral center=1

The present invention also relates to 2,6-dimethyl-4-oxoheptanonic acid of formula (3) not only as a precursor for the synthesis of whisky lactone of formula (1a) but also as a novel analogue of 2,6-dimethyl-5-oxoheptanonic acid (a constituent of well known essential oil of Mentha x piperita) which has wide applications in flavouring foodstuffs, soft and alcoholic beverages, perfumery and pharmaceutical industries.

BACKGROUND OF THE INVENTION


In particular, two chiral centered γ-butyrolactones having substituents such as an alkyl or alkenyl group are in more demand and they exist as either cis and/or trans isomer in nature (Bryan, V. and Chan, T. H., Tetrahedron Lett., 37(30), 5341-5342 (1996)). For example, whisky lactone, chemically known as 5-butyl-4-methyl-dihydro-2(3H)-furanone (also known as 3-methyl-4-octanolic or 4-hydroxy-3-methyloctanolic acid lactone) (1a) is a natural optically active disubstituted butyrolactone which is widely recognized and it has been identified as the most valuable additive responsible for the flavor of high quality whisky, wine, cognac and scotch etc (Nishimura, K. and Masuda, M., J. Food Sci. 36, 819 (1971); Kepner, R. E., Webb, A. D. and Muller, C. J., Am. J. Enol. Viticult, 23, 144 (1972); Etienne, M. and Manfred, S., Tetrahedron Lett., 25(40), 4491-4494 (1984) and Pérez-coello, M. S., Sanchez, M. A., Garcia, E., Gonzalez-vinas, M. A., Sanz, J. and Cabezaudo, M. D., J. Agric. Food Chem., 48, 885-889 (2000)). Whisky lactone (1a) (also known as oak lactone or Quercus lactone) is a diastereomer of 3-methyl-4-octanolides (5-butyl-4-methyl-dihydro-2(3H)-furanone) which is found in whisky, wine,
brandy and in extract of oak barrels. This lactone is important constituent of aging flavour and have so far been considered to be formed during aging from unknown precursors in wood barrels (Chem Abstr. 114, 181996w, 1991) which finally contributes to the flavour impact of the oak. The effect of oak barrels on the quality of wines are also influenced by several factors such as tree age, growth rate of tree and barrel size etc (Towey, J. P. and Waterhouse, A. L., Am. J. Enol. Viticult., 47(1), 1–20 (1996)).

In particular, the quality of the alcoholic beverages is affected largely by the presence of whisky lactone which is produced during the aging process. A wine aged for a longer period of time is realized to be of higher quality and is further influenced by several factors such as climatological conditions, fermentation, distillation, maturation and blending (Plaza, E. G., Munoz, R. G., Roa, J. M. L. and Martinez, A., J. Agric., Food Chem., 48, 736–741 (2000)). One of the isomers of this interesting whisky lactone (1a) is identified for the first time in whisky by Soumalainen and Nykanen in 1969 (Soumalainen, H. and Nykanen, L., Nährungsmittelindustrien, 23, 1–15 (1970), however, later on cis and trans isomers have been determined by several workers in matured cognac, Jamaican rum and whiskies and found that the aroma threshold for the cis isomer is 92 ppb while for the trans-isomer is 460 ppb (Masuda, M. and Nishimura, K., Phytochemistry, 10, 1401–1402 (1971); Gunther, C. and Mosandl, A., Liebigs Ann. Chem. 2112–2122, (1986); Gunther, C. and Mosandl, A., Z. Lebensm. Unters. Forsch., 185, 1–4 (1987) and Maarse, H. and Berg F. V. D. In Understanding Natural Flavours, edited by Piggott, J. R. and Paterson, A.; Blackie Academic & Professional, New York, pp. 243–267 (1994)). In fact, four stereoisomers of whisky lactone (1b) are known with the following taste and odor such as (3R,4R)-5-butyl-4-methyl-dihydro-2(3H)-furanone with sweet, fresh, bright coconut smell (10% ethanol) and creamy, soft, coconut taste (10 ppm in aqueous saccharin); (3S,4S)-5-butyl-4-methyl-dihydro-2(3H)-furanone with weak coconut note earthy, moldy afterodor, hay note odor and herbaceous, reminiscent of coconut taste; (3S,4R)-furanone derivative with spicy celery note, weak coconut, distinct green walnut note odor and sweet creamy fatty, reminiscent of coconut; (3R,4S)-furanone derivative with intensive coconut note, afterodor, reminiscent of celery odor and weak spicy, herbaceous taste (Schreier, P., In: Bioflavour '87, Analysis. Biochemistry Biotechnology, Proceedings of the International Conference, Würzburg, Federal Republic of Germany, Sep. 29–30, 1987, Walter de Gruyter, New York, 55–74, (1988) and Eric, M., Raymond, B., Christine, L.G. and Jean-Louis, P., J. Agric.Food Chem., 48, 4306–4309, (2000)). A mixture of cis- and trans-whisky lactones also exhibits a repellent activity against mosquitoes and flies (Shono, Y. and Tsukasa, Y., Nippon Kokai Tokkyo Koho, 63-8203). However, considering all the above factors, it was realized that the productivity of oak lactone by aging whisky in oak barrel for several months/years in controlled temperature and other conditions is a laborious job, therefore, synthesis of whisky lactone (1a) has been developed by several chemists and some of the reported conventional methods include the following:


It, therefore, becomes an object of the invention to eliminate the disadvantages existing with above known methods and others and to provide a rapid and economical method for the preparation of γ-butyrolactone of formula (1) as an analogue of whisky lactone (nicknamed as Quercus lactone or oak lactone) of formula (1a), via oxidation of dihydrodacetone (4) into 2,6-dimethyl-4-oxo-heptanoic acid (3) followed by reduction of 3 and lactonisation into γ-butyrolactone (1). In addition, γ-butyrolactone (1) with coconut flavour can also find application in flavouring the food stuffs since its analogue i.e. coconut aldehyde (γ-nonalactone, F.E.M.A. No. 2751) (1b) is responsible for flavouring a wide range of food stuffs including baked goods and confectionery.

