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3,312,226

SMOKING TOBACCO COMPOSITION

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This invention relates to smoking compositions. More particularly, the present invention relates to a composition for embodying predetermined flavors in tobacco, which flavors can be maintained and preserved during subsequent processing and storage of the tobacco.

It is an object of this invention to permit the incorporation of a flavor into a tobacco product, which flavor will not be lost or altered during subsequent manufacturing steps or during storage of the tobacco product.

It is a further object of this invention to permit the incorporation of a material in tobacco, which material will release one or more flavorants into the tobacco smoke which results when the tobacco containing said material is smoked.

It is a further object of the present invention to control the amount of flavor or flavors released during the smoking of a tobacco product to insure uniformity of flavor during the entire smoking process.

It is a still further object of the present invention to provide a flavoring composition which is uniquely suited for use in tobacco products.

One of the more specific objects of the present invention is to incorporate menthol in a tobacco product in such a manner that it will not be released prior to the time that the tobacco product is smoked but will be readily and efficiently released as the tobacco product is smoked.

Another of the more specific objects of the present invention is to incorporate an additive in tobacco which, when the tobacco is smoked, will not only release menthol but will also release one or more additional flavorants.

Numerous methods of adding flavorants to tobacco smoke are known. However, none of the known methods has been found to be completely satisfactory, particularly when the flavorant is menthol.

When menthol per se is added to tobacco, the loss of menthol during the manufacturing process and during storage is very high, due to the highly volatile nature of the menthol. In addition to the undesirable loss of the menthol, objectionably strong and irritating menthol vapors are often encountered in the actual manufacturing process wherein the menthol is incorporated in cigarettes.

When menthol has been adsorbed on an adsorbent, such as activated charcoal or fuller's earth, and applied to tobacco, the menthol yield when such tobacco is smoked has been found to be very low. In addition, such a process results in the incorporation in the tobacco of a foreign material which can give an undesirable appearance to the tobacco and which can result in uneven burning of the tobacco.

Menthol has also been incorporated in tobacco in the form of a clathrate. However, such a method for incorporating menthol in tobacco has been not only expensive but also inefficient, since the yield of menthol when tobacco containing such clathrates is burned has been found to be very low.

Esters of menthol, such as menthol succinate and menthol borates have also been prepared and added to tobacco. However, the yield of menthol when the tobacco product containing such succinates or borates is smoked has also been found to be very low. Furthermore, such a method results in the addition to the smoke of undesirable combustion products.

The present invention overcomes the disadvantages of

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the prior art and provides for the incorporation of a flavorant, and particularly menthol, in tobacco in such a manner that the flavorant is not lost during manufacturing and storage and yet is readily released when the tobacco is smoked. If desired, more than one flavorant may, in accordance with the invention, be simultaneously incorporated in the tobacco.

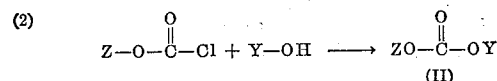
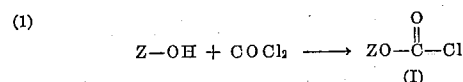
The additives which are employed in accordance with the present invention are relatively non-volatile. As a result, it is possible to prevent the loss of a flavorant during manufacturing operations or during storage of the product. For example 1-menthyl chlorocarbonate, when applied to tobacco, is stable and it not lost due to volatilization under conditions of either high or low humidity. In addition, since the flavorant is not present in free form on the filler, it is possible to prevent the transfer of the flavorant from one cigarette to another or from one pack of cigarettes to another.

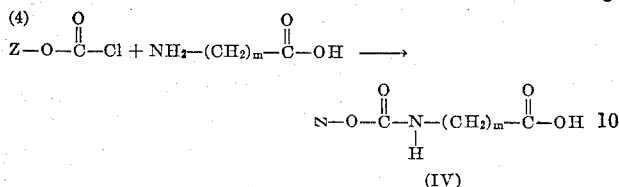
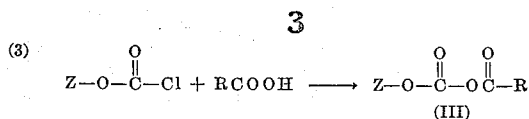
The present invention also results in a tobacco product from which a high delivery of flavorants into the smoke can be achieved without the production of undesirable pyrolysis products and without the addition of undesirable foreign materials to the tobacco.

