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TOBACCO PRODUCT

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This invention relates to a tobacco product and has for an object the provision of a composition and process for improving the flavor and aroma of tobacco and tobacco smoke.

It is well known in the tobacco art that the flavor and aroma of the tobacco product and the smoke from the tobacco are very important considerations in so far as the ultimate consumer is concerned. Considerable efforts are exerted by the manufacturers of tobacco products to provide a product that will be acceptable to the consumer, particularly as regards flavor and aroma characteristics. It has been the common practice in the tobacco industry to prepare blends of domestic and oriental tobaccos in order to provide smoking tobacco which has a pleasing flavor and aroma before and during smoking. However, such a procedure is costly and may at times become prohibitive in the event that certain types of tobacco may be in short supply. Accordingly it is a further object of this invention to provide a new class of additive materials which when applied to the tobacco products improve and enhance the flavor and aroma of these products and the smoke emitted therefrom thereby increasing or enhancing the pleasure and other values that may be derived by the consumer from the use of these products.

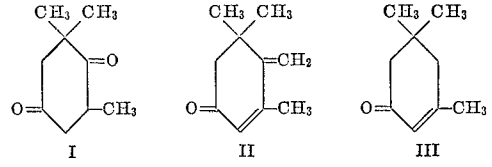
A further object of this invention is the provision of a process for enhancing or otherwise improving the flavor, aroma and other qualities of certain domestic, oriental, reconstituted or synthetic tobaccos which may be deficient in said flavor or aroma or other qualities.

An additional object of this invention is to provide a process of preparing a smoking tobacco or product which when smoked has an enhanced flavor or aroma.

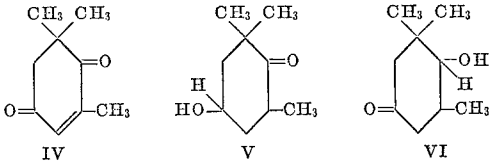
A still further object of this invention is the provision of smoking products, such as cigarettes, cigars or pipe tobacco, and a process for forming same whereby the flavor and aroma before and during smoking are improved or enhanced.

Further and additional objects will appear from the following description and the appended claims.

In accordance with one embodiment of this invention, a tobacco product is provided to which has been added or which has been treated with a small amount of a compound selected from the group of cyclohexenones and cyclohexanones having the formulae:



I II III



IV V VI

The following specific examples illustrate the preparation and characteristics of representative tobacco additives of the present invention. It will be appreciated, however, that this invention is not limited to the specific process by which the compounds may be prepared. All temperatures are expressed in degrees centigrade.

Example I.—Synthesis of 3,5,5-trimethyl-2-cyclohexene-1,4-dione (Formula IV)

2,6,6 - trimethyl-2-cyclohexene-1-ol-4-one was oxidized with chromium trioxide according to the method of Isler et al., United States Patent No. 2,917,539.

The above keto alcohol, 10 grams, obtained by fermentation of isophorone was mixed with 13 milliliters of acetic acid and 35 milliliters of water. To this mixture was added dropwise 4.7 grams of chromium trioxide in 13 milliliters of water with stirring and cooling so that the temperature did not rise above 30°. The mixture was stirred overnight at 15–30°. The reaction mixture was saturated with ammonium chloride and extracted twice with 60 milliliters of 1:1 pentane-hexane solution. The extracts were dried over sodium sulfate and concentrated. Distillation in vacuo of the residue gave 5 grams of 3,5,5-trimethyl-2-cyclohexene-1,4-dione, boiling point 88–90°/8 mm.

Example II

The compound 2,6,6 - trimethyl - 1,4-cyclohexanedione (Formula I) is prepared by the reduction of the product of Example I using zinc and glacial acetic acid. It is a solid melting at 63–64° and has the following characterization:

Nuclear magnetic resonance spectrum: τ=8.88(3), 8.884(3, doublet), 8.80(3), 7.40(2), and 7.13(1).
Infrared spectrum: 1710, 1237, 1208, 932, 907, 885, 775, and 748 cm.$^{-1}$.
Mass spectrum: parent peak at 154 mass units.

Example III.—Synthesis of 4-methylene-3,5,5-trimethyl-2-cyclohexen-1-one (Formula II)

(*a*) *Preparation of safranol.*—A mixture of 8.2 grams of safranol containing a small amount of cyclocitral and 0.7 gram of sodium borohydride in 25 milliliters of water was allowed to stand two hours. Dilute hydrochloric acid was then added, and the mixture was extracted with ether; the ether solution was dried, and the solvent was removed in vacuo. A mixture of alcohols (7.2 grams) was recovered.

(*b*) *Rearrangement of safranol to 4-methyleneisophorol.*—The 7.2 grams of safranol containing some cyclogeranoil was dissolved in 175 milliliters of cold acetone, and 70 milliliters of 0.5 N sulfuric acid was added. The solution was kept at 0° for three days. The solution was then added to 250 milliliters of saturated sodium bicarbonate solution; this mixture was extracted with ether, and the ether solution was washed with bicarbonate solution and dried. The residue after evaporation of the ether was chromatographed on silicic acid; three main fractions were obtained with weights of 1.36, 1.37 and 0.83 grams. The first fraction was cyclogeraniol, the second 4-methyleneisophorol, and the third was not identified.

