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3,372,699
TOBACCO

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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 insofar 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.

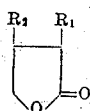
An additional object of this invention is to provide a process of preparing a smoking tobacco or product which when smoked has an enhanced flavor or aroma.

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 compounds having the formula:



Formula I

wherein R_1 and R_2 are selected from the group consisting of hydrogen and alkyl radicals with at least one of said substituents being an alkyl group. Preferably the alkyl groups are lower alkyl radicals of from 1 to 4 carbon atoms. The compounds falling within the scope

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of the generic Formula I are gamma-lactones having an alkyl substituent in either the alpha or beta position. Thus, preferably R_1 and R_2 can be methyl, ethyl, propyl, isopropyl, butyl and isobutyl.

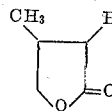
The following specific examples illustrate the preparation and characteristics of representative tobacco additives of the present invention. It will be appreciated, however, that this invention is not limited to the specific processes by which the compounds may be prepared. All temperatures are expressed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

Example I.—The synthesis of β -methylbutyrolactone

(a) *Hydrobromination of methallyl chloride.*—A solution of 10 grams of methallyl chloride in 50 milliliters of hexane was saturated with dry hydrogen bromide at 0–10° in the presence of 1% benzoyl peroxide. The solution was washed several times with water and the organic layer was dried and concentrated, yielding 18.1 grams of 1-bromo-3-chloro-2-methylpropane.

(b) *Preparation of 5-chloro-3-methylbutanoic acid.*—A mixture of 18.1 grams of 1-bromo-3-chloro-2-methylpropane, 6.9 grams of potassium cyanide, 60 milliliters of water and 3 milliliters of ethanol was refluxed four hours. The resulting solution was acidified with 50 milliliters of concentrated hydrochloric acid and refluxed an additional five hours. The aqueous layer was extracted with ether and the ether layers were dried and concentrated, yielding 13.8 grams of crude acid.

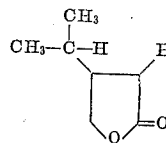
(c) *Lactonization of 5-chloro-3-methylbutanoic acid.*—A mixture of 13.8 grams of crude 5-chloro-3-methylbutanoic acid and 300 milliliters of 50% sodium carbonate solution was refluxed 14 hours and the resulting solution was acidified with 5 N hydrochloric acid and extracted with ether. The residue from the reaction was analyzed by vapor phase chromatography using a 10' x 1/4" column packed with 10% diethylene glycol adipate on 80–100 mesh carrier. The main peak was shown by infrared and nuclear magnetic resonance spectra to have the formula:



Example II.— β -Isopropylbutyrolactone

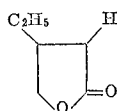
Forty-two grams of 2-isopropyl-1,3-propanediol, boiling point 118–120°/10 mm. was saturated with anhydrous hydrogen bromide at 0 to –5° (about 4 hours) and the reaction mixture was distilled to give 26.8 grams of 1-bromo-2-isopropylpropanol, boiling point 102–106°/7 mm.

To a solution of 12 grams of potassium cyanide in 75 milliliters of water were added 32.8 grams of 1-bromo-2-isopropylpropanol in 15 milliliters of ethanol. The solution was refluxed for 15 hours, cooled, diluted with 20 milliliters of water and acidified with 50% hydrochloric acid. Extraction with ether and distillation of the residue produced 12.5 grams of β -isopropylbutyrolactone, boiling point 73–76°/1 millimeter having the formula:



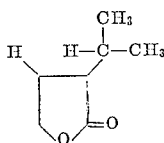
Example III.— β -Ethylbutyrolactone

The title compound, boiling point $59^{\circ}/1$ mm., is prepared according to the procedure of Example II using 2-ethyl-1,3-propanediol in lieu of the 2-isopropyl-1,3-propanediol. The intermediate 1-bromo-2-ethylpropanol had a boiling point of $84-86^{\circ}/8$ mm. The final tobacco additive has the formula:



Example IV.— α -Isopropylbutyrolactone

Ethylhydrogenisopropylsuccinate (26.1 grams, 0.14 mole) was added dropwise to a solution of 5.5 grams of lithium aluminum hydride in 400 milliliters of absolute ether. After the addition was completed, the solution was refluxed for two hours, cooled, acidified with dilute hydrochloric acid, and extracted with ether. Concentration of the ether layer and distillation of the residue gave 7.1 grams of α -isopropylbutyrolactone, boiling point $96-100^{\circ}/8$ millimeters, having the formula:



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 alkyl 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 provides a smooth smoke. The tobacco additive of Example II imparts an aroma and flavor which some characterize as a sweet, rich tobacco taste with the tobacco additive of Example III being similar; and that of Example IV imparts a sharp woody note.

In accordance with this invention, a compound embraced by generic formula I or mixtures thereof is added to tobacco or applied to a smoking article or its component parts in amounts of about 0.001 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 β -isopropyl-

butyrolactone 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 sweet rich tobacco similar to that found in a tobacco-curing shed, 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 U.S. 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.

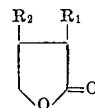
While this invention is principally useful in the manufacture of cigarette tobacco, it is also suitable for use in connection with the manufacture of pipe tobacco, cigars or other tobacco products. Furthermore, the compounds may be added to certain tobacco substitutes of natural or synthetic origin and 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 packaging 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.

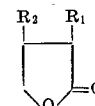
I claim:

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 of compounds having the formula



wherein R_1 and R_2 are selected from the group consisting of hydrogen and alkyl groups with at least one of such substituents being alkyl.

2. 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 of compounds having the formula



wherein R_1 and R_2 are selected from the group consisting of hydrogen and alkyl groups of 1 to 4 carbon atoms with at least one of such substituents being alkyl.

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3. The product recited in claim 1 wherein the amount of said compound is between about 0.001 and 2.0 percent by weight of the product.

4. 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.

5. A tobacco product having added thereto and dispersed therein a small amount sufficient to improve the flavor thereof of β -methylbutyrolactone.

6. A tobacco product having added thereto and dispersed therein a small amount sufficient to improve the flavor thereof of β -isopropylbutyrolactone.

7. A tobacco product having added thereto and dispersed therein a small amount sufficient to improve the flavor thereof of β -ethylbutyrolactone.

8. A tobacco product having added thereto and dispersed therein a small amount sufficient to improve the flavor thereof of α -isopropylbutyrolactone.

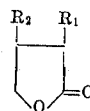
9. The product recited in claim 5 wherein the amount of flavorant is between about 0.001 and 2.0 percent by weight of said product.

10. The product recited in claim 6 wherein the amount of flavorant is between about 0.001 and 2.0 percent by weight of said product.

11. The product recited in claim 7 wherein the amount of flavorant is between about 0.001 and 2.0 percent by weight of said product.

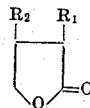
12. The product recited in claim 8 wherein the amount of flavorant is between about 0.001 and 2.0 percent by weight of said product.

13. A process for improving the flavor of a tobacco product which comprises adding thereto a small amount of a compound selected from the group of compounds having the formula



wherein R_1 and R_2 are selected from the group consisting of hydrogen and alkyl groups with at least one of such substituents being alkyl.

14. A process for improving the flavor of a tobacco product which comprises adding thereto a small amount of a compound selected from the group of compounds having the formula



wherein R_1 and R_2 are selected from the group consisting of hydrogen and alkyl groups of 1 to 4 carbon atoms with at least one of such substituents being alkyl.

15. The process of claim 13 wherein the compound is added in an amount from about 0.001 to 2.0 percent by weight of the product.

References Cited

UNITED STATES PATENTS

2,766,147	10/1956	Rowland	131-17
2,872,360	3/1959	Teague	131-264 X

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