In accordance with the present invention, a flavor-containing carbonate is incorporated in a tobacco product, and particularly in the filler of cigarettes.

The flavor-containing carbonate of this invention may be any organic carbonate which (1) contains at least one flavor moiety, i.e. the flavor-imparting portion of a flavorant, which portion, when present in burning tobacco, is converted to a flavorant; (2) is substantially non-volatile under normal atmospheric conditions, e.g. temperatures of 20-30° C. and atmospheric pressures; (3) is capable of being broken down to yield a flavor moiety when incorporated in and subjected to the temperature of burning tobacco, which will generally be temperatures of from about 100° C. to about 1200° C. The organic carbonate may for example be a dicarbonate or a halocarbonate. The carbonates can be readily prepared from the reaction of suitable alcohols with carbonyl chloride which, when the reaction is conducted at low temperatures, results in the production of chloro-carbonic esters. At higher temperatures and in the presence of excess alcohol, a diester of carbonic acid is produced.

The following equations are illustrative of the preparation of certain dicarbonates and halocarbonates which may be employed in accordance with the present invention. In the equations, Z is the flavorful moiety of a flavor alcohol, Z-OH, for example menthol, and Y is a radical derived from an alcohol which may, for example, be a simple alcohol such as a lower alkyl alcohol. Preferably, however, Y is the flavor moiety of a flavor alcohol, Y-OH, which may be identical to Z-OH or may be a different flavor alcohol. Thus, as used in this specification Z will always represent a flavorful moiety but Y may represent either a flavorful moiety or a non-flavorful moiety. Furthermore, as used herein, "flavor alcohol" means an alcohol which contains a flavor moiety and is either itself a flavorant or will release a flavorant when incorporated in and subjected to the temperatures of burning tobacco. Metal alkoxides, wherein the hydrogen of the alcohol is replaced by a monovalent metal, such as sodium or potassium, may be substituted for the flavor alcohols.



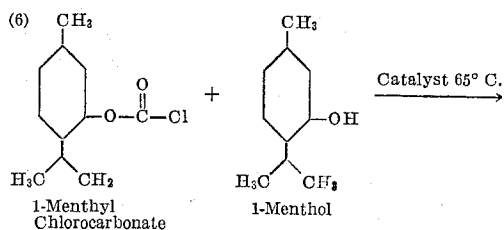
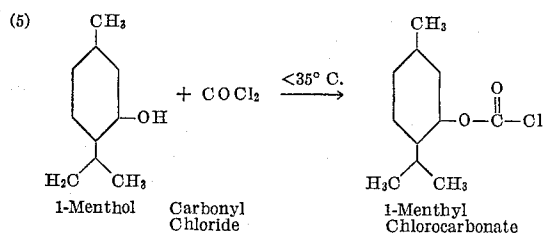


Equation 1 illustrates the preparation of a halocarbonate which can be employed in accordance with the present invention. The product of this equation, Formula I, can be employed as the flavor-containing carbonate or it can be reacted with a flavor alcohol, as illustrated in Equation 2 to produce a dicarbonate. When the flavor alcohol is the same as the flavor alcohol employed to produce the chlorocarbonate, only one flavorful moiety will be present in the resulting dicarbonate. However, when a different flavor alcohol is employed, the resulting dicarbonate will have two flavor moieties.

The flavor-containing halocarbonate produced in accordance with Equation 1 may also be reacted with a flavor-containing carboxylic acid, as in Equation 3 or with a flavor-containing amino acid, as in Equation 4. The alkylacylcarbonate (Formula III) produced in accordance with Equation 3 and the acyloxy amino acid derivative (Formula IV) produced in accordance with Equation 4 are also useful as flavor-containing carbonates in accordance with the present invention.

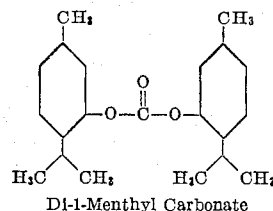
In Equations 1-4, Z and Y are, as set forth above, flavor moieties and may be an organic radical, for example an aromatic or aliphatic group, containing from 1 to about 100 carbon atoms, R is an organic radical, for example an aromatic, aliphatic or cycloaliphatic group, containing from 1 to about 100 carbon atoms, and m is an integer having the value of from about 1 to 10. In Equation 4, the (CH₂)_m portion of the molecule can be exactly as it is shown or one or both of the hydrogens at one or more positions along the chain can be replaced by an organic radical, for example a straight or branched chain aliphatic radical having from 1 to 8 carbon atoms or more.

