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New York, N.Y.

[54] TOBACCO COMPOSITION ON THE LIKE
INCORPORATING A POLYISO-PRENOID
ALCOHOL FLAVORSANT
4 Claims, No Drawings

[52] U.S. Cl. 131/17,
131/144
[51] Int. Cl. A24b 15/04
[50] Field of Search 131/2, 17,
140, 144, 143

[56]

References Cited

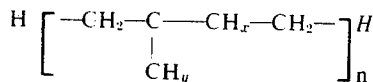
UNITED STATES PATENTS

3,139,888 7/1964 Grossman et al. 131/17

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ABSTRACT: Tobacco products having improved flavor and smoking characteristics comprising tobacco and at least one polyisoprenoid hydrocarbon with optional polyisoprenoid esters and/or alcohols, the effective polyisoprenoid hydrocarbon may be represented by the formula.



Where n is an integer of from about 4 to 15 inclusive and the dotted line means that, at the position indicated, the bond can be either a single or a double bond, one double bond being present, and where x is an integer of 1 or 2 and y is an integer of 2 or 3, the value of x and y being determined by the position of the double bond.

TOBACCO COMPOSITION ON THE LIKE INCORPORATING A POLYISOPRENOID ALCOHOL FLAVORSANT

This application is a division of copending U.S. Pat. Application Ser. No. 433,174, filed Feb. 16, 1965, now Pat. No. 3,542,677.

This invention has to do with a tobacco or tobacco-like product having improved smoking characteristics. More particularly, it relates to enhancing the flavor of tobacco containing products.

The term "tobacco" as used in this specification and the appended claims means and includes, where the context permits, natural tobaccos such as burley, Turkish tobacco, Maryland tobacco, Virginia tobacco, and flue-cured tobacco; tobacco-like products such as reconstituted tobacco or homogenized tobacco; and tobacco substitutes intended to replace natural tobacco such as various vegetable leaves such as lettuce and cabbage leaves.

Recently there has been a marked trend in the tobacco industry toward the use of filters in cigarettes, cigars and pipes. In addition, applications have been found for reconstituted or homogenized tobaccos and for synthetic wrappers and fillers as substitutes for natural tobacco. Such smoking products have suffered from poor flavor characteristics. They are generally regarded as harsh, as containing undesirable aroma quality, and, they otherwise are considered as having poorer smoking flavor and less pleasant smoking character. These innovations have magnified the need for improving the flavor of tobacco products and otherwise enhancing the smoking characteristics of these products. Of course, improved flavor may be obtained by the use of higher grade tobacco leaf. However, this is costly and there has been increased use of tobacco stems and poorer grades of tobacco in various blends.

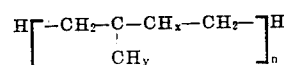
It has been suggested that various additives or flavoring agents be incorporated with, or added to, tobacco products to improve their flavor. For example, Grossman et al., U.S. Pat. No. 3,139,888, suggests the use of certain isoprenoid alcohols to impart a desirable flavor note to tobacco. These additives and agents have not been uniformly successful, and have suffered from a number of disadvantages. The main disadvantages of the additives and agents previously proposed for this purpose have been their relative unavailability, high cost, and inferior aroma and flavor. For example, the polyisoprenoid alcohol additives suggested by Grossman et al., U.S. Pat. No. 3,139,888, have heretofore been obtainable by the extraction of naturally occurring plant sources or by noncommercial laboratory procedures. These processes are expensive. Furthermore, these polyisoprenoid alcohols possess a flowery fragrance and impart undesirable aroma characteristics to tobacco products.

Accordingly, it is an object of this invention to improve the flavor and smoking characteristics of tobacco products.