OBJECTS OF THE INVENTION

The main object of the present invention is to provide a novel compound 5-isobutyl-3-methyl-4,5-dihydro-2(3H)-furanone of formula (1) as an analogue of whisky lactone or oak lactone or quercus lactone of formula (1a).

Yet another object of the invention is to provide a process for the preparation of compound 5-isobutyl-3-methyl-4,5-dihydro-2(3H)-furanone of formula (1) starting from dihydrodacetone of formula (4) which is, in fact, a natural acyclic monoterpen ketone (dihydroacetone) found readily in Tagetes species.

Yet another object of the invention is to provide a simple process for industrial scale purification of starting material i.e. dihydrodacetone of the formula (4) from oil of Tagetes species.

Yet another object of the invention is to provide a simple process for proper utilization of dihydrodacetone of the formula (4) into high valued products.

Yet another object of the invention is to provide a simple and efficient oxidizing reagent which utilizes the double bond of dihydrodacetone to generate a novel 2,6-dimethyl-4-oxo-heptanoic acid of formula (3) with one chiral center.

Yet another object of the invention is to provide a rapid method of converting dihydrodacetone of the formula (4) into 2,6-dimethyl-4-oxo-heptanoic acid of formula (3) in microwave oven ranging from 5–30 minutes.

Yet another object of the invention is to accelerate oxidation of double bond of dihydrodacetone of the formula (4) into 2,6-dimethyl-4-oxo-heptanoic acid of formula (3) by using alumina as a catalyst.

Yet another object of the invention is to provide a simple process for the preparation of 2,6-dimethyl-4-oxo-heptanoic acid of formula (3) in high yield for its use as a multipurpose additive in food and tobacco products since its analogue such as 2,6-dimethyl-5-oxo-heptanoic acid (found as a constituent of an important Italo-Milcham peppermint oil i.e. Mentha spicata L.) is well known for its wide application in perfumery, flavour and pharmaceutical industry.

Yet another object of the invention is to convert 2,6-dimethyl-4-oxo-heptanoic acid of formula (3) into corresponding 4-hydroxy acid or 4-oxo ester or the like.

Yet another object of the invention is to provide an industrial method for the preparation of 5-isobutyl-3-methyl-4,5-dihydro-2(3H)-furanone of formula (1) in high yield and purity via reduction of above obtained 2,6-
dimethyl-4-oxo-heptanoic acid of formula (3) with metal hydride followed by lactonisation into 5-isobutyl-3-methyl-4,5-dihydro-2(3H)-furanone of formula (1).

Yet another object of the present invention is to introduce two chiral centred 5-isobutyl-3-methyl-4,5-dihydro-2(3H)-furanone of formula (1) since it is well established that chiral discrimination is an important principal of odor perception.

Yet another object of the invention is to provide 5-isobutyl-3-methyl-4,5-dihydro-2(3H)-furanone of formula (1) as a new product for the first time for its wide application in flavours, perfumery, cosmetics, soft and alcoholic beverages and pharmaceutical industries for its biological activities.

Yet another object of the invention is to provide coconut flavoured 5-isobutyl-3-methyl-4,5-dihydro-2(3H)-furanone of formula (1) for its use as a substitute of several other coconut flavoured γ-butyrolactones for its application in food and beverage or the like.

Yet another object of the invention is to provide an analogue of coconut aldehyde (aldehyde C-18) of formula (1b) which is one of the most frequently used lactones in perfumery and flavour industries (F.E.M.A. No. 2781).

SUMMARY OF THE INVENTION

The present invention provides a method for preparing 3,5-disubstituted γ-butyrolactone, via which two chiral centered lactone of formula (1) with coconut flavour, an analogue of natural whisky lactone (also named as Quercus lactone or oak lactone) of formula (1a) responsible for high quality of whisky, wine, brandy and scotch is formed by utilizing a natural inexpensive acyclic monoterpenic ketone (dihydroactagone) of formula (4), isolated from the oil of Tagetes sp., which was smoothly oxidized with periodate/potassium permanganate to 2,6-dimethyl-4-oxo-heptanoic acid of formula (3). The reduction of oxo acid (3) with sodium borohydride followed by lactonisation in acidic medium furnished a new two chiral centered 5-isobutyl-3-methyl-4,5-dihydro-2(3H)-furanone of formula (1) which has wide applications in flavouring food stuffs, soft and alcoholic beverages, perfumery and pharmaceutical industries.

The present invention also relates to 2,6-dimethyl-4-oxo-heptanoic acid of formula (3) not only as a precursor for the synthesis of whisky lactone of formula (1a) but also as a novel analogue of 2,6-dimethyl-5-oxo-heptanoic acid (a constituent of well known essential oil of Mentha spicata) which has wide applications in flavouring food stuffs, soft and alcoholic beverages, perfumery and pharmaceutical industries.

BRIEF DESCRIPTION OF THE ACCOMPANYING DRAWING

FIG. 1 is 1H NMR (300 MHz) spectra of 2,6-dimethyl-4-oxo-heptanoic acid (in CDCl₃) of the reaction product of Example I containing the compound having the structure: #STR1##.

FIG. 2 is 13C NMR (75.4 MHz) spectra of 2,6-dimethyl-4-oxo-heptanoic acid (in CDCl₃) of the reaction product of Example I containing the compound having the structure: #STR1##.

FIG. 3 is 1H NMR (300 MHz) spectra of 5-isobutyl-3-methyl-4,5-dihydro-2(3H)-furanone (in CDCl₃) of the reaction product of Example II containing the compound having the structure: #STR2##.

FIG. 4 is 13C NMR (75.4 MHz) spectra of 5-isobutyl-3-methyl-4,5-dihydro-2(3H)-furanone (in CDCl₃) of the reaction product of Example II containing the compound having the structure: #STR2##.

FIG. 5 is the GC of 5-isobutyl-3-methyl-4,5-dihydro-2(3H)-furanone of the reaction product of Example II containing the compound having the structure: #STR2##.

