

PATENT SPECIFICATION

(11)

S. 22687
Singapore
1 317 584

NO DRAWINGS

1 317 584

- (21) Application No. 13865/70 (22) Filed 23 March 1970
(61) Patent of Addition to No. 1 312 483 dated 19 April 1971
(23) Complete Specification filed 19 April 1971
(44) Complete Specification published 23 May 1973
(51) International Classification A24B 15/00
(52) Index at acceptance
A2C 18A3 18C1 20A 20D 20E1 20E2 20EX 20H1 20HX 20J2
(72) Inventor ROBERT CRAIG ANDERSON

(19)



9

(54) IMPROVED SMOKING MIXTURE

(71) We, IMPERIAL CHEMICAL INDUSTRIES LIMITED of Imperial Chemical House, Millbank, London S.W.1., a British Company, do hereby declare the invention, 5 for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:

This invention relates to a smoking mixture suitable for incorporation in cigars, 10 cigarettes and smoking pipes, in total or partial replacement of the tobacco constituent of the mixtures normally used.

Smoking mixtures normally consist 15 mainly of, or contain a high proportion of natural tobacco and the opinion is now widely held that the smoking of tobacco, especially in cigarette form, increases the incidence of lung cancer and bronchitic 20 ailments. The replacement of tobacco in smoking mixtures by cellulose and other smoke-producing materials has been proposed but such proposed mixtures have not been acceptable to smokers because the 25 smoke flavour was excessively different from that of tobacco smoke.

It is an object of this invention to provide a smoking mixture comprising an organic combustible material as smoke producing fuel which will give a smoke resembling tobacco smoke in flavour. 30

In our co-pending U.K. Patent Application No. 13862/70 (Serial No. 1312483) we describe how this object can be fulfilled to 35 some extent by the inclusion of protein in the smoking mixture. The protein serves to reduce or eliminate the intensity of the off-odours and off-flavours of the smoke from the substitute smoking material and 40 also imparts a tobacco-like flavour and after-taste to the smoke. Such an improved smoking mixture containing protein to mask the unpleasant character of the smoke produced therefrom is not, however, entirely 45 satisfactory since the protein itself can give

rise to off-notes or odours.

It is a further object, therefore, of this invention to improve on the protein-containing smoking mixtures of our aforesaid Application No. 13862/70 (Serial No. 50 1312483) by including an ingredient which masks the protein off-notes of the smoke from said mixtures.

According to the present invention there is provided a smoking mixture comprising 55 an organic combustible material as smoke producing fuel, protein and phenyl acetic acid and/or 2-phenyl ethanol.

The amount of protein to smoke-producing fuel is in the range 1:1 to 1:60, 60 preferably 1:8 to 1:30.

The smoking mixture of the invention may comprise a minimum of 25% by weight of the aforesaid smoke-producing fuel up to 7% by weight of protein, up to 0.02% 65 of phenyl acetic acid and/or 0.2% of 2-phenyl ethanol.

The organic combustible material may be a smoke-producing carbohydrate.

The smoke-producing carbohydrate may 70 advantageously comprise α -cellulose, cellulose derivatives, sugars, starch, alginate, pectin or natural gum.

The organic combustible material may be a modified carbohydrate. 75

The modified carbohydrate may advantageously be prepared as disclosed in our Patent No. 1,113,979 by subjecting it to a catalysed degradation process at a temperature of 100-250°C until the weight of 80 the degraded material is less than 90% of the dry weight of the original carbohydrate.

Preferred degradation catalysts include sulphuric acid, sulphamic acid and ammonium sulphamate. 85

A preferred modified carbohydrate is thermally degraded cellulose.

The modified carbohydrate may also be oxidised cellulose. The organic combustible material may be a solid condensate as de- 90

scribed in our co-pending U.K. Application No. 22270/69 (Serial No. 1298354) and produced by acid or base catalysed condensation of a compound of the formula

5 $R^1CO \cdot CH_2 \cdot CH_2 \cdot COR^2(I)$ (or a precursor thereof), wherein R^1 and R^2 , which may be the same or different, each represents a hydrogen atom, or an alkyl, hydroxyalkyl or formyl group. Said condensate may be

10 prepared from succinaldehyde, acetyl acetone or a precursor of (I) which is a compound containing a furan ring structure.

The protein should preferably be pure and it is desirable, but not essential, that any residual fat should be removed from the protein, for example, by extraction with an organic solvent. Especially valuable proteins include animal protein, for example, albumin, casein, gelatine, peptone,

20 haemoglobin or wool protein and vegetable proteins, for example, protein from maize (zein), wheat (gluten and gliadin), soyabean or groundnut. Protein acid hydrolysates and protein enzyme hydrolysates, particularly casein acid hydrolysate and casein enzyme hydrolysate, are also suitable.