As a specific illustration of Reactions 1 and 2, the treatment of 1-menthol with carbonyl chloride leads to the formation of 1-menthyl chlorocarbonate. When the 1-menthyl chlorocarbonate is contacted at higher temperatures, with an excess of 1-menthol, di-1-menthyl carbonate is formed as shown in the following equations:



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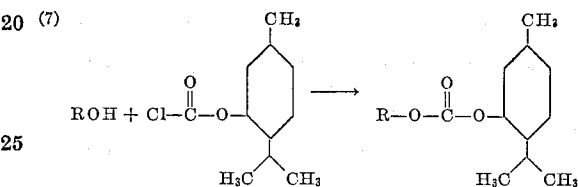
(6)



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Other flavor alcohols may be employed instead of menthol in Equations 1 and 2. In addition, alcohols which are flavorants and/or alcohols that decompose to yield flavorants can be reacted with 1-menthyl chlorocarbonate. The resulting compounds can decompose to the parent alcohol (a flavorant in itself) and to 1-menthol or they can rearrange to give a second flavorful note plus menthol. A general equation for the reaction of alcohols with 1-menthyl chlorocarbonate is given below.

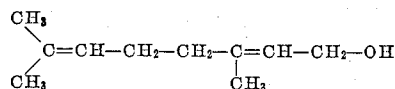
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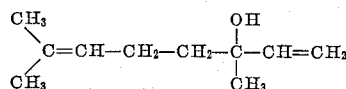
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Glycols, glycerol, terpene alcohols, sugars and the like are illustrative of flavor alcohols which can be employed in accordance with the present invention. These include, for example:

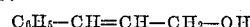
Geraniol:



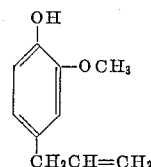
Linalool:



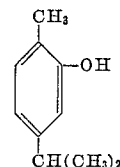
Cinnamyl alcohol:



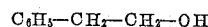
Eugenol:



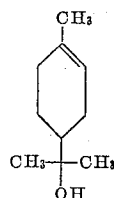
Carvacrol (Isoeuthymol):



Phenylethyl alcohol:



Terpineol:

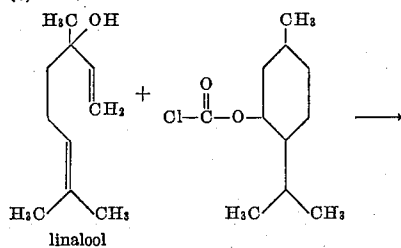


As an illustration of how one of these compounds can be employed, linalool when reacted with 1-menthyl chlorocarbonate forms a compound which rearranges when

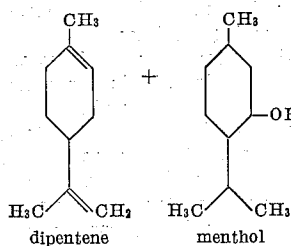
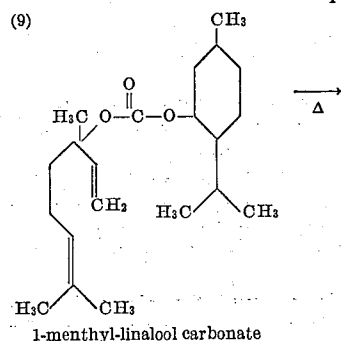
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the tobacco product is burned to give menthol and dipentene in the smoke. These reactions are shown below:

(8)



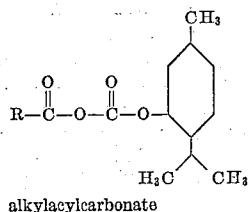
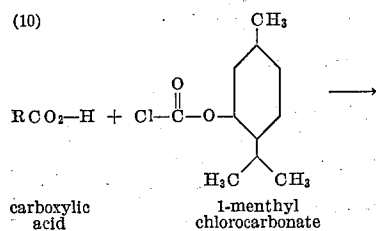
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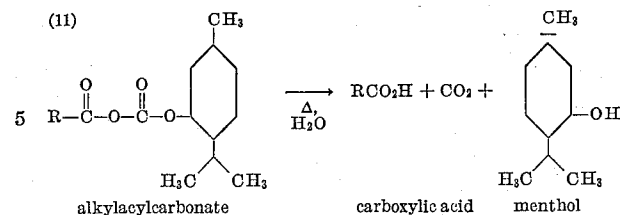
Terpineol-, geraniol-, and nerol- 1-menthyl carbonates also degrade to give dipentene and 1-menthol.