FIG. 6 is the electro spray (ES) mass spectrum of 5-isobutyl-3-methyl-4,5-dihydro-2(3H)-furanone (MW 156) of the reaction product of Example II containing the compound having the structure: #STR2##.

Accordingly, the present invention provides “a novel method for converting dihydroactagone into coconut flavoured 5-isobutyl-3-methyl-4,5-dihydro-2(3H)-furanone with two chiral centers: an analogue of natural whisky lactone and coconut aldehyde” of the general formula (1), namely:

the said method comprising:

a. adsorbing Tagetes minuta oil on an adsorbent selected from but not limited to alumina and silica gel;
b. loading adsorbent obtained from step (a) on a soxhlet extractor and washing with a non polar solvent (cold percolation) selected from but not limited to petroleum ether and hexane for removing the non polar constituents;
c. isolating the dihydroactagone rich fractions using hot hexane percolation with increasing percentage of ethyl acetate starting from 0.2 up to 5%;
d. concentrating the above separated fractions under reduced pressure to obtain a dihydroactagone rich concentrate;
e. loading above concentrate on alumina or silica gel column and eluting the column with a non polar solvent selected from but not limited to hexane and petroleum ether;
f. eluting the column further with increasing percentage of ethyl acetate up to 5% in hexane;
g. concentrating the resulting hexane-ethylacetate fractions under reduced pressure to obtain dihydroactagone of formula (4) in 99% or more purity;
h. providing dihydroactagone of formula (4) in organic solvent selected from the group consisting of ether selected from but not limited to tetrahydrofuran, dimethoxyethane and dioxane, alcohol selected from but not limited to butanol and t-butyl alcohol, ketone selected from but not limited to acetone and methyl ethyl ketone;
i. oxidising dihydroactagone of formula (4) in the presence of an oxidizing reagent selected from but not limited to sodium meta-periodate, potassium meta-periodate and ruthenium tetroxide in the range of 2 to 8 times mole of dihydroactagone of formula (4) and a co-oxidizing agent selected from but not limited to potassium permanganate and sodium permanganate in the range of 0.1 to 0.6 times mole of dihydroactagone (starting material) of formula (4);
j. adding a base selected from the group consisting of but not limited to sodium carbonate, sodium bicarbonate, potassium carbonate, lithium carbonate and sodium hydroxide...
in the range of 1 to 4 times mole of dihydrotagetone of formula (4) to the above said solution obtained in step (i);
k. maintaining the temperature between -15°C. to +90°C. during oxidation steps from (h) to (j) for a period ranging
followed by extraction and chromatography to obtain 2,6-
dimethyl-4-oxo-heptanoic acid of formula (3) with 68% yield or more;
n. providing 4-oxo acid of formula (3) under stirring in a
solvent selected from the group consisting of ether but not
limited to tetrahydrofuran, dimethoxyethane and dioxane,
alcohol but not limited to methanol, butanol and t-butyl
alcohol, chlorinated solvent but not limited to dichloro-
rane and chloroform;
o. adding a base selected from the group consisting of
but not limited to sodium bicarbonate or sodium hydroxide or
potassium carbonate to the resultant solution to make salt of
the oxy acid of formula (3);
p. reducing salt of oxo acid of formula (3) to hydroxy acid
of formula (2) in the presence of a reducing agent selected
from but not limited to sodium borohydride or lithium aluminum hydride in the range of 0.5–3 times mole at
a reaction temperature varying from -15 to 50°C;
q. removing the organic solvent under vacuum and acidifying
the cooled aqueous alcoholic solution at -10°C. to 0°C.
with an acid selected from the group but not limited to
hydrochloric acid, sulfuric acid, acetic acid, acetic anhy-
dride for a period ranging between 30 minutes to 8 hours
and finally isolating the product 5-isobutyl-3-methyl-4,5-
dihydro-2(3H)-furanone also known as 4-isobutyl-2-
methyl-4-butanolide) of formula (1) in 76% or more yield
by extraction and chromatography in usual manor.

In an embodiment of the present invention Soxhlet extrac-
tor using cold hexane percolation removes oelimine, one
of the main and unwanted constituent, from tagetes oil.

In an embodiment of the present invention Soxhlet extrac-
tion of alumina or silica gel bound tagetes oil with hot
percolation of non polar solvent removes polymerized material efficiently since it remains stuck on alumina or silica.

In an embodiment of the present invention a simple process is employed for the industrial scale purification of
dihydrotagetone of formula (4) from oil of Tagetes species.

In an embodiment of the present invention a simple process is employed for the proper utilization of dihydrotagetone of formula (4) into high valued products.

In an embodiment of the present invention said process is capable of preparing 2,6-dimethyl-4-oxo-heptanoic acid of formula (3) on commercial scale.

In an embodiment of the present invention 2,6-dimethyl-
4-oxo-heptanoic acid of formula (3) has acidic sweet aroma.

In an embodiment of the present invention compound 2,6-dimethyl-4-oxo-heptanoic acid of formula (3) has bifunctionality which is capable of converting compound 3 into several value added products.

In an embodiment of the present invention an alternative process is employed for the rapid conversion of dihydrotagetone of formula (4) into 2,6-dimethyl-4-oxo-heptanoic acid (3) in microwave oven ranging from 5–30 minutes where the rate of reaction is further accelerated by adding alumina.

In an embodiment of the present invention is to provide a simple process for preparation of 2,6-dimethyl-4-oxo-
heptanoic acid of formula (3) as an analogue of 2,6-
dimethyl-5-oxo-heptanoic acid which is found as a constituent of an important Italo-Mitcham peppermint oil i.e. Mentha x piperita L. and well known for its wide application in perfumery, flavour and pharmaceutical industry.

In an embodiment of the present invention 2,6-dimethyl-
4-oxo-heptanoic acid of formula (3) is a novel compound and useful as a chiral synthone for its application in asymmetric chemistry.