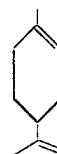
In accordance with this invention it has been found that tobacco materials can be improved in flavor and smoking characteristics by incorporating in, or adding to, such materials a small but effective amount of a polyisoprenoid hydrocarbon or a mixture of a plurality of such polyisoprenoid hydrocarbons. I have also found that tobacco and tobacco-like materials are enhanced in flavor when admixed with a composition comprising certain polyisoprenoid hydrocarbons and certain polyisoprenoid esters or polyisoprenoid alcohols or mixtures of said esters and alcohols. Tobacco or tobacco-like products containing such a polyisoprenoid hydrocarbon, or mixtures of a polyisoprenoid hydrocarbon and a polyisoprenoid ester or a polyisoprenoid alcohol, are less harsh, more mild and have a strong tobacco taste and character and this improved taste, character and flavor is noted although the tobacco is smoked through a filter. Still another aspect of this invention is the preparation of mixtures of the tobacco flavoring agents found useful in accordance with this invention by refining, without degrading, as by molecular distillation, crude mixtures of polyisoprenoid hydrocarbons and polyisoprenoid

esters formed as a residue after removing the conventionally distillable components of the reaction products obtained by the controlled polymerization of isoprene.

The polyisoprenoid hydrocarbons found effective as tobacco flavoring agents are represented by the formula



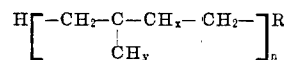
where n is an integer of from about 4 to 15 inclusive and the dotted line means that, at the position indicated, the bond can be either a single or a double bond, one double bond being present, and where x is an integer of 1 or 2 and y is an integer of 2 or 3, the value of x and y being determined by the position of the double bond. The data obtained from Nuclear Magnetic Resonance studies confirms the presence of vinylidene methylene groups as well as of trisubstituted double bonds. Further, some or all of the hydrocarbons formed may contain a terminal menthadiene group of the formula



in place of the terminal hydrogen shown in the above formula. Accordingly the formula given as representative of the polyisoprenoid hydrocarbons contemplates and includes all of the foregoing variations. Such polyisoprenoid hydrocarbons include: 2, 6, 10, 14-tetramethyl- 2, 6, 10, 14-hexadecatetraene; 2, 6, 10, 14, 18, 22, 26-heptamethyl- 2, 6, 10, 14, 18, 22, 26cosaheptaene; 2, 6, 10, 14, 18, 22, 26, 30, 34-nona-methyl- 2, 6, 10, 14, 18, 22, 26, 30, 34-hexatriacontanonaene; 2, 6, 10, 14, 18, 22, 26, 30, 34, 38, 42, 46-dodecamethyl- 2, 6, 10, 14, 18, 22, 26, 30, 34, 38, 42, 46-octatetra-dodecaene, and the double bond isomers thereof. A plurality of such hydrocarbons may be used, and preferably the molecularly distilled or purified mixture of hydrocarbons obtained from the controlled polymerization of isoprene is employed as the tobacco flavoring agent.

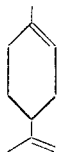
The polyisoprenoid hydrocarbons of this invention and mixtures of such hydrocarbons with polyisoprenoid esters or polyisoprenoid alcohols have been found to possess a combination of properties which make them particularly suitable as tobacco flavoring agents in that such compositions are effective at low concentrations and thus will not bleed into tobacco wrapping materials, are low in cost, are stable on prolonged storage, resemble natural constituents of tobacco, are not known to decompose on smoking to give materials which are physiologically harmful, and their flavor enhancement is deliverable through a filter. The use of the polyisoprenoid hydrocarbons of this invention and mixtures thereof as tobacco flavoring agents also permits greater flexibility in experimenting with new blends of tobacco.