Flavorful carboxylic acids such as lactic, pyruvic, citric, and tartaric acids will react with 1-menthyl chlorocarbonate in this manner, and break down to give the flavorful acid and menthol in smoke. This is illustrated below:

(10)



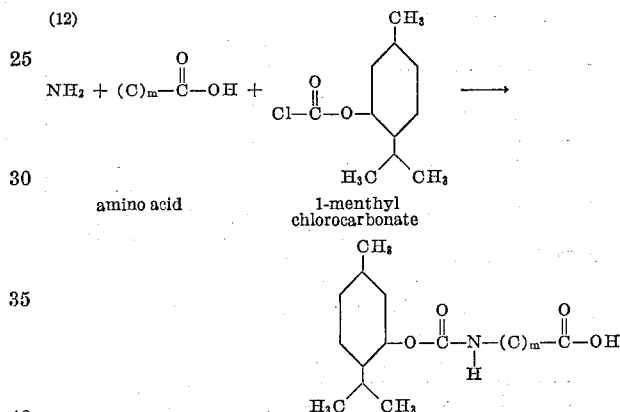
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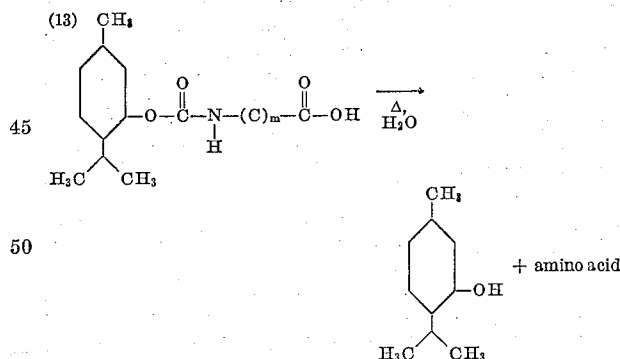
10 In a similar manner, the sugar acids such as glycolic acid, levulinic acid, and gluconic acid and amines such as diphenylamine and methylantranilate (which provide a floral and rose note, respectively) will react and the formed product will decompose to give flavorful and aromatic substances into the smoke.

15 Amino acids, particularly lysine, glycine, proline and asparagine will also react with 1-menthyl chlorocarbonate giving compounds which will decompose during pyrolysis of the tobacco product into which they have been incorporated to yield menthol and other flavorful and aroma-producing materials in the smoke. This is illustrated by the following equations:

(12)



(13)



wherein m is an integer having the value of from about one to ten. In a similar manner, tobacco alkaloids can be reacted with 1-menthyl chlorocarbonate and added to tobacco.

60 The above-described reactions can be conducted under the usual conditions which are employed for the preparation of such carbonates. More specifically, the reaction of Equation 1 can be conducted at a temperature of from about -60°C . to about 60°C . for a period of from about 15 minutes to about 24 hours. Generally, the reaction is conducted at atmospheric pressure, although higher or lower pressures may be employed. A solvent or cosolvent, such as benzene, chloroform, dioxane and toluene may be employed if desired. If desired, from about 1 to about 10 percent of a catalyst, such as pyridine or antipyrine, may be employed during the reaction.

70 The reaction of Equation 2 can be conducted at a temperature of from about -60° to about 85°C . for a period of from about 30 minutes to about 24 hours. Generally, the reaction is conducted at atmospheric pressure,

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although higher or lower pressures may be employed. A solvent or cosolvent, such as benzene or dioxane may be employed if desired. If desired, from about 1 to about 10 percent of a catalyst, such as pyridine or antipyrine, may be employed during the reaction.

The reaction of Equation 3 can be conducted at a temperature of from about -60° to about 100° C. for a period of from about 30 minutes to about 24 hours. Generally, the reaction is conducted at atmospheric pressure, although higher or lower pressures may be employed. A solvent or cosolvent, such as benzene, tetrahydrofuran or toluene, may be employed if desired. If desired, from about 1 to about 10 percent of a catalyst, such as pyridine or triethylamine, may be employed during the reaction.