In an embodiment of the present invention compound 2,6-dimethyl-4-oxo-heptanoic acid of formula (3) is useful as a versatile intermediate for converting it into corresponding 4-oxo ester, 4-hydroxy ester and 4-hydroxy acid for various formulations in flavour, perfumery and pharmaceutical industries.

In an embodiment of the present invention compound 2,6-dimethyl-4-oxo-heptanoic acid of formula (3) is useful for various formulations in flavouring food stuffs, soft and alcoholic beverages, herbal and tobacco products, perfumery and pharmaceutical aids in desired and acceptable amount varying from ppm to mg based on the type of products.

In an embodiment of the present invention a simple process is employed for the preparation of novel 5-isobutyl-
3-methyl-4,5-dihydro-2(3H)-furanone of formula (1) in high yield and high purity.

In an embodiment of the present invention a simple process is employed for the preparation of novel 5-isobutyl-
3-methyl-4,5-dihydro-2(3H)-furanone of formula (1) as an analogue of whisky lactone or oak lactone or quercus lactone.

In an embodiment of the present invention compound 5-isobutyl-3-methyl-4,5-dihydro-2(3H)-furanone of formula (1) is capable of providing aroma of whisky lactone to high quality alcoholic beverages which generally comes after aging wine, brandy scotch and whisky in a specific oak barren for several months/years, however, this invention removes all the hassle.

In an embodiment of the present invention compound 5-isobutyl-3-methyl-4,5-dihydro-2(3H)-furanone of formula (1) is capable of undergoing conversion into several value added products including furans and pyrrolidones etc.

In an embodiment of the present invention compound 5-isobutyl-3-methyl-4,5-dihydro-2(3H)-furanone of formula (1) is a stable compound.

In an embodiment of the present invention compound 5-isobutyl-3-methyl-4,5-dihydro-2(3H)-furanone of formula (1) has coconut like aroma.

In an embodiment of the present invention compound 5-isobutyl-3-
methyl-4,5-dihydro-2(3H)-furanone of formula (1) is an
analogue of whisky lactone (1a) is useful in formulation of high quality alcoholic beverages including wine, brandy scotch and whisky.

In an embodiment of the present invention coconut aroma of 5-isobutyl-3-methyl-4,5-dihydro-2(3H)-furanone of formula (1) is useful as multipurpose additive for various formulations in flavouring food stuffs, soft and alcoholic beverages, herbal and tobacco products, perfumery and pharmaceutical aids in desired and acceptable amount varying from ppm to mg based on the type of products.

In an embodiment of the present invention a simple process is employed for the preparation of novel 5-isobutyl-
3-methyl-4,5-dihydro-2(3H)-furanone of formula (1) since an analogue of coconut aldehyde (commonly known as gamma lactone) (1b) is one of the most frequently used lactones in perfumery and flavour industries (F.E.M.A. No. 2781).

**DETAILED DESCRIPTION OF THE INVENTION**

Alcoholic beverages contain various kinds of flavour compounds with flavour ranging from strong to mild
In particular, the quality of the alcoholic beverages is affected largely by the source of material, enzymatic action, process, storage, transportation and aging process (Simpson, P., J. Vitam. Nutr., 17, 274–287 (1978) and Maga, J. A., Food Rev. Intern., 5, 39–99, (1989)). Aging techniques are widely used to produce wine, whisky, miso and fermented foods and special aging methods are devised for each product (Paterson, A. and Piggott, J. R., In: The Distilled Beverage Flavour, recent Developments, Ellis Horwood, Chichester, 151–169 (1989)). Oak barrels have been used to store and age wine and other beverages for centuries for enhancing the flavour of the beverage by incorporating flavour components from oak into the beverages in which cis-oak lactone has a superior scent than trans-isomer (Otsuka, K., Zenibayashi, Y., Itoh, M. and Totsuka, A., Agric. Biol. Chem., 38, 485, (1974)). White oak obtained from United States and France is of highest quality as these barrels do not leak and finally produce high quality wines including sherries as well as distilled spirits—principally whiskies, bourbon and scotch although other countries do produce some oak barrels too. Some oak species found in France and across Europe are Quercus pedunculata (Q. robur), Q. sessilis (Q. petraea or Q. sessiliflora) while Q. alba, Q. cerris are found through out United States (Alamo, M. D., Bemal, J. L. and Cordoves, C. G., J. Agric. Food Chem., 48, 4613–4618 (2600)). However oak lactone (whisky lactone) is found in traces in plant kingdom and for these reasons several methods of preparation of whisky lactone have been reported and some of the important methods are:

1) 1,4-addition of lithium dimethylcuprate to the butanolate derivative, but this is not a convenient synthesis (Vigneron, J. P., Meric, R. and Dhaenens, M., Tetrahedron Lett., 21, 2057 (1980) as it requires expensive reagents.

2) A method utilizing substituted carboxylic acid and butylene oxide derivatives as a starting material, however, this method requires highly flammable lithium metal to complete the series of reaction, therefore, not applicable on an industrial scale. Moreover, the starting materials i.e acid and oxide derivatives need to be synthesized in several steps (Moret, E. and Schlosser, M., in Tetrahedron Lett., 25: 4491–4494, (1984) and Fujita, T., Watanabe, S. and Suga, K. in Aust. J. Chem., 27: 2205–2208, (1971)).

3) A method using acyclic vinyl sulfoxide as a starting material involves many steps and several expensive reagent combinations (Marino, J. P. and Pradilla de la R., R.F., Tetrahedron Letters, 26(44), 5381–5384 (1985)).

4) A method utilizing ribonolactone as a starting material, however, ribonolactone is expensive and the reaction steps are complicated (Ortuno, R. M., Merce, R. and Font, J., Tetrahedron, 43, 4497–4506 (1987)).

5) A method utilizing optically active lactone as a starting material, however, lactone is expensive and the reaction steps are complicated (Bloch, R. and Gilbert, L., J. Org. Chem. 52, 4603–4605 (1987)).

6) A method utilizing 2-butenyl carbonate as a starting material, however, this method requires the use of specific reagent such as (–) sparteine but even then the optical purity of lactone derivative obtained is not sufficiently high (Hoppe, D. and Zschage, O., Angew. Chem. Int. Ed. Engl., 28, 69–71 (1989)).

