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SMOKING TOBACCO PRODUCT HAVING MENTHYL KETO ESTER ADDITIVE

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This invention relates to an improved smoking tobacco product having improved smoking taste, flavor and aroma characteristics and to the method of making same.

The use of natural and synthetic menthol as an additive in smoking tobacco products is of increasing importance to the tobacco products industry. However, the use of menthol has several disadvantages and difficulties when used as an additive for tobacco products. Due to its volatility, the manufacturing rooms of the tobacco products factory and also the unmentholated tobacco products are frequently contaminated by menthol, thus resulting in low plant efficiency and a relatively short shelf life for products made to contain closely regulated quantities of menthol. Also, when smoking a mentholated tobacco product, regardless of the uniformity in menthol loading, the smoker often experiences a nonuniform menthol response.

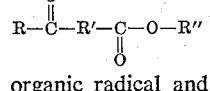
It is a primary object of my invention to overcome the disadvantages and difficulties heretofore encountered and to provide an improved tobacco product having an individual and distinctive taste, flavor and aroma and method of making the product wherein the menthol-containing additive imparts a desirable and pleasing cooling flavor and aroma which substantially eliminates the problem of contamination of the manufacturing rooms and unmentholated products, in which the tobacco products have a protracted shelf life, and in which enhanced organoleptic properties of the additive or its desirable menthol component is released into the smoke stream on smoking the tobacco product at a satisfactorily regulated rate.

My invention contemplates incorporating in tobacco or in a smoking tobacco product an aliphatic-beta, gamma or delta keto acid ester of synthetic, natural, or mixtures of natural and synthetic menthol whereby the tobacco product has improved characteristics of flavor, enhanced taste and in which the organoleptic harshness associated with some grades of tobacco is substantially reduced.

Examples of the keto acids which may be employed as components of the menthyl esters contemplated in this invention are acetoacetic acid, levulinic acid, and γ acetylbutyric acid.

As stated above, I may employ either natural or synthetic or mixtures of natural and synthetic menthol as the menthol component of the menthyl keto esters. Examples of the menthyl esters of aliphatic keto beta, gamma and delta acids which may be used as tobacco products additive in accordance with my invention are menthyl acetoacetate, menthyl levulinate and menthyl γ acetylbutyrate.

The menthyl esters that have been found to improve the flavoring characteristics of tobacco products when incorporated therein may be characterized by the formula:



in which R is an organic radical and which is organoleptically pleasing and beneficial or is at least neutral, R' is an aliphatic carbon chain containing 1 to 3 carbon atoms, and R'' is menthyl (either natural or synthetic or mixtures thereof).

The menthyl esters may be prepared in any desired fashion, thus the keto acids may be reacted directly with

menthol and the menthyl ester fractionally distilled under high vacuum conditions.

Specific examples of the preparation of a suitable menthyl ester of a beta, gamma or delta keto acid which may be used as tobacco products additive in accordance with my invention are as follows:

Example 1

Menthyl acetoacetate may be prepared as follows: 15.6 gms. of menthol are dissolved in 200 cc. of toluene contained in a 250 ml. round bottom reaction flask. To this solution is added 13.0 gms. of ethyl acetoacetate and 10.0 mgms. of p-toluenesulfonic acid catalyst. The mixture is refluxed for 12 hours, water at 90° C. being circulated through the outer shell of the reflux condenser. At the end of this time the reaction mixture is washed with two 25 ml. fractions of 2% sodium bicarbonate solution to remove the catalyst. The toluene solvent is then removed by flash evaporation and the product eluted with chloroform from a 20 x 1 cm. column of neutral alumina. After evaporation of the chloroform, the product is recrystallized from petroleum ether at —20° C. M.P. 33° C.; M.P. literature 30–32° C. Yield 21.08 gms.; 90% of theoretical.

Example 2

Menthyl levulinate is prepared as follows: 11.6 gms. of levulinic acid is dissolved in 50 cc. of benzene in a 125 cc. single neck round bottom flask. To this mixture is added 15.6 gms. of menthol and 0.05 cc. of concentrated sulfuric acid catalyst. The esterification is carried out azeotropically and the mixture refluxed for 8½ hours to collect the theoretical 1.8 cc. of water. After this time, the reaction mixture is washed with two 25 cc. portions of 2% sodium bicarbonate solution and one 25 cc. fraction of distilled water. The product is distilled at 0.075 mm. HgA. pressure; B.P. 79–80° C.;

$$[\alpha]_D^{23}=60.45°$$

$\eta_D^{20}=1.4594$; $d_4^{23}=0.972$; yield 21.0 gms. 82.7% of theoretical. Elemental analysis: 2,4-dinitrophenylhydrazone, calculated for $C_{21}H_{30}O_6N_4$: C, 58.05; H, 6.96; N, 12.90. Found: C, 57.83; H, 6.84; N, 13.4.