The reaction of Equation 4 can be conducted at a temperature of from about -20 to about 60° C. for a period of from about 10 minutes to about 30 hours. Generally, the reaction is conducted at atmospheric pressure, although higher or lower pressures may be employed. A solvent or cosolvent, such as benzene, xylene, toluene, chloroform, or dioxane, may be employed if desired. If desired, from about 1 to about 10 percent of a catalyst, such as pyridine or antipyrine, may be employed during the reaction.

The reaction of Equation 5 can be conducted at a temperature of from about -60° to about 60° C. for a period of from about 15 minutes to about 24 hours. Generally, the reaction is conducted at atmospheric pressure, although higher or lower pressures may be employed. A solvent or cosolvent, such as benzene, xylene, toluene or dioxane, may be employed if desired. If desired, from about 1 to about 10 percent of a catalyst, such as pyridine or antipyrine, may be employed during the reaction.

The reaction of Equation 6 can be conducted at a temperature of from about 50° to 150° C. for a period of from about 1 hour to about 24 hours. Generally, the reaction is conducted at atmospheric pressure, although higher or lower pressures may be employed. A solvent or cosolvent, such as benzene, xylene, toluene or dioxane, may be employed if desired. If desired, from about 1 to about 10 percent of a catalyst, such as pyridine or antipyrine, may be employed during the reaction.

The reaction of Equation 7 can be conducted at a temperature of from about -60° to about 150° C. for a period of from about 30 minutes to about 24 hours. Generally, the reaction is conducted at atmospheric pressure, although higher or lower pressures may be employed. A solvent or cosolvent, such as benzene, toluene or xylene, may be employed if desired. If desired, from about 1 to about 10 percent of a catalyst, such as pyridine or antipyrine, may be employed during the reaction.

The reaction of Equation 8 can be conducted at a temperature of from about -60° to about 60° C. for a period of from about 15 minutes to about 24 hours. Generally, the reaction is conducted at atmospheric pressure, although higher or lower pressures may be employed. A solvent or cosolvent, such as benzene, toluene or xylene, may be employed if desired. If desired, from about 1 to about 10 percent of a catalyst, such as pyridine or antipyrine, may be employed during the reaction.

The reaction of Equation 9 can be conducted at a temperature of from about 100° to about 1200° C. for a period of from about 0.1 second to about 24 hours. Generally, the reaction is conducted at atmospheric pressure, although higher or lower pressures may be employed.

The reaction of Equation 10 can be conducted at a temperature from about -60° to about 100° C. for a period of from about 30 minutes to about 2 hours. Generally, the reaction is conducted at atmospheric pressure, although higher or lower pressures may be employed. A solvent or cosolvent, such as benzene, toluene or xylene, may be employed if desired. If desired, from about 1 to about 10 percent of a catalyst, such as pyridine or antipyrine, may be employed during the reaction.

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The reaction of Equation 11 can be conducted at a temperature of from about 100° to about 1200° C. for a period of from about 0.1 second to about 24 hours. Generally, the reaction is conducted at atmospheric pressure, although higher or lower pressures may be employed. A solvent or cosolvent, such as benzene, toluene or xylene, may be employed if desired. If desired, from about 1 to about 10 percent of a catalyst, such as pyridine or antipyrine, may be employed during the reaction.

The reaction of Equation 12 can be conducted at a temperature of from about -20° to about 60° C. for a period of from about 10 minutes to about 30 hours. Generally, the reaction is conducted at atmospheric pressure, although higher or lower pressures may be employed. A solvent or cosolvent, such as benzene, xylene, toluene, chloroform or dioxane, may be employed if desired. If desired, from about 1 to about 10 percent of a catalyst, such as pyridine, may be employed during the reaction.

The reaction of Equation 13 can be conducted at a temperature of from about 100° to about 1200° C. for a period of from about 0.1 second to about 24 hours. Generally, the reaction is conducted at atmospheric pressure, although higher or lower pressures may be employed. A solvent or cosolvent, such as benzene, toluene or xylene, may be employed if desired. If desired, a catalyst, such as pyridine, may be employed during the reaction.

The materials produced in accordance with the above reactions may, after they have been produced, be purified in the usual manner, for example by distillation, crystallization and the like.