The smoking mixtures of the invention will, in addition to protein and the smoke-producing constituent and phenyl acetic acid and/or 2-phenyl ethanol contain other ingredients such as are normally used in smoking mixtures to impart desired physical properties and burning characteristics. For

30 example, the mixtures may comprise glow-controlling catalysts, materials to improve ash coherence and colour, nicotine, flavourants, medicaments and humectants or film-forming binding agents. The mixtures may also be blended with tobacco, which may be in the form of comminuted tobacco or tobacco shred.

Alkali metal compounds may advantageously be used as glow-controlling catalysts and salts of ammonia, alkali metals or alkaline earth metals may be used as ash improvers.

Alkali or alkaline earth carbonates or porous fillers may be incorporated in the smoking mixtures to give an open texture and facilitate combustion.

More particularly, additional compounds may comprise:

1. Fillers—e.g. calcium carbonate, magnesium carbonate.
2. Humectants—e.g. glycerol, ethylene glycol, polyethylene glycol.
3. Film-forming agents—e.g. methyl cellulose, sodium carboxymethyl cellulose, pectins, gums.
4. Glow-controlling catalysts—e.g. potassium citrate, calcium carbonate, magnesium carbonate.
5. Ash cohesion agents—e.g. citric acid, sodium hydrogen phosphate, tobacco

extracts.

6. Solanesol, other similar unconjugated polyisoprenoids and derivatives thereof (e.g. esters, acids and hydroxyl derivatives), may be added to obtain a sweetish aroma from the burning mixture and to give a pleasant after-note in the mouth of the smoker.

The smoking mixture containing modified carbohydrate or aldol condensation product may additionally contain as a minor component one or more carbohydrate materials such as cellulose fibre, starch or sugar, to improve flavour and physical properties.

The smoking mixture is preferably prepared in a form simulating tobacco. Thus it is preferred to form the mixed ingredients into a sheet and cut or shred into the physical form required. If the smoke-producing material used is already sheeted, the smoking mixture may be prepared by merely treating the sheet with the remaining ingredients. In the usual case, where the smoke-producing material is in divided form, the ingredients may be admixed and subsequently sheeted. To facilitate sheeting, it is advantageous to incorporate a solution of a binding agent, for example, water-soluble cellulose ether, polyvinyl alcohol or a water-soluble gum, in the mixture.

The invention is further illustrated by the following Examples in which all parts and percentages are by weight.

The Examples illustrate that the desired effect can be achieved by adding phenyl acetic acid or 2-phenyl ethanol to the smoking mixture or by adding both compounds together to the mixture. The Examples also illustrate that the addition of these compounds has little positive influence on the flavour of tobacco substitute material if protein is absent. The effect is thus synergistic between the substitute compounds and protein. In each of the Examples the flavour and after-taste of the product were assessed by a panel of smokers. Furthermore, in each Example, those ingredients which are not soluble, are to be considered as being finely divided.

Example 1

2.8 parts glycerol, 0.8 part citric acid and 1.0 part potassium citrate were dissolved in 20 parts distilled water and the resultant solution added to a stirred solution of 2 parts of sodium carboxymethyl cellulose in 80 parts hot distilled water. 1.27 parts casein (fat-free) 2 parts calcium carbonate and 10.13 parts of a material made by heating α -cellulose impregnated with 0.51 part of ammonium sulphamate for 4 hours at 200°C, were added to the above solution and the slurry stirred until homogeneous (approximately 1 hour).

(11)

The slurry was cast on glass plates to form a film 0.006 in. thick which was shredded. The shred was sprayed with an ether solution containing 0.004 part of

- 5 phenyl acetic acid, the ether removed by evaporation and the shred made into cigarettes. When smoked, the cigarettes had a tobacco-like flavour and after-taste. This effect was superior to that from a similar cigarette without phenyl acetic acid added.

- 10 In a similar composition in which the casein was replaced by ammonium sulphate, thus eliminating any protein flavour effect yet keeping the smoke pH the same as before, phenyl acetic acid produced a smoke in which no tobacco flavour was evident.

Example 2

- 2.8 parts of glycerol, 0.8 part citric acid and 1.0 part potassium citrate were dissolved in 20 parts distilled water and the resultant solution added to a stirred solution of 2 parts of sodium carboxymethyl cellulose in 80 parts hot distilled water. 0.88 part casein (fat-free), 2 parts of calcium carbonate and 10.52 parts of a material made by heating α -cellulose impregnated with 0.51 part of ammonium sulphamate for 4 hours at 200°C, were added to the above solution and the slurry stirred until homogeneous (approximately 1 hour).