7) A method using chiral methyl-β-formyloxalylate derivative as a starting material involves many steps and several expensive reagent combinations (Thomas, K., Agnes, J. and Hans, U. R., Chem. Ber., 122, 2165–2175 (1989)).

8) A method using cyclopentane derivative as a starting material involves many steps and several expensive reagent combinations (Suzuki, Y., Wakako, M., Hiyoduki, I., Koichi, N. and Toshio, H., Tetrahedron Letters, 33(34), 4931–4932 (1992)).

9) A method using a naturally occurring geraniol as a starting material involves many steps and several expensive reagent combinations (Taber, D. F. and Jonathan, B. H., J. Org. Chem., 59, 4004–4006 (1994)).

10) A method using levoglucosenone is made to react with methyl lithium in the presence of copper iodide in order to introduce a methyl group into an enone group of levoglucosenone and to obtain 1,6-anhydro-3,4-dideoxy-4-C-methyl-beta-D-erythro-b-hexopyranos-2-olose. This methyl compound is oxidized in acetic acid for lactone formation and (3S, 48)-5-hydroxy-3-methylpentan-4-olide is thus obtained. Further, this lactone is made to react with tosyl chloride in anhydrous pyridine to obtain (3S, 48)-3-methyl-5-tosylxypentan-4-olide (tosylate). The obtained tosylate is alkylated with n-propyl lithium in the presence of copper iodide to obtain (3S, 48)-3-methyl-5-tosylxypentan-4-olide. Here, the (3S, 48)-3-methyl-5-tosylxypentan-4-olide may be treated with potassium carbonate to cleave lactone once, thereby obtaining epoxide. After that, the obtained epoxide is alkylated to form lactone ring again to obtain (3S, 48)-3-methyl-5-tosylxypentan-4-olide (whisky lactone) (1a), however, this method has various limitations and can not be upscaled on industrial basis (Takashi, E., Katsuya, M., Hajime, Y., Koshi, K., Hiroshi, K. and Hajime, M., Heterocycles, 31(9), 1585–1588 (1990) and Takashi, E., Hajime, M., Hiroshi, K. and Koshi, K., U.S. Pat. No. 5,216,177 (1993)).

All the above methods including patent have various limitations and none of them have been found suitable for the economical production of whisky lactone (4-butyl-3-methyl-butyrolactone) (1a) and therefore, it was realized that any synthetic analogue of (4-isobutyl-2-methyl-butyrolactone or 5-sec-butyl-3-methyl-4,5-dihydro-2H-furanone) (1) that can be prepared more readily. In seeking a simple synthesis of (1) from a cheaper material and reagents, dihydrogeratone (4) (isolated from Tagetes oil) (Lawrence, B. M., Powell, R. H., Smith T. W. and Kramer, S., Perfumer and Flavorist, 10(9), 56–58 (1985)) appears as a simple and economical starting material in which dihydrotagetone undergoes oxidation and reduction followed by lactonization to afford γ-butyrolactone (1) which, in fact, would offer the advantages of simplicity and directness and can be applied for large scale preparations. In addition, this new butyrolactone (1) may have better applications than whisky lactone (1a) since in many cases the structural modifications towards analogue formations are proved to be more biologically active and useful than parent compounds (William, D. K., Jeffrey, C. B., Dalia, R. J., Kenneth, G. H., Sidney, M. H., Gregory, G., Mary, J. C., Francis, L. M., Lee, F. F., Randall, K. J. and Robert, P. H., J. Med. Chem., 34, 98–107 (1991); Xuechao, W., Yuxin, C., Xinfu, P. and Yaozu, C., J. Indian Chem. Soc., 73, 217–219 (1996); Vivek, K., Thomas, F. M. and Jane, V. A., J. Med. Chem. 43, 5050–5054 (2000) e.g a δ-nonalactone (mol. formula= C16H16O2, six membered lactone), an isomer of the well known γ-nonalactone (mol. formula= C13H16O2, five membered lactone) (1b) is commercially available and having extensive use in perfumery compositions as a modifier for coconut aldehyde (Flavour and Extract Manufactures Association, F.E.M.A. No. 2781) (1a) (Steffen, A., In: Perfume and Flavor Chemicals, Part II, Allured Publishing Corporation, Carol Stream, Ill. 60188–2787, (1994)).

Dihydrotangetone (2,7-dimethyl-oct-1-en-5-one or 2,6-dimethyldec-7-en-4-one) (4), a natural acyclic monoterpene ketone, which is used in the present invention can be easily

Tagetes minuta (family: Asteraceae) is most suitable essential oil bearing plant and therefore, cultivated in several countries such as Egypt, France, Morocco, Brazil, United States and India for its applications in perfumery, flavour and pharmaceutical industries (Chalchat, J. C., Garry, R. P. and Muhayimana, A., J. Essential Oil Research, & (4), 375–386 (1995). Out of one hundred and four components, the major constituents of the essential oil of T. minuta are tagetone, E/Z-ocimene, E/Z-ocimene, germacrene, limonene, trans-anethole and desired dihydrotagetone (Hethelyi, E., Danos, B. Tetenyi, P. and Koczka, I., Flavour and Fragrance J., 1(4–5), 169–173 (1986); Wilson, R. A., zampino, M. J., Mookherjee, B. D., owerkerk, A. and Monteleone, M. G., U.S. Pat. No. 4,960,754 (1990). Tagetes oil has been known from long due to its pleasant and powerful odors and flavours, however, tagetes oil does not have very attractive price in market and also has storage problem as it constitutes undergo polymerization such as the highly conjugated tagetone, ocimene and ocimene etc. Polymerization deteriorates the quality of the tagetes oil as well as makes it unsuitable for any kind of applications, therefore, an economic temptation arose to utilize this acyclic monoterpene ketone (dihydrotagetone) (4) for its conversion into high valued products which appeared to us γ-butyractone (1). Among the Tagetes spp., the chemical composition of T. minuta oil is well documented with the seasonal variation and the percentage of dihydrotagetone is detected upto 60% or more (Singh, B., Sood, R. P., Singh, V., J. Essential Oil Research, 4(5), 525–526 (1992); Thappa, R. K., Agarwal, S. G., Kalia, N., K., Ramesh, K. and Kapoor, R., J. Essential Oil Research, 5(4), 375–379 (1993); Singh, B. and Sood, R. P., Indian Perfumer, 39, 102–106 (1995); Chisowa, E. H., Hall, D. R. and Farman, D. I., J. Essential Oil Research, 10(2), 183–184 (1998); Dargie, S. N. and Mehta, V. D., Phytochemistry, 48(2), 395–396 (1998); Raol, V. S. P., Spandl, M. A., Folin, M. A., Riedel, V. and Prakash, E. V. S., J. Essential Oil Research, 11(2), 259–261 (1999).