The flavor-containing carbonates of the present invention can, when they are liquids, be applied to the tobacco by directly spraying them on the tobacco. They can also be applied by dispersing or dissolving them in a suitable carrier, for example water, organic solvents and other carriers known in the art or may be directly mixed or otherwise combined with the tobacco by methods which are known in the art.

When a liquid carrier is employed, the mixture is preferably air-dried or otherwise treated to remove the carrier after the flavor-releasing carbonate has been thoroughly admixed with the tobacco.

The flavor-containing carbonates of the present invention can, if desired, be incorporated in cigarette paper or in cigar wrappers or the like. However, it will generally be more desirable to incorporate these materials in the tobacco itself. So long as the tobacco-containing carbonates are placed in a tobacco product in such a manner that they are in contact with the tobacco when it is burned, they will release their flavors upon smoking of the tobacco product.

While the amount of flavor-containing carbonate to be added to the tobacco product will vary depending upon the amount of flavor which is desired in the smoke, the flavor-containing carbonates will generally be added to the tobacco in an amount of from about 0.1 to 10% by weight of the total tobacco composition.

The following examples are illustrative:

Example 1

About 312 grams of Brazilian 1-menthol was reacted at a temperature of $10-20^{\circ}$ C. with 198 grams of carbonyl chloride, in the presence of 150 ml. of benzene as a cosolvent and of a catalytic amount (5 grams) of pyridine. This mixture was stirred for a total of 3 hours and allowed to stand overnight. At the end of this period, the reaction was terminated and 1-menthyl chlorocarbonate was recovered as a slightly yellow, mobile liquid. Rectification of the recovered material by vacuum distillation resulted in a distillate which was a water-white liquid, having a boiling point of $105^{\circ}-106^{\circ}$ at 12 mm. Hg. About 424 grams (97 percent over-all yield) of 1-menthyl chlorocarbonate was obtained. This material was stable

to boiling water, aqueous acids, and to bases and was found to be soluble in a variety of organic solvents, including such classes as alcohols, ethers, hydrocarbons, esters and halogenated hydrocarbons. It was stable to heat, at least to a temperature of 150° C. It was found to react slowly with lower alcohols (e.g. ethanol) and to react vigorously with organic bases to form a complex. This complex was readily decomposed with water at elevated temperatures to yield 1-menthol.

Example 2

Shredded tobacco (5 lbs.) containing no menthol (but otherwise having been prepared for the manufacture of commercially available mentholated cigarettes) was placed in a rotary drum and sprayed with a solution consisting of 11.1 g. of 1-menthyl chlorocarbonate in 200 ml. absolute ethanol. The sprayed tobacco was then stored for three days in polyethylene bags to allow the additive to penetrate the tobacco shreds. The material was air-dried on screens at 60 percent relative humidity and a temperature of 75° F., until the solvent alcohol was lost. No appreciable odor of menthol was observed on the tobacco. The tobacco was made into cigarettes, using a standard machine which added to the shredded tobacco section of the cigarette a 15 mm. (in length) filter. Control cigarettes were prepared in a similar fashion, except that 3.2 milligrams of Brazilian 1-menthol were added to each cigarette, in place of the 1-menthyl chlorocarbonate. The two batches of cigarettes were separately stored in glass jars at 38° F.

The test and control cigarettes were smoked on a standard smoking machine. The smoke was collected on Cambridge filter pads and extracted with ethanol. The details of the tests are given below:

Lucite holders were packed with Cambridge filter pads and individually weighed on a Gram-atic balance. The holders were inserted into the correct ports on a standard 20 port smoking machine, the ports being selected by randomization, or patterning of the test samples and checked for leaks.

The cigarettes were then inserted to a depth of 5 mm. in the hole of the dental dam cover of the Lucite holders, lit with an electric lighter or alcohol torch and smoked to a 25 mm. butt length. Four monitor samples were smoked along with the test cigarettes for every smoking run.

As the char line of a cigarette reached the 25 mm. mark, the coal was cut off with a pair of surgical scissors. The dead butt was left in the dental dam until a clearing puff was taken before another cigarette was inserted. When the fourth cigarette on each port had finished smoking and a clearing puff had been taken, the Lucite holders were removed from the machine.

After all the cigarettes had been smoked, the individual Lucite holders were re-weighed immediately. The Cambridge filter pads were then removed from the holders with a pair of tweezers and folded carefully without touching the deposited T.P.M. with the hands. The pad was folded so that the T.P.M. was on the inside and the Lucite holder was wiped out, using tweezers to hold the pad. The removed Cambridge filter pads were extracted, in each case, with 100 ml. of ethanol.