- 30 The slurry was cast on glass plates to form a film 0.006 in. thick which was shredded. The shred was sprayed with an ether solution containing 0.004 part of phenyl acetic acid, the ether removed by evaporation and the shred made into cigarettes. When smoked, the cigarettes had a tobacco-like flavour and after-taste. The same result was obtained when the 0.004 part of phenyl acetic acid was incorporated into the slurry mix instead of spraying on to the shred.

- 45 This Example illustrates the desired effect with a lower proportion of casein included in the smoking mixture.

Example 3

- 2.8 parts of glycerol, 0.8 part citric acid and 1.0 part potassium citrate were dissolved in 20 parts distilled water and the resultant solution added to a stirred solution of 2 parts of sodium carboxymethyl cellulose in 80 parts hot distilled water. 0.88 part casein (fat-free), 2 parts of calcium carbonate and 10.52 parts of a material made by heating α -cellulose impregnated with 0.51 part of ammonium sulphamate for 4 hours at 200°C, were added to the above solution and the slurry stirred until homogeneous (approximately 1 hour).

- 60 The slurry was cast on glass plates to form a film 0.006 in. thick which was shredded. The shred was sprayed with an ether solution containing 0.002 part of

phenyl acetic acid, the ether removed by evaporation and the shred made into cigarettes.

When smoked, the cigarettes had a tobacco-like flavour and after-taste.

This Example illustrates the desired effect being obtained with a lower proportion of phenyl acetic acid in the smoking mixture.

Example 4

2.8 parts glycerol, 0.8 part citric acid and 1.0 part potassium citrate were dissolved in 20 parts distilled water and the resultant solution added to a stirred solution of 2 parts of sodium carboxymethyl cellulose in 80 parts hot distilled water. 0.88 part casein (fat-free), 2 parts of calcium carbonate and 10.52 parts of a material made by heating α -cellulose impregnated with 0.51 part of ammonium sulphamate for 4 hours at 200°C, were added to the above solution and the slurry stirred until homogeneous (approximately 1 hour).

The slurry was cast on glass plates to form a film 0.006 in. thick which was shredded. The shred was sprayed with an ether solution containing 0.02 part 2-phenyl ethanol, the ether removed by evaporation and the shred made into cigarettes.

When smoked, the cigarettes had protein off-notes and rank notes reduced with tobacco-like after-taste and a hint of tobacco flavour present.

Example 5

2.8 parts glycerol, 0.8 part citric acid and 1.0 part potassium citrate were dissolved in 20 parts distilled water and the resultant solution added to a stirred solution of 2 parts of sodium carboxymethyl cellulose in 80 parts hot distilled water. 0.88 part casein (fat-free), 2 parts of calcium carbonate and 10.52 parts of a material made by heating α -cellulose impregnated with 0.51 part of ammonium sulphamate for 4 hours at 200°C, were added to the above solution and the slurry stirred until homogeneous (approximately 1 hour).

The slurry was cast on glass plates to form a film 0.006 in. thick which was shredded. The shred was sprayed with an ether solution containing 0.008 part of 2-phenyl ethanol, the ether removed by evaporation and the shred made into cigarettes.

When smoked, the cigarettes had a faint tobacco-like flavour and tobacco-like after-taste. Rank protein off-notes were considerably reduced compared to the same composition without 2-phenyl ethanol.

Example 6

2.8 parts glycerol, 0.8 part citric acid and 1.0 part potassium citrate were dissolved in 20 parts distilled water and the resultant solution added to a stirred solution of 2

parts of sodium carboxymethyl cellulose in 80 parts hot distilled water. 0.88 part casein (fat-free), 2 parts of calcium carbonate and 10.52 parts of a material made by heating α -cellulose impregnated with 0.51 part of ammonium sulphamate for 4 hours at 200°C, were added to the above solution and the slurry stirred until homogeneous (approximately 1 hour).

- 10 The slurry was cast on glass plates to form a film 0.006 in. thick which was shredded. The shred was sprayed with an ether solution containing 0.002 part phenyl acetic acid and 0.02 part 2-phenyl ethanol, the ether removed by evaporation and the shred made into cigarettes.

When smoked, the cigarettes had a sweet tobacco-like flavour and tobacco-like after-taste. The flavour was superior to the similar composition without 2-phenyl ethanol (Example 3) and protein off-odours were not so readily detected.