The polyisoprenoid hydrocarbons of this invention may also be admixed with other polyisoprenoid materials such as polyisoprenoid esters or alcohols or mixtures of such polyisoprenoid esters and alcohols, and such mixtures of polyisoprenoid hydrocarbon and other polyisoprenoid materials have been found to be particularly convenient compositions to be used as tobacco flavoring agents. The polyisoprenoid alcohols and esters found useful in this invention may be illustrated by the formula



wherein n is an integer of from 4 to 15, inclusive, and R is an hydroxyl radical or a lower aliphatic carboxylic acid ester radical such as formate, acetate, propionate and the like, and the dotted line means that, at the positions indicated, the bond can be either a single or a double bond, one double bond being

present, and where x is an integer of 1 or 2 and y is an integer of 2 or 3, the value of x and y being determined by the position of the double bond. The data obtained from Nuclear Magnetic Resonance studies confirms the presence of vinylidene methylene groups as well as of trisubstituted double bonds. Further the esters or alcohols formed may contain a terminal menthadiene group of the formula



in place of the terminal hydrogen shown in the above formula. Accordingly, the formula given as representative of the polyisoprenoid esters or alcohols contemplates and includes all of the foregoing variations. Such polyisoprenoid alcohols include: 2, 6, 10, 14-tetramethyl- 2, 6, 10, 14-hexadecatetraene- 16- ol; 2, 6, 10, 14, 18, 22, 26-heptamethyl- 2, 6, 10, 14, 18, 22, 26-octacosaeptaene- 28 - ol; 2, 6, 10, 14, 18, 22, 26, 30, 34-nonomethyl- 2, 6, 10, 14, 18, 22, 26, 30, 34-hexatriacontanonaen- 36 - ol; 2, 6, 10, 14, 18, 22, 26, 30, 34, 38, 42, 46 -octatetracontadodecaen- 48 - ol; and the double bond isomers thereof. The polyisoprenoid esters include the lower alkyl esters of the aforesaid alcohols. It should be understood that the useful polyisoprenoid alcohols represented by the above formula may include additional hydroxy groups in the chain or may have tertiary hydroxy groups rather than terminal hydroxy groups and such materials would be considered equivalent for purposes of this invention.

Although the mixture of polyisoprenoid hydrocarbon and other polyisoprenoid material may contain a broad range of amounts by weight of polyisoprenoid hydrocarbon to other polyisoprenoid material, it is convenient that the polyisoprenoid hydrocarbon be present to a minor amount (less than 50 percent) by weight and the other polyisoprenoid materials in the mixture be present in a major amount (50 percent or more) by weight since such compositions may be directly prepared by the controlled polymerization of isoprene in a lower aliphatic carboxylic acid. More particularly, the polyisoprenoid hydrocarbon may range from about 15 to 40 percent by weight of the total polyisoprenoid mixture and the other polyisoprenoid material may range from about 85 to 60 percent by weight of the total mixture.

The tobacco flavoring agent composition of this invention may be incorporated in, or added to, tobacco products by any of the usual techniques including mechanical mixing, spraying, impregnation, coating and other methods which will readily occur to those skilled in the art. The amount of the tobacco flavoring agent composition used will depend upon a number of factors including: the particular flavor or smoking character desired, the particular tobaccos used, and the specific flavor agent composition employed. Generally the flavor agent mixture of this invention is admixed with tobacco or tobaccolike materials in amounts of from about 0.25 to 6.0 percent by weight based on the weight of the tobacco materials. At proportions below about 0.25 percent little improvement is detected and at proportions much above about 6.0 percent nontobacco or objectionable flavors or aromas may be produced.

The tobacco enhancing compositions of this invention may also be combined with other flavor or aroma enhancing materials to obtain a more esthetically pleasing combination of qualities. These can be formulated as desired to obtain the particular nuances required. Other smoke enhancing ingredients with which the compositions of this invention may be combined include: essential oils, balsam, fruit flavors such as peach, walnut and cherry, coriander, coumarin, lactic acid and phenylacetic acid. Minty, spicy, nutty, fruity, and many other nuances can be added to the compositions of this invention.