The menthol was identified and the delivery determined by gas chromatography. The delivery of menthol for the test cigarettes was 420 micrograms/cigt. This represented about an 18% delivery since the 11.1 grams of additive was roughly equivalent to 8 grams of menthol.

Physical tests revealed no significant difference in nicotine delivery, TPM delivery, and "smoke pH" between the test cigarettes and the control cigarettes.

Chemical analyses of the smoke and particulate matter corresponded to those of the control except as noted previously.

Test cigarettes containing 1-menthyl chlorocarbonate were placed in open glass jars and subjected to the follow-

ing cyclic conditions: (1) 110° F. and 25% R.H. for 8 hrs.; (2) 110° F. and 85% R.H. for 16 hrs. and this cycle repeated for the number of days shown in Table I and then the cigarettes were stored at 75° F. and 65% R.H. for about two days, until smoked. The amount of menthol in the smoke was measured as above.

The results were compared with production mentholated cigarettes subjected, as controls, to identical conditions. The results are given in Table I below:

10 TABLE I.—MENTHOL DELIVERIES UPON SMOKING CONTROL VS. TEST CIGARETTES IN OPEN JARS

Time (days)	Mg. Menthol/Cigarette	
	Control Cigarettes	Test Cigarettes
0	0.54	0.40
1	0.39	0.37
2	0.39	0.32
3	0.33	0.28
4	0.32	0.31
5	0.33	0.31

Percent Loss Menthol after 5 days:

Control cigarettes

$$\frac{.54-.33}{.54} \times 100 = \frac{21}{.54} = 39\%$$

Test cigarettes

$$\frac{.40-.31}{.40} \times 100 = \frac{9}{.40} = 22.5\%$$

Production mentholated cigarettes having the same production date were obtained. These cigarettes contained 3.2 milligrams menthol/cigt. Half the cigarettes were removed from their packs and were replaced with test cigarettes containing 3.6 milligrams 1-menthyl chlorocarbonate/cigt. Both sets of cigarettes were subjected to the following cyclic conditions:

(1) 110° F.—85% R.H.—4.5 days.

(2) 75° F.—65% R.H.—2.5 days.

(3) 110° F.—25% R.H.—4.5 days.

(4) 75° F.—65% R.H.—2.5 days.

(5) 110° F.—85% R.H.—4.5 days.

Table II is a summary of the data obtained:

45 TABLE II.—MENTHOL DELIVERIES UPON SMOKING PRODUCTION CIGARETTES VS. TEST CIGARETTES—STORAGE TESTS

Time (weeks)	Mg. Menthol/Cigarette	
	Production Cigarettes	Test Cigarettes
0	0.54	0.40
1	0.49	0.37
2	0.47	0.36
3	0.44	0.36
4	0.46	0.36
5	0.35	0.36
6	0.42	0.40
7	0.37	0.36
8	0.39	0.37
9	0.38	0.37
10	0.38	0.37

Example 3

Test cigarettes containing 3.6 milligrams of 1-menthyl chlorocarbonate/cigt. were smoked in comparison with two commercially available mentholated cigarettes (Alpine brand cigarettes and Salem brand cigarettes) by two smoking panels, one consisting of 34 members and the second consisting of 31 members. The results of these tests show that the 34 member panel had a 20 to 14 preference for the test cigarettes over the Alpine brand cigarettes. The test cigarettes were also found by the 31 member panel to have more tobacco flavor and less harshness than the Salem brand cigarettes, as shown by a preference of 21 to 10.

Example 4

Carbonyl chloride (198 grams) was reacted with 312 grams of 1-menthol, in the same manner as in Example 1, to produce 1-menthyl chlorocarbonate. Five pounds of commercial shredded tobacco filler which contained no menthol (Alpine filler except that the menthol normally added to the shredded tobacco had not been added) was sprayed in a manner similar to that described in Example 1 with 25.0 grams of 1-menthyl chlorocarbonate which were dissolved in 200 ml. of absolute alcohol. The sprayed product was then stored for three days in polyethylene bags to allow the 1-menthyl chlorocarbonate to penetrate the tobacco shreds. The material was then air-dried on screens at 60% relative humidity and a temperature of 75° until the absolute ethanol had all evaporated. Test cigarettes were prepared and smoked in comparison with control cigarettes in the same manner as is set forth in Example 1. Similar results were obtained except that the menthol delivery into the smoke of the test cigarettes was found to be 820 micrograms per cigarette. Subjective evaluation of the cigarettes showed that the menthol was readily detected in the smoke and had transferred well into the smoke.