Example 7

- 25 2.8 parts glycerol, 0.8 part citric acid and 1.0 part potassium citrate were dissolved in 20 parts distilled water and the resultant solution added to a stirred solution of 2 parts of sodium carboxymethyl cellulose in 30 80 parts hot distilled water. 0.56 parts casein (fat-free), 2 parts of calcium carbonate and 10.84 parts of a material made by heating α -cellulose impregnated with 0.51 part of ammonium sulphamate for 4 hours at 200°C, were added to the above solution and the slurry stirred until homogeneous (approximately 1 hour).

- 35 The slurry was cast on glass plates to form a film 0.006 in. thick which was shredded. The shred was sprayed with an ether solution containing 0.001 part of phenyl acetic acid and 0.01 part 2-phenyl ethanol, the ether removed by evaporation and the shred made into cigarettes.

- 45 When smoked, the cigarettes had weak tobacco-like flavour and tobacco-like after-taste.

Example 8

- 1.34 parts of glycerol and 0.78 part of 50 potassium citrate dissolved in 4 parts distilled water were mixed with 1.92 parts of sodium carboxymethyl cellulose dissolved in 96 parts hot distilled water. 6.72 parts of magnesium carbonate (magnesite), 3.86 55 parts of calcium carbonate, 0.2 part of protein casein (extracted for 48 hours with petroleum ether at 30°C) and 5.18 parts of a material made by heating α -cellulose with ammonium sulphamate for up to 4 hours at 200°C, were added to the above mixture and the whole stirred until homogeneous (about 1 hour).

- 65 The slurry was cast on glass plates to give a film 0.006 in. thick which was dried, removed from the plates, cut and shredded.

This shred was sprayed with a solution of 0.0002 part of phenyl acetic acid and 0.0004 part of 2-phenyl ethanol in 50 parts of ether and the solvent allowed to evaporate.

When the treated shred was made into 70 cigarettes in a 50:50 blend with flue-cured tobacco shred and smoked in comparison with a similar blend with the two additives missing, there was a marked preference for the former cigarette because of the presence 75 of a fuller flue-cured tobacco flavour.

Assimilar result was obtained in similar exercises using either 70:30 or 85:15 shred/shred blends of the modified cellulose mixture and flue-cured tobacco respectively. 80

Example 9

1.34 parts of glycerol and 0.78 part of potassium citrate dissolved in 4 parts distilled water were mixed with 1.92 parts of 85 sodium carboxymethyl cellulose dissolved in 96 parts hot distilled water. 6.72 parts of magnesium carbonate (magnesite), 3.86 parts of calcium carbonate, 0.2 part of protein casein (extracted for 48 hours with petroleum ether at 30°C) and 5.18 parts of a material made by heating α -cellulose with ammonium sulphamate for up to 4 hours at 200°C, were added to the above mixture and the whole stirred until homogeneous 95 (about 1 hour).

The slurry was cast on glass plates to give a film 0.006 in. thick which was dried, removed from the plates, cut and shredded. This shred was sprayed with a solution of 100 0.0002 part of phenyl acetic acid, 0.0004 part 2-phenyl ethanol and 0.01 part of carotene (as a mixture of isomers 85% β -, 15% α -carotene) in 35 parts of carbon tetrachloride and the solvent allowed to 105 evaporate.

When the treated shred was made up into cigarettes in a 50:50 blend with flue-cured tobacco shred and smoked in comparison with a similar blend with phenyl acetic acid 110 and 2-phenyl ethanol missing, there was a marked preference for the former cigarette because of the presence of a fuller flue-cured tobacco flavour.

A similar result was obtained in similar 115 exercises using a 70:30 shred/shred blend of modified cellulose mixture and flue-cured tobacco respectively.

Example 10

1.34 parts of glycerol and 0.78 part of potassium citrate dissolved in 4 parts of distilled water were mixed with 1.92 parts of sodium carboxymethyl cellulose dissolved in 96 parts hot distilled water. 6.72 parts 125 of magnesium carbonate (magnesite), 3.86 parts of calcium carbonate, 0.38 part of the protein casein (extracted for 48 hours with petroleum ether at 30°C) and 5.0 parts of rice starch were added to the above mixture 130

and the whole stirred until homogeneous (about 1 hour).

The slurry was cast on glass plates to give a film 0.006 in. thick which was dried, removed from the plates, cut and shredded. The shred was sprayed with a solution of 0.0002 part of phenyl acetic acid and 0.002 part of 2-phenyl ethanol in 50 parts ether and the solvent allowed to evaporate. When the treated shred was made into cigarettes and smoked in comparison with a similar cigarette not containing the two sprayed-on additives, there was a marked preference for the cigarette with the additives because of its improved tobacco character.

Example 11

1.34 parts of glycerol and 0.78 part of potassium citrate dissolved in 4 parts of distilled water were mixed with 1.92 parts of sodium carboxymethyl cellulose dissolved