(*c*) *Oxidation of 4-methyleneisophorol.*—4 - methyleneisophorol (1.37 grams) was oxidized with 1 gram of chromium trioxide in 35 milliliters of pyridine at room temperature. After four hours the reaction mixture was hydrolyzed with water and the product was extracted into ether. After washing the ether solution with 2 N hydrochloric acid the ether solution was dried and the ether removed by distillation. The residue (1.03 grams) was 4-methylene - 3,5,5-trimethyl - 2-cyclohexen - 1-one as shown by the infrared and nuclear magnetic resonance spectra.

Infrared spectrum: 1665, 1590, 1280, 1250 and 905 cm.$^{-1}$
Nuclear magnetic resonance spectrum: τ=4.00 (1, HC=), 4.52 (2, H_2C=), 7,62 (2, CH_2C=O), 7.90 (3, CH_3C=), and 8.77 (6, geminal methyls).

Example IV

The compound of Formula III is 3,5,5-trimethyl-2-cyclohexenone, a commercially available chemical known as isophorone. It has the following characterization:

Infrared spectrum: 1660, 1300, 1280, 1250, 1150, 1115, 1020, 900, 855 cm.$^{-1}$

Nuclear magnetic resonance spectrum: τ=4.00 (1, CH_3—), 8.07 (3, CH_3C=), 7.83 (4, CH_2C=), and 4.20 (1, HC=).

Mass spectrum: parent peak at 138 mass units.

Boiling point: 211–214°.

Example V.—Synthesis of 4-hydroxy-2,2,6-trimethylcyclohexanone (Formula V) and 4-hydroxy-3,3,5-trimethylcyclohexanone (Formula VI)

One gram of 4 - ketodihydroisophorone was reduced with 65 milligrams of sodium borohydride in 20 milliliters of 50% aqueous methanol. After standing three hours, the mixture was acidified and extracted with ether. The ether extract was dried, concentrated, and chromatographed on silicic acid. Three fractions contained ketoalcohols, and four fractions contained two isomeric diols. Separation of the ketoalcohol fractions by gas chromatography gave 70% of 4-hydroxy - 3,3,5-trimethylcyclohexanone and 30% of 4-hydroxy - 2,2,6-trimethylcyclohexanone. The 4-hydroxy - 3,3,5-trimethylcyclohexanone has the following properties:

Infrared spectrum: 3430, 1708, 1280, 1076, 1055, 1032, 988, and 857 cm.$^{-1}$

Nuclear magnetic resonance spectrum: τ=6.73 (1, doublet), 7.46 (2), 7.82 (2), 8.93 (6), and 9.16 (3, doublet).

The compound 4 - hydroxy - 2,2,6 - trimethylcyclohexanone has the following properties:

Infrared spectrum: 3430, 1710, 1128, 1095, 1060, 1030 and 994 cm.$^{-1}$

Nuclear magnetic resonance spectrum: τ=5.92 (1), 8.64 (OH), and 8.86–9.07 (9).

Analysis.—Calcd. for $C_9H_{16}O_2$: Mol. wt., 156. Found: Mass, 156.

It has been found that the tobacco additives of the invention when incorporated into tobacco products impart a flavor and aroma both before and during smoking which many smokers consider to be desirable in smoking products. However, it is pointed out that the methods for defining or characterizing the quality of a flavor or aroma in the tobacco art are almost purely subjective and different smokers may define the same flavor quite differently. Also, the compounds included within the broad scope of this invention may impart different flavors or aromas depending upon the substituents therein. Thus, the compounds comprehended by this invention, by subjective tests, impart characteristic flavors which are desirable in tobacco products and the smoke therefrom even though the exact character thereof cannot be described on the basis of known standards. The tobacco additive of Example I imparts an aroma or flavor which is reminiscent of pine which many smokers consider to be desirable in a tobacco product. The tobacco additive of Example II imparts an aroma and flaxor which some characterize as camphoraceous; the tobacco additive of Example III has a sweet note; the tobacco additive of Example IV is methol-like with a camphor note; and the compounds of Example V both have very delicate burnt sugar notes.

In accordance with this invention, an above indicated compound or a mixture of compounds is added to tobacco or applied to a smoking article or its component parts in amounts of about 0.0005 to 2.0 percent by weight of the product. Preferably the amount of additive is between about 0.01 and 0.5 percent by weight in order to provide a tobacco product having a desired flavor and aroma.

However, the amount used will depend upon the amount of flavor and aroma desired and the particular compound or mixture thereof that is used. The additive may be incorporated at any step in the treatment of the tobacco but is preferably added after aging, curing and shredding and before the tobacco is formed into cigarettes. Likewise, it will be apparent that only a portion of the tobacco need be treated and the thus treated tobacco may be blended with other tobaccos before the cigarettes or other smoking articles are formed. In such case the tobacco treated may have the additive in excess of the amounts above indicated so that when blended with other tobaccos the final product will have the percentage within the indicated range.