Example 5

Linalool (11 grams) was reacted with 7.7 grams of 1-menthyl chlorocarbonate at a temperature of 35° C. and an atmospheric pressure of 14.7 p.s.i.a. for a period of 18 hours to form 1-menthyl linalool carbonate, which was found to be a solid material having a melting point of 106.6° F., which was soluble in organic solvents, such as ethanol, benzene, ethyl acetate, and which was stable to boiling water.

The 1-menthyl linalool carbonate (1 gram) was dissolved in 35 ml. of ethanol and the resulting solution was sprayed on commercial tobacco filler containing no menthol, in a manner that 3.2 milligrams of menthol of the 1-menthyl linalool carbonate were incorporated in each cigarette (Alpine filler which had not been treated with menthol). The sprayed tobacco was stored for three days in polyethylene bags to permit the 1-menthyl linalool carbonate to penetrate the tobacco shreds. The resulting treated tobacco was then air-dried on screens at 60% relative humidity and a temperature of 75° F. until the absolute ethanol had evaporated. The treated filler was made into cigarettes on a standard machine which incorporated a 15 mm. filter in each cigarette. Control cigarettes were prepared in a similar fashion, except that 3.2 milligrams of Brazilian 1-menthol was employed in each cigarette in place of the 1-menthyl linalool carbonate.

The test and control cigarettes were smoked on a standard smoking machine and the smoke was collected in the same manner as described in Example 1. The menthol was extracted from the TPM pads with ethanol and was identified by gas chromatography. Vacuum distillation followed by gas chromatography was employed in order to characterize the flavorants that were present in the smoke. By infrared, ultraviolet and mass spectrometry, dipentene was identified as the second flavorant which had been released by pyrolysis of the cigarette containing 1-menthyl linalool carbonate.

Subjective evaluation of the test cigarettes showed that

these cigarettes had the distinctive flavor of menthol and of the citrus peel, a flavor which is characterized by dipentene. Thus, the evaluation shows that the 1-menthyl linalool carbonate was broken down upon pyrolysis of tobacco into two flavorants: menthol and dipentene, both of which had transferred well into the smoke phase from the test cigarettes.

Example 6

Shredded tobacco (300 lbs.) containing no menthol (but otherwise having been prepared for the manufacture of commercially available mentholated cigarettes) was placed in a rotary drum and sprayed with a solution consisting of 840 g. of 1-menthyl chlorocarbonate in 6 liters absolute ethanol. The sprayed tobacco was then stored for three days in polyethylene bags to allow the additive to penetrate the tobacco shreds. The material was air-dried on screens at 60 percent relative humidity and a temperature of 75° F., until the solvent alcohol was lost. No appreciable odor of menthol was observed on the tobacco. The tobacco was made into cigarettes, using a standard machine which added to the shredded tobacco section of the cigarette a 15 mm. (in length) filter. Control cigarettes were prepared in a similar fashion, except that 3.2 mgs. of Brazilian 1-menthol were added to each cigarette, in place of the 1-menthyl chlorocarbonate. The two batches of cigarettes were packaged in plastic packs.

The test and control cigarettes were smoked on a standard smoking machine. The smoke was collected on Cambridge filter pads and extracted with ethanol. The chemical and physical data obtained were as follows:

Smoke	Experimental	Control
TPM, mg./cigarette.....	26.4	26.7
Nicotine, mg./cigarette.....	1.27	1.20
Menthol, mg./cigarette.....	0.47	0.48

We claim:

1. A smoking tobacco composition comprising smoking tobacco and from about 0.1 to 10% by weight, based on the total tobacco composition of 1-menthyl chlorocarbonate.
2. A smoking tobacco composition comprising smoking tobacco and from about 0.1 to 10% by weight, based on the total tobacco composition of 1-menthyl linalool carbonate.
3. A smoking tobacco composition comprising smoking tobacco and from about 0.1 to 10% by weight, based on the total tobacco composition, of a member selected from the group consisting of 1-menthyl chlorocarbonate and 1-menthyl linalool carbonate.

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