A mixture of polyisoprenoid hydrocarbons of suitable chain length may be prepared by the controlled polymerization of isoprene. In this respect ionic polymerizations have been found to be more capable of being controlled, and, therefore, are to be preferred in the practice of this invention. Further, a useful mixture of such polyisoprenoid hydrocarbons and polyisoprenoid esters may be obtained directly by the controlled polymerization of isoprene in a lower aliphatic carboxylic acid. In these processes the reaction mixture containing the desired polyisoprenoid materials of suitable chain length (C_{20} to C_{75}) are refined and separated from the nondistillable portions in such a manner that the mixture is purified under low thermal hazard conditions so that the material is not decomposed or degraded. Molecular distillation procedures have been found to be particularly effective in separating the desired polyisoprenoid materials from the undistillable objectionable portions of the reaction product components without degradation of the desired polyisoprenoid materials. For this purpose the spinning disc type of molecular still has been especially effective since it permits purification with the least thermal degradation, although, the "wiping" or "brush-type" molecular still may be used. In general, the controlled polymerization is of the ionic type, and is preferably carried out by reacting isoprene in a lower aliphatic carboxylic acid and in the presence of a strong acid catalyst, preferably having a PH of 3 or less, under controlled temperatures and for a period sufficient to obtain the desired chain length. Any lower aliphatic carboxylic acid may be used, although lower alkyl monocarboxylic acids such as acetic acid, propionic acid and the like are preferred. The strong acid catalyst used may be a mineral acid such as sulfuric acid and phosphoric acid, or acidic resins such as sulfonated polystyrene resins like "Amberlyst 15" made and sold by Rohm & Haas Company, and other strong acid equivalents.

The temperature and duration of the reaction will depend on a variety of factors including the economics of reaction time and conversions desired, and the specific organic acid and catalyst employed. Generally, the reaction temperature may vary from about 15° C. to 70° C. and temperatures of from about 25° C. to 35° C. are preferred since at lower temperatures the reaction time is slowed, while at temperatures much above 70° C. the reaction may proceed too quickly, higher polymers of isoprene may be formed, and a closed system and super atmospheric pressure is required to prevent loss of the volatile isoprene. The time of the reaction may vary from about 5 to 200 hours, and preferably the reaction period is from about 50 to 170 hours.

When the reaction is completed, the acid catalyst is neutralized by the addition of a basic material or removed from the reaction mixture, and the unreacted materials and the reaction products of chain length below about C_{20} may be separated by any conventional method. Preferably, such products are separated by distillation. The residue from this separation containing products of about C_{20} and higher chain lengths is further refined by molecular distillation, and the distillate from this molecular distillation contains a tobacco flavor agent composition comprising the polyisoprenoid hydrocarbons and polyisoprenoid esters of this invention and the composition obtained may be used as such without further treatment. Since the polyisoprenoid hydrocarbons are the preferred flavoring agents the polyisoprenoid esters formed in the reaction may be converted, if desired, to the corresponding polyisoprenoid hydrocarbons by pyrolysis or any of the conventional processes.

In still another embodiment, the said composition of polyisoprenoid hydrocarbons and polyisoprenoid esters may be saponified by reaction with a strong alkali-material to yield a mixture of polyisoprenoid hydrocarbons and polyisoprenoid alcohols, which may also contain some polyisoprenoid esters depending upon the degree of hydrolysis obtained. The saponified mixture after purification by molecular distillation is also effective as a tobacco enhancing agent.

The following example will serve only to illustrate the invention more fully; and accordingly they are not to be construed as limiting the scope of the invention.

Example 1

To a flask fitted with reflux condenser, stirrer and thermometer 3,000 gm. of isoprene, 3,000 gm. of glacial acetic acid, 15 gm. of water, 7 gm. of hydroquinone, and 10 gm. of concentrated sulfuric acid were charged. The mixture was stirred for 6 hours and then allowed to stand at about 25° C., for 120 hours, stirring for 30 minutes after each 24 hours period.

The catalyst was then neutralized by adding 50 gm. of anhydrous sodium acetate to the mixture, and the mixture was stirred until the salt was dissolved. The reflux condenser was replaced with a distillation head and the reaction mixture heated to 50° C. A fraction weighing 703 gms. was obtained. This fraction was essentially pure isoprene.

The pressure was reduced to 100 mm. Hg and the temperature of the reaction mixture held at 50° C. A fraction weighing 545 gms. was obtained. This mixture consisted of 235 gms. (43 percent) of acetic acid and 310 gms. (57 percent) of isoprene.