As dihydrotagetone (4) is a starting material, therefore it becomes very important to develop a simple and economical process for the separation of dihydrotagetone from the mixtures of closely related several constituents (tagetone, ocimene etc) of Tagetes minuta oil which is done by two step process Le Soxhlet extraction of tagetes oil adsorbed on solid support followed by column chromatography. Soxhlet extraction of solid (taken in thimble) occupies an important place amongst the technologies for separation of the desired constituents from its various constituents. However the high impurity of tagetes oil on solid support such as silica gel, alumina or celite and Soxhlet extraction with non polar solvents such as petroleum ether and hexane was found extremely successful to separate dihydrotagetone from remaining constituents since more polar constituents and polymerized materials bound on solid supports does not react with non polar solvents. It is worthwhile to mention that initial cold percolation of alumina bound tagetes oil using hexane allows removal of non polar constituents mainly ocimene, however, warm percolation (with hexane) of Soxhlet extractor brings down yellow coloured dhydrotagetone rich fractions. The fractions were monitored on TLC plate and the desired fractions were combined and eluted with hexane with an increasing amount of ethyl acetate (upto 5%) afforded dihydrotagetone as a sweet smelling liquid in above 99% purity by gas chromatography (tr=16.25 min) performed on a Shimadzu-GC-14B/C-R7A gas chromatograph with the following conditions: carboxax column; 0.25 mm/60 m; injector 250°C; FID detector 250°C; temp. programme 40 (hold for 2 min.) to 190°C (hold for 20 min.) 5°C min⁻¹. In this two step purification, Soxhlet extraction is found very efficient to take out dihydrotagetone even from polymerized tagetes oil since dihydrotagetone does not polymerize and its percentage found to be higher in old polymerized oil. Therefore, above developed Soxhlet extraction appears as an effective process otherwise purification of tagetes oil directly on column chromatography is tedious and time consuming and does not allow to obtain dihydrotagetone in high purity. On the basis of IR and NMR spectrum of liquid (4) and its comparison with those reported in the literature, the liquid was confirmed to be dihydrotagetone (4). This by-functional alkene (4) appears as an attractive intermediate in which without protecting ketone function, it is possible to change the terminal double bond into corresponding acid with Lemieux-von Rudloff reagent (Lemieux, R. U., von Rudloff, E., Can. J. Chem., 33, 1701 (1955)).


The olfactory assessment of this interesting oxo acid (3) is performed as per Indian standard of 2284, 1997 (Bureau of Indian Standard, Manak Bhavan, 9 Bahadur Shah Zafar Marg, New Delhi). The
The odour effect of oxo acid (3) perceived by the olfactory nerves on smelling a strip impregnated with an oil is giving the impression of sweetness with the acidity thereby enabling its use as multipurpose additives in the areas of soft drinks, desserts and tobacco products etc since several related 4-oxoacid derivatives such as 4-oxovaleric acid and volatile acidic fraction of sun-cured Nicotiana tabacum (Greek tobacco) (Kimland, B., Aasen, A. J., Almqvist, S. O., Arestro, P. and Enzell, C. R., Phytochemistry 12:835–847 (1973)) are well known for their wide applications in flavour industries especially as tobacco additives to improve the tobacco flavour. In addition, 2,6-dimethyl-4-oxo-heptanoic acid (3) can also be converted into corresponding 4-oxoesters and 4-hydroxyacids which are found in plant kingdom and are widely used in flavour industries (Taylor, A. J. and Mottram, D. S., in: Flavour Science, Recent Developments, The Royal Society of Chemistry, Cambridge CB4 4WF, U.K., 151–162 (1996)). However, keeping in view all the information collected from literature, 2,6-dimethyl-4-oxo-heptanoic acid (3) appears itself an interesting product having a wide scope for vigorous biological activities as well as new additive for various applications particularly in flavour industries. With the world market expansion towards natural flavours, there is clearly a huge potential for the application of new and simple techniques for production in which microwave assisted (Bose, A. K., Bank, I. K., Lavinskata, N., Jayaraman, M. and Manhas, M. S. Chemtech, 27, 18, (1997)) oxidation of dihydrogaretone (conducted for the first time) appeared as a rapid, economical and high yielding method for production of 4-oxo acid (3) (81% yield) higher than the above conventional method (71% yield). It is worthwhile to mention that the addition of alumina during microwave oxidation of dihydrogaretone provide not only a solid support (McKillop, A., Young, D. W., Synthesis, 481–500 (1979) and Clark, J. H., Gullen, S. R., Barlow, S. J. and Bastock, T. W. J. Chem. Soc. Perkin Trans 2, 1117, (1994)) but it also accelerates the oxidation reaction (Example II).

Reduction of this 4-keto acid (3) with metal hydride such as sodium borohydride, lithium aluminium hydride furnished hydroxy acid (2) (without isolation) which when treated directly with strong acid such as hydrochloric acid, sulphuric acid, paratoluene sulphonie acid, acetic anhydride, trifluoro acetic acid or the like gave coconut flavoured γ-butyrolactone (1) as diastereoisemic mixture (68:32) on the basis of NMR (Example III and FIG. 4) and GC (FIG. 5).