In accordance with one specific embodiment of this invention, an aged, flue-cured and shredded tobacco is sprayed with a 1% ethyl alcohol solution of 3,5,5-trimethyl-2-cyclohexene-1,4-dione in an amount to provide a tobacco containing 0.25 percent by weight of the additive on a dry basis. Thereafter the alcohol is removed by evaporation and the tobacco is manufactured into cigarettes by the usual techniques. It has been found that the cigarette when prepared as indicated has a desired and pleasing flavor, an aroma which to some people is reminiscent of pine and is detectable and pleasing in the main and side smoke streams when the cigarette is smoked.

The additives falling within the scope of this invention may be applied to the tobacco by spraying, dipping or otherwise, utilizing suitable suspensions or solutions of the additive. Thus water or volatile organic solvents, such as alcohol, ether, acetone, volatile hydrocarbons and the like, may be used as the carrying medium for the additive while it is being applied to the tobacco. Also, other flavor- and aroma-producing additives, such as those disclosed in United States Patents Nos. 2,766,145, 2,905,575, 2,905,576, 2,978,365 and 3,041,211 may be incorporated into the tobacco with the additive of this invention.

While this invention is principally useful in the manufacture of cigarette tobacco, it is also suitable for use in connection with the manufacture of pipe tobacco, cigars or other tobacco products. Furthermore, the compounds may be added to certain tobacco substitutes of natural or synthetic origin and by the term "tobacco" as used throughout this specification is meant any composition intended for human consumption by smoking or otherwise, whether composed of tobacco plant parts or substitute materials or both.

Also, the invention has been particularly described with reference to the addition of the compounds directly to tobacco. However, it will be apparent that the compound may be applied to the paper of the cigarette or to the wrapper of a cigar. Also it may be incorporated into the filter tip, the packing material or the seam paste employed for gluing the cigarette paper. Thus a tobacco product is provided which includes the specified additives and tobacco although in every instance the compound need not be admixed with the tobacco as above specifically described.

While several particular embodiments of this invention are shown above, it will be understood, of course, that the invention is not to be limited thereto, since many modifications may be made, and it is contemplated, therefore, by the appended claims, to cover any such modifications as fall within the true spirit and scope of this invention.

We claim:

1. A tobacco product having added thereto an amount sufficient to alter the flavor or aroma of the tobacco product of a compound selected from the group consisting of 3,5,5-trimethyl-2-cyclohexene-1,4-dione, 2,6,6-trimethyl-1,4-cyclohexanedione, 4-methylene-3,5,5-trimethyl-2-cyclohexen-1-one, 3,5,5 - trimethyl-2-cyclohexenone, 4-hydroxy-2,2,6-trimethylcyclohexanone and 4-hydroxy-3,3,5-trimethylcyclohexanone.

2. The product recited in claim 1 wherein the amount of said compound is between about 0.0005 and 2.0 percent by weight of the product.

3. The product recited in claim 1 wherein the amount of said compound is between about 0.01 and 0.5 percent by weight of the product.

4. A tobacco product having added thereto and dispersed therein a small amount sufficient to improve the flavor thereof of 3,5,5-trimethyl-2-cyclohexene-1,4-dione.

5. A tobacco product having added thereto and dispersed therein a small amount sufficient to improve the flavor thereof of 2,6,6-trimethyl-1,4-cyclohexanedione.

6. A tobacco product having added thereto and dispersed therein a small amount sufficient to improve the flavor thereof of 4-methylene-3,5,5-trimethyl-2-cyclohexen-1-one.

7. A tobacco product having added thereto and dispersed therein a small amount sufficient to improve the flavor thereof of 3,5,5-trimethyl-2-cyclohexenone.

8. A tobacco product having added thereto and dispersed therein a small amount sufficient to improve the flavor thereof of 4-hydroxy-2,2,6-trimethylcyclohexanone.

9. A tobacco product having added thereto and dispersed therein a small amount sufficient to improve the flavor thereof of 4-hydroxy-3,3,5-trimethylcyclohexanone.

10. The product recited in claim 4 wherein the amount of flavorant is between about 0.0005 and 2.0 percent by weight of said product.

11. The product recited in claim 5 wherein the amount of flavorant is between about 0.0005 and 2.0 percent by weight of said product.

12. The product recited in claim 6 wherein the amount of flavorant is between about 0.0005 and 2.0 percent by weight of said product.

13. The product recited in claim 7 wherein the amount of flavorant is between about 0.0005 and 2.0 percent by weight of said product.

14. The product recited in claim 8 wherein the amount of flavorant is between about 0.0005 and 2.0 percent by weight of said product.

15. The product recited in claim 9 wherein the amount of flavorant is between about 0.0005 and 2.0 percent by weight of said product.

References Cited

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