The pressure was reduced to 3 mm. Hg and the temperature of the reaction mixture held at 50° C. A fraction weighing 2,462 gms. was obtained. This fraction consisted of 2,157 gms. (87.6 percent) of acetic acid and 305 gms. (12.4 percent) of prenyl acetate.

The pressure was held at 3 mm. Hg and the temperature of the reaction mixture slowly raised to 200° C. The distilled fraction was condensed and collected. It weighed 942 gms. The residue weighed 256 gms.

The distilled fraction was washed once with an equal volume of water then once with an equal volume of 3 percent sodium carbonate and subjected to fractional distillation and the results obtained are summarized in the following table:

Compound type	Weight collected (gms.)	Yield percent
C ₅ compounds.....	433	14.4
C ₁₀ compounds.....	600	20.0
C ₁₅₋₂₀ compounds.....	153	5.1
Residue.....	256	8.5

The residue obtained was distilled on a spinning disc molecular distillation unit. The resulting distilled product comprised a mixture of C₂₀ to C₇₅ polyisoprenoid hydrocarbons and a mixture of C₂₀ to C₇₅ polyisoprenoid acetates, in which the ester portion of the mixture represented about 70 percent by weight.

Example 2

The crude residue product of example 1 prior to molecular distillation comprising a mixture of polyisoprenoid hydrocarbons and polyisoprenoid acetates was saponified as follows:

To 5 kg. of the residue of example 1 was added 8 liters of 20 percent potassium hydroxide in methanol. The reaction mass was then neutralized with hydrochloric acid, washed with water and extracted with petroleum ether. The ether extract was dried and flash evaporated leaving 4,176 grams of a crude mixture. The crude mixture was then molecularly distilled on a spinning disc molecular distillation unit and yielded a composition composed of about 30 percent by weight of C₂₀ to C₇₅ polyisoprenoid hydrocarbons and 70 percent by weight of C₂₀ to C₇₅ polyisoprenoid alcohols.

Example 3

The saponified product of example 2 was further refined and separated in the following manner:

The 4,176 grams of saponified product of example 2 was diluted with 6 liters of *n*-hexane, and 20 pounds of activated

silica gel was slowly added with stirring. The silica gel was filtered and washed with 10 liters of hexane. The wash and filtrate were combined and flash evaporated, yielding 1,817 grams of hydrocarbon material.

1.4 kg. of this hydrocarbon fraction was distilled on a spinning disc molecular distillation unit. 11 fractions weighing 878 grams were obtained. These fractions are listed as follows:

Hydrocarbon fraction Number	Boiling point, °C. (rotor temp.)	Pressure (mm. Hg)	Refractive index (n _D 20)	Iodine Number
1.....	84	0.032	1.5093	276.3
2.....	85	0.017	1.5096	-----
3.....	100	0.020	1.5105	270.2
4.....	106	0.018	1.5107	-----
5.....	110	0.016	1.5114	261.7
6.....	125	0.018	1.5126	-----
7.....	145	0.025	1.5148	239.1
8.....	152	0.028	1.5167	-----
9.....	140	0.020	1.5159	240.9
10.....	170	0.050	1.5188	-----
11.....	185	0.200	1.5196	223.6

The silica gel was then washed with 12 liters of methanol. The wash liquid was flash evaporated leaving 1,396 grams of crude alcohols. This material was distilled on a spinning disc molecular still. Twelve fractions weighing 916 grams were obtained.

Alcohol fraction Number	Boiling point, °C. (rotor temp.)	Pressure (mm. Hg)	Refractive index (n _D 20)	Iodine Number
1.....	75	0.031	1.4971	304.3
2.....	80	0.017	1.4979	-----
3.....	84	0.011	1.4988	304.9
4.....	84	0.011	1.4984	-----
5.....	87	0.009	1.5000	306.6
6.....	90	0.008	1.5007	-----
7.....	96	0.008	1.5021	296.7
8.....	100	0.007	1.5042	-----
9.....	110	0.007	1.5060	302.1
10.....	116	0.007	1.5074	-----
11.....	124	0.007	1.5094	294.7
12.....	125	0.007	1.5104	-----

Example 4

To a flask fitted with reflux condenser, stirrer and thermometer was added 3,000 gms. of isoprene, 3,000 gms. of propionic acid, 15 gms. of water, 7 mgs. of hydroquinone and 10 gms. of concentrated sulfuric acid. The mixture was stirred for 30 minutes at the end of each 24 hour period. The reaction was then stopped by neutralizing the catalyst by adding and dissolving 58 grams of anhydrous sodium propionate.

The reflux condenser was replaced with a distillation head and condenser and the reaction mixture heated to 50° C.

A fraction of pure isoprene weighing 702 grams was obtained. The pressure was reduced to 100 mm. Hg and the temperature of the reaction mixture maintained at 50° C. A fraction weighing 1,304 grams and consisting of 29 grams (2.2 percent) propionic acid and 1,275 grams (97.8 percent) of isoprene was obtained.

The pressure was reduced to 3 mm. Hg and the temperature of the reaction mixture maintained at 50° C. A fraction weighing 2,982 grams and consisting of 179 grams (6 percent) prenyl propionate and 2,803 grams (94 percent) of propionic acid was obtained.

The pressure was held at 3 mm. Hg and the temperature of the liquid slowly raised from 50° C. to 200° C. A fraction weighing 277 grams was collected. As residue, 192 grams were obtained.

The distilled fraction was washed once with 600 mls. of water and then once with 600 mls. of 3 percent aqueous sodium carbonate, and subjected to fractional distillation. The results obtained are summarized in the table below:

Compound type	Weight collected (gms.)	Yield, percent
C ₅ compounds.....	196	6.5
C ₁₀ compounds.....	141	4.7
C ₁₅₋₂₀ compounds.....	1.1	0.04
Residue.....	192	6.4

The residue was distilled on a spinning disc molecular distillation unit. The resulting product comprised a mixture of C₂₀ to C₇₅ polyisoprenoid hydrocarbons and a mixture of C₂₀ to C₇₅ polyisoprenoid propionates, in which the ester proportion of the mixture represented about 70 percent by weight.

The resulting product may then be saponified as in example 2 and separated into hydrocarbon and alcohol fractions in accordance with the procedure set forth in example 3.

Example 5

A series of tobacco blends were prepared by spraying a solution of each of the flavoring agents shown in the following tabulation onto a tobacco blend consisting of 55 percent weight of Type 12 Virginia tobacco and 45 percent by weight of Burley tobacco. The solution comprised 1 percent of each of said flavoring agents based on the weight of the tobacco blend and equal parts by volume of hexane and special denatured number 4 ethyl alcohol. Each of the sprayed tobacco blends was air dried, made into cigarettes and allowed to age over night. The cigarettes were then smoked and evaluated by a panel of experts and the results indicated in the tabulation were noted.

Flavoring agent employed	Evaluation comment
1. Unflavored tobacco blend.....	Standard of comparison.
2. The molecularly distilled product of Example 1 comprising a mixture of C ₂₀ to C ₇₅ polyisoprenoid hydrocarbons and a mixture of C ₂₀ to C ₇₅ polyisoprenoid acetates.	Stronger tobacco taste, no significant off taste, less harsh.
3. The composite hydrocarbon fraction of Example 3.	Strong tobacco flavor. Milder flavor. Smoother than flavoring agent 2 of this tabulation.
4. Hydrocarbon fraction 1 of Example 3.	Do.
5. Hydrocarbon fraction 3 of Example 3.	Do.
6. Hydrocarbon fraction 5 of Example 3.	Do.
7. Hydrocarbon fraction 7 of Example 3.	Do.
8. Hydrocarbon fraction 9 of Example 3.	Do.
9. Hydrocarbon fraction 11 of Example 3.	Do.
10. Alcohol fraction 1 of Example 3.	Strong flowery-chemical off taste.
11. Alcohol fraction 3 of Example 3.	Do.
12. Alcohol fraction 5 of Example 3.	Do.
13. Composite of 12 alcohol fractions of Example 3.	Some off taste. Hot suitable at concentrations greater than 1% by weight.
14. The molecularly distilled product of Example 2 comprising a mixture of C ₂₀ -C ₇₅ polyisoprenoid hydrocarbons and C ₂₀ -C ₇₅ polyisoprenoid alcohol.	Less harsh and smoother than control but not as good as flavoring agents Nos. 2 and 3 of this tabulation.

Example 6

To a 3 neck flask fitted with reflux condenser, stirrer and thermometer was added 3,000 gms. of isoprene, 3,000 gms. of glacial acetic acid, 15 gms. of water, 7 gms. of hydroquinone and 41.6 gms. of Amberlyst-15 sulfonated polystyrene ion-exchange resin. The mixture was stirred continuously for 126 hours.

50 gms. of anhydrous sodium acetate was then added to the mixture and stirred until dissolved. The reflux condenser was replaced with a distillation head and condenser and the reaction mixture heated to 50° C. A fraction pure isoprene weighing 592 gms. was obtained.

The pressure was reduced to 100 mm. Hg. and the temperature of the reaction mixture held at 50° C. A fraction weighing 672 gms. and consisting of 325 gms. (48.4 percent) of acetic acid and 347 gms. (51.6 percent) of isoprene was obtained. The reaction mixture was cooled and filtered under slight vacuum to remove the ion-exchange resin. The filtrate was distilled at a pressure of 3 mm. Hg. maintaining the reaction mixture at 50° C. A fraction weighing 2,363 gms. and consisting of 1,914 gms. (81 percent) of acetic acid and 449 gms. (19 percent) of prenylacetate was obtained. The pressure was held at 3 mm. and the temperature of the liquid slowly raised from 50° C. to 200° C. A fraction weighing 569 gms. was obtained. A residue of 55 gms. was obtained.

The residue of 55 gms. may then be distilled on a spinning disc molecular still. The resulting distilled product comprises a mixture of C₂₀ to C₇₅ polyisoprenoid hydrocarbons and a mixture of C₂₀ and C₇₅ polyisoprenoid acetates, in which the ester portion of the mixture represents about 70 percent by weight.

The distilled mixture is blended with tobacco at 1 percent by weight concentration and enhances the flavor of the blend when smoked.

Example 7

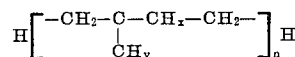
4 kg. of the residue obtained in example 1 was distilled on a spinning disc molecular distillation unit as described in example 1, yielding 3.2 kg. of distilled product comprising a mixture of C₂₀ to C₇₅ polyisoprenoid hydrocarbons and a mixture of C₂₀ to C₇₅ polyisoprenoid acetates, in which the ester portion of the mixture represented about 70 percent by weight.

This material was passed through a steel tube packed with porcelain saddles at 400° C. in a nitrogen atmosphere, yielding about 1,400 gms. of a product consisting substantially of polyisoprenoid hydrocarbons having the same chain length as the hydrocarbons of example 3. The crude pyrolysate mixture was washed with aqueous sodium bicarbonate solution to remove the acetic acid formed in the pyrolysis, dried, and distilled on a spinning disc as in example 3. There were obtained 11 fractions in a temperature range of 85° C. to 185° C. (rotor temperature) at approximately 0.03 to 0.02 mm. Hg. pressure. Of these fractions, the first three were discarded. The remainder of about 1,100 gms. were evaluated and were found to have substantially the same smoking properties as the product of example 3.

The invention in its broader aspects is not limited to the specific compositions, combinations and improvements described, but departures may be made therefrom within the scope of the accompanying claims and their equivalents without departing from the principles of the invention and without sacrificing its chief advantages.

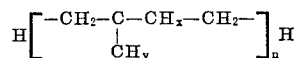
What I claim is:

1. A tobacco product having improved flavor and smoking characteristics comprising tobacco and from about 0.25 to 6.0 percent by weight based on the weight of the tobacco of a polyisoprenoid hydrocarbon represented by the formula

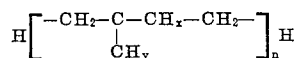


wherein n is an integer of from about 4 to 15 inclusive, and wherein each dotted line is selected from the group consisting of a single bond and a double bond, one double bond being present, and wherein x is an integer of from 1 to 2 and y is an integer of from 2 to 3, the value of x and y being determined by the position of said double bond.

2. A tobacco product comprising tobacco and from about 0.25 to 6.0 percent by weight based on the weight of tobacco of a mixture of (i) a polyisoprenoid hydrocarbon represented by the formula



where n is an integer of from about 4 to 15 inclusive and wherein each dotted line is a bond selected from the group consisting of a single bond and a double bond, a double bond being present, and wherein x is an integer of from 1 to 2 and y is an integer of from 2 to 3, the value of x and y being determined by the position of said double bond, and (ii) a polyisoprenoid compound represented by the formula



wherein n is an integer of from about 4 to 15 inclusive, and wherein each dotted line is a bond selected from the group consisting of a single bond and a double bond, a double bond being present, and wherein x is an integer of from 1 to 2 and y is an integer of from 2 to 3, the value of x and y being determined by the position of said double bond, and R is a radical

selected from the group consisting of hydroxyl and lower aliphatic carboxylic acid ester radicals.

3. A tobacco product as defined in claim 2, wherein said polyisoprenoid compound is a polyisoprenoid alcohol.

4. A smoking product as defined in claim 2, wherein said polyisoprenoid compound is a polyisoprenoid lower aliphatic carboxylic acid ester.

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UNITED STATES PATENT OFFICE
CERTIFICATE OF CORRECTION

Patent No. 3,589,372 Dated June 29, 1971

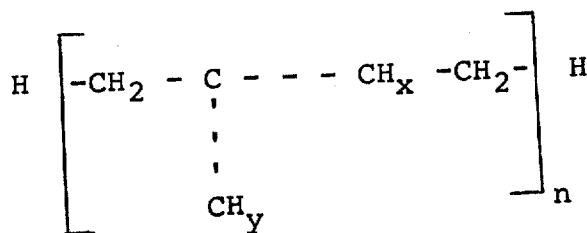
Inventor(s) ERNST T. THEIMER

It is certified that error appears in the above-identified patent and that said Letters Patent are hereby corrected as shown below:

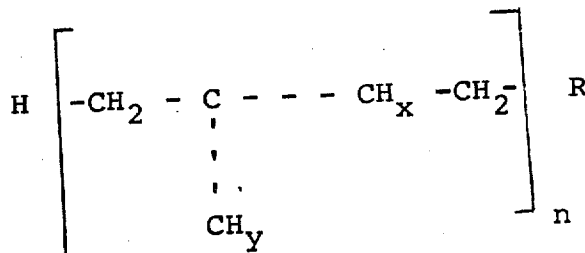
Title page, correct the title to read:

--TOBACCO COMPOSITION OR THE LIKE INCORPORATING A
POLYISO-PRENOID ALCOHOL FLAVORANT--

Title page, in the Abstract, Col. 2, line 7, Col. 8, line 45, (claim 1), and at Col. 8, line 57, (claim 2), the formula should read:



Col. 2, line 68, and at Col. 8, line 67, (claim 2), the formula should read:



Col. 2, line 35, "cosaheptaene" should read --octa-cosaheptaene--

Col. 5, line 63, "either" should read --ether--.

Col. 7, line 43, "hot" should read --not--

Col. 8, line 19, "400°C. In a" should read --400°C. in a--

Signed and sealed this 21st day of March 1972.

(SEAL)
Attest:

EDWARD M. FLETCHER, JR.
Attesting Officer

ROBERT GOTTSCHALK
Commissioner of Patents