This semisynthetic butyrolactone (1) is as good as the natural whisky lactone (1a) and has added advantage that it can be made available in any quantity required utilizing cheaper starting material and inexpensive reagents and also in less time with overall good yield. In addition, this coconut flavoured γ-butyrolactone (1) appears as a multipurpose additive for flavouring a variety of food products of coconut flavour. Scanning of literature reveals that there are some lactones such as 3-methyl-n-γ-lactone (F.E.M.A. 3103) and γ-nonyllactone (coconut aldehyde) (F.E.M.A. 2781) or the like which have aroma like coconut and the Flavour and Extract Manufactures Association (F.E.M.A.) monograph cites their uses in a wide range of food stuffs including baked goods and confectionery (Steffen, A., in: Perfume and Flavor Chemicals, Part I & II, Allured Publishing Corporation, Carol Stream, Ill. 60188-2787, (1994)). The above coconut aldehyde (also known as γ-nonyllactone or 4-hydroxynonanoic acid, lactone or prunolide or γ-nonalactone or γ-n-amy butyrolactone or abricolin or aldehyde C-18) (F.E.M.A. 2781) is one of the most frequently used lactones in perfumes and flavours and its field of application reaches from the finest luxury perfumes to inexpensive masking colours, from gardenia flowers bases to coconut candy flavours. Its creamy, coconuty, delicately fruity (in extreme dilution) and floral-musk odour of excellent tenacity is often utilized along with that of undecanole (so-called aldehyde C-14) in garden, tuberose, honeysuckle, stephanotis, plumeria, jasmine and many other heavy floral types. In flavours, the coconut aldehyde finds even more extensive use, and often in much higher concentration. Besides coconut, this material is used in imitation of bitter-almond, in fruit blends (fixative), berry complexes, nut variations, American cherry flavour etc. in which concentration may vary from 10 to 50 ppm in the finished product (G.R.A.S. F.E.M.A. No. 2781). This important coconut aldehyde can be prepared by either from methylaureylate plus hexanol with di-tertiary-butylperoxide or from undecylenic acid with malonic acid condensation or from nonoic acid (obtained from castor oil) by lactonization. The importance of coconut flavour can also be understood as a combination of diluted coconut water concentrate with equal amount of milk, water and sugar give a delicious drink. Recently, coconut water concentrate is developed by Miracle Food Processors International Ltd. (MFPI) and the quality was certified by Central Food Technology Research Institute (CFTRI), Mysore (India) and at a research center KIN in Germany. Considering all the above factors, it appeared to us that our new γ-butyrolactone (1) with coconut flavour is not only an analogue of whisky lactone (1a) but can also be used as a substitute to coconut aldehyde or aldehyde C-18 (1b) which occurs in peaches, apricots and coconut (Bennett, H., in: Concise Chemical and Technical Dictionary, Fourth Edition, ed., Edward, A., Publishing Ltd., 41 Bedford Square London, WC1B, 3DQ, 48, (1986) and Orlando, S. in: Handbook of Perfumes and Flavours, Chemical Publishing Co., Inc., New York, N.Y. 152 (1990)).

![Chemical Structure](image)

**EXAMPLES**

The following examples are given by way of illustration of the present invention and should not be considered to limit the scope of the present invention.

**Example I**

(a) Isolation of dihydrogaretone(4) from Tagetes minuta oil: The large scale purification of tagetes oil is achieved by adsorbing it on alumina (6–10 times more than the oil, w/w) for leading on a soxhlet extractor (30 cm long and 6.5 cm diameter or more). The cold percolation of extractor with
hexane removes non polar impurities such as ocinene. However, warm hexane percolation of Soxhlet extractor with an increasing proportion of ethylacetate unto 0.1 to 2% gave mainly dihydrogaretone rich fractions. The fractions were monitored on TLC plate and the desired fractions were combined and solvent was removed under vacuum to afford sweet smelling dihydrogaretone richer fractions (38 to 43% w/w or more depending upon the % of dihydrogaretone in tagetes oil) which were finally purified on silica gel column and the column was eluted with an increasing amount of hexane/ethyl acetate (100:0 to 95:5). The fractions having same 

Example II

###STR2##

**Synthesis of 2,6-dimethyl-4-oxo-heptanoic acid (3):**

(i) (by conventional method): To a vigorously stirred solution of dihydrogaretone (4) (5 g, 33 mmole) in 1,4-dioxane-water (250 mL, 4:1) is added potassium carbonate (7 g) followed by powdered potassium metaperiodate (15 g, 70 mmol) and then 0.25 g of powdered potassium permanganate. To the above mixture 5 to 6°C C and the mixture was then cooled to effect complete precipitation. The precipitate was collected, washed with water, and then dried under reduced pressure. The crude product was purified by column chromatography using silica gel and eluting with ether/petroleum ether (9:1). The product was obtained as a colorless oil (2.0 g, 56%).

(ii) (by microwave method): A mixture of dihydrogaretone (4) (3.53 g, 22.92 mmole), sodium bicarbonate (1.8 g, 21.43 mmole), sodium metaperiodate (22.34 g, 106.78 mmole), alumina (0.8–1.2 g) and THF-water or acetone-water (20 mL, 1:3) was taken in a 250 mL Erlenmeyer flask fitted with a loose funnel at the top. The flask was shaken well for a while and then placed inside a microwave oven (Kenstar) operating at medium power (600 W) and irradiated for 2–20 minutes in parts. After completion of the reaction (monitored by TLC), the contents of the flask were poured into saturated solution of sodium bisulphite to destroy excess metaperiodate and resulting mixture was passed through a bed of Celite and further washed with aqueous acetone (40 mL×3, 3:1). The filtrate and washings were combined and the solution concentrated under reduced pressure to remove acetone. The mixture was cooled to 0 to 8°C C and acidified with dilute acid and then extracted with ethylacetate (3x25 mL). The ethylacetate layers were then combined, washed with saturated sodium bisulphite and sodium chloride (5x15 mL), dried over anhydrous sodium sulphate and then filtered. The solvent was removed to afford a crude product which was purified on silica column as above which afforded 2,6-dimethyl-4-oxo-heptanoic acid (3) in 81% yield as a viscous liquid. The physical and spectral data of this product is found similar as above (Example II-i).