Example 3

Menthyl γ acetylbutyrate is prepared as follows: 9.8 gms. of γ acetylbutyric acid is dissolved in 30.0 cc. of benzene in a 50 cc single neck round bottom flask. To this mixture is added 11.7 gms. of menthol and two drops of concentrated sulfuric acid catalyst. The esterification is carried out azeotropically and refluxed for 15 hours to collect 1.1 cc. (1.3 theoretical) of water. After this time, the reaction mixture is washed with 50 cc. of 2% sodium bicarbonate solution and 25 cc. fraction of distilled water. The product is distilled at 0.075 mm. Hg A. pressure; B.P. 84° C.; $[\alpha]_D^{23}=57.79$; $\eta_D^{20}=1.4605$; $d_4^{23.5}=0.978$; yield 17.0 gms.; 84.5% of theoretical. Elemental analysis: 2,4-dinitrophenylhydrazone, calculated as $C_{22}H_{32}O_6N_4$: C, 58.91; H, 7.19; N, 12.49. Found: C, 58.88; H, 7.24; N, 12.57.

It should be understood that in the Examples 1 to 3 set forth above, any organic aliphatic keto acid in accordance with my invention may be used and substituted for the acids indicated in the examples and the proportions adjusted in accordance with the molecular weight thereof. The menthyl esters may be incorporated in the tobacco product either alone or in mixture with menthol. The proportion of the menthyl ester incorporated in the tobacco product may be widely varied in accordance with taste, but I have found that satisfactory results are obtained if the proportion by weight on a dry basis is between 0.005 gm. and 0.017 gm. of the ester per gram of smoking tobacco.

Any convenient method for incorporating the menthyl keto ester in a tobacco product may be employed. Thus the ester may be dissolved in a suitable solvent and either sprayed on the cured, cased and blended tobacco or the tobacco dipped therein. Also, a stabilized aqueous suspension of the ester may be prepared which, in turn, may be sprayed on the cured, cased and blended tobacco or the tobacco dipped therein. Under certain conditions the menthyl ester may be applied by a suitable applicator to the paper or leaf wrapper of a tobacco product.

Specific examples of methods of incorporating the menthyl keto esters in a tobacco product are as follows:

Example 4

Approximately 5.4 oz. of menthyl acetoacetate is dissolved in one pint of absolute ethanol and the solution is sprayed on approximately 30 pounds of cured, cased and blended commercial tobacco. The tobacco so treated is manufactured into cigarettes using normal factory procedures and equipment.

Example 5

Approximately 0.27 oz. of menthyl levulinate is dissolved in one pint of absolute ethanol and the solution is sprayed on approximately 30 pounds of cured, cased and blended commercial tobacco. The tobacco so treated is manufactured into cigarettes using normal factory procedures and equipment.

Example 6

Approximately 7.10 oz. of menthyl γ acetylbutyrate is dissolved in one pint of absolute ethanol and the solution is sprayed on the cured, cased and blended commercial tobacco. The tobacco so treated is manufactured into cigarettes using normal factory procedures and equipment.

The tobacco products made in accordance with the specific examples given herein have been evaluated and tested organoleptically, the smoking products were found to deliver smoke having a pleasing and cooling taste and aroma characteristics of menthol. Additionally, the flavor was noted to have a highly pleasing quality which lacked the harshness generally associated with some grades of tobacco and when smoked the individual keto esters employed had distinctive taste, flavor and aroma characteristics.

Modifications may be made in the described embodiments of my invention and in the illustrated examples without departing from the scope of the invention set forth in the accompanying claims.

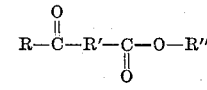
I claim:

1. A smoking tobacco product having improved and distinctive taste, flavor and aroma characteristics comprising a smoking tobacco having incorporated therein an ester of menthol and an organic keto acid whereby the hardshness associated with the smoke of a tobacco product is substantially eliminated.

2. A smoking tobacco product having improved and distinctive taste, flavor and aroma characteristics comprising a smoking tobacco having between 0.005 and 0.017 gm. of a menthyl keto ester whereby the harshness associated with the smoke of a tobacco product is substantially eliminated.

3. A smoking tobacco product having improved and distinctive taste, flavor and aroma characteristics comprising a smoking tobacco having incorporated therein a material selected from the group consisting of menthyl acetoacetate, menthyl levulinate and menthyl γ acetylbutyrate whereby the harshness associated with the smoke of a tobacco product is substantially eliminated.

4. A smoking tobacco product having improved and distinctive taste, flavor and aroma characteristics comprising a smoking tobacco having incorporated therein an ester of menthol and an organic keto acid having the formula



in which R is an organic radical, R′ is an aliphatic carbon chain containing 1 to 3 carbon atoms and R″ is menthyl.

5. A smoking tobacco product having improved and distinctive taste, flavor and aroma characteristics comprising a smoking tobacco having incorporated therein menthyl acetoacetate whereby the harshness associated with the smoke of a tobacco product is substantially eliminated.

6. A smoking tobacco product having improved and distinctive taste, flavor and aroma characteristics comprising a smoking tobacco having incorporated therein menthyl levulinate whereby the harshness associated with the smoke of a tobacco product is substantially eliminated.

7. A smoking tobacco product having improved and distinctive taste, flavor and aroma characteristics comprising a smoking tobacco having incorporated therein menthyl γ acetylbutyrate whereby the harshness associated with the smoke of a tobacco product is substantially eliminated.

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