**Example III**

**Synthesis of 5-isobutyl-3-methyl-4,5-dihydro-2(3H)-furanone (4-isobutyl-2-methyl-4-butanolide) (1):**

A cold solution (~5 to 8° C) of sodium borohydride (0.6 g) in 2N-sodium hydroxide (1.5 mL) was added dropwise very slowly to a stirred solution of 2,6-dimethyl-4-oxo-heptanoic acid (3) (1.0 g) in methanol (20 mL) and stirring was continued overnight. Methanol was evaporated and more water (3–4 mL) was added to the residue and the mixture was once again cooled to ~5 to 5°C C and then acidified with cold dilute hydrochloric acid and then extracted with ether (3x50 mL). The combined extracts were washed with brine, dried (Na₂SO₄) and then concentrated to give a crude liquid which was loaded on silica gel column and the column was eluted with an increasing amount of hexane/ethyl acetate (100:0 to 95:5). The fractions were monitored on TLC plate and then the desired fractions were combined and solvent was removed under vacuum to afford 4-isobutyl-2-methyl-4-butanolide (trans-cis:68:32 on NMR basis) of formula (1) in 76% yield as a colourless oil with coconut flavour; Rₖ=0.80 (10% ethylacetate in hexane); IR (film) 3292, 1772, 1558, 1457, 1362, 1173, 1135, 1004, 1024, 926 cm⁻¹; ¹H NMR δ 4.63–4.55 (ddq, 0.32H, cis-5, 1H, 3, CH₃), 4.46–4.38 (dd, 0.68H, trans-C-5), 2.73–2.62 (m, 1H, 3, CH₃), 2.55–2.47 (m, 1H, 7, CH₂), 2.00–2.04 and 1.84–1.78 (2 m, 2H, 4, CH₂), 1.67–1.61 and 1.50–1.41 (2 m, 2H, 6, CH₂), 1.36–1.26 and 1.27–1.25 (2 m, 3H, 10, CH₃), 0.96–0.95 and 0.94–0.93 (2 m, 6H, 8, CH₂, and 9, CH₃) (FIG. 3); ¹³C NMR δ 179.42 (d, C-2), 76.55 (d, C-5), 44.43 (d, C-3), 37.69 (s, C-4), 35.69 (d, C-7), 33.77 (s, C-6), 21.92 (d, C-10), 15.65, 14.89 (2s, C-8 and C-9) (FIG. 4); GC of compound 1 appeared at tᵣ=30.24 and tᵣ=30.44 also confirms the presence of isomers (FIG. 5); EIMS m/z 156 [M⁺] (13.3, 99(100%), 84(5.4), 71(28.5), 56(14.7), 42(12.8) (FIG. 6). The main advantages of the present invention are:

1. A simple process for industrial scale purification of dihydrogaretone of formula (4) from oil of Tagetes species.
2. A simple process for proper utilization of dihydrogaretone of formula (4) into high valued products.
3. A simple process for the preparation of 2,6-dimethyl-4-oxo-heptanoic acid of formula (3) as a novel product with one chiral center. i.e. 2,6-dimethyl-4-oxo-heptanoic acid (3).

4. A simple process is obtained for rapid conversion of dihydrotagetone of formula of (4) into 2,6-dimethyl-4-oxo-heptanoic acid of of formula (3) in microwave oven ranging from 5–30 minutes where the rate of reaction is further accelerated by adding alumina as a catalyst.

5. A process for the preparation of 2,6-dimethyl-4-oxo-heptanoic acid of formula (3) in high yield as a multipurpose additive in food and tobacco products as its analogue 2,6-dimethyl-5-oxo-heptanoic acid (found as a constituent of an important Italo-Mitcham peppermint oil i.e. Mentha piperita L) has wide application in perfumery, flavour and pharmaceutical industry.

6. A simple process for the preparation for 5-isobutyl-3-methyl-4,5-dihydro-2(3H)-furanone of formula (1) in high yield and high purity.

7. A simple process for the preparation of 5-isobutyl-3-methyl-4,5-dihydro-2(3H)-furanone of formula (1) as an analogue of whisky lactone or oak lactone or quercus lactone of formula (1a).

8. A simple process for the preparation of 5-isobutyl-3-methyl-4,5-dihydro-2(3H)-furanone of formula (1)) as an analogue of coconut aldehyde (1b).

9. The use of 5-isobutyl-3-methyl-4,5-dihydro-2(3H)-furanone as a starting material which is capable of undergoing conversion into new kind of corresponding furans, cyclopentenones, butenolides and pyrrolidone or the like.

10. Providing a novel product i.e. 5-isobutyl-3-methyl-4,5-dihydro-2(3H)-furanone of formula (1) in sufficient quantity for its applications in perfumery, flavour and pharmaceutical industries.

What is claimed is:

1. The compound 2,6-dimethyl-4-oxo-heptanoic acid.

2. Soft drink formulations comprising the compound 2,6-dimethyl-4-oxo-heptanoic acid in a desired and acceptable amount varying from ppm to mg along with other drink compositions.

3. Herbal formulations comprising the compound 2,6-dimethyl-4-oxo-heptanoic acid in a desired and acceptable amount varying from ppm to mg along with other herbal compositions.

4. Alcoholic beverage formulations comprising the compound 2,6-dimethyl-4-oxo-heptanoic acid in a desired and acceptable amount varying from ppm to mg along with other beverage compositions.

5. Confectionery formulations comprising the compound 2,6-dimethyl-4-oxo-heptanoic acid in a desired and acceptable amount varying from ppm to mg along with other confectionery compositions.

6. Tobacco product formulations comprising the compound 2,6-dimethyl-4-oxo-heptanoic acid in a desired and acceptable amount varying from ppm to mg along with other tobacco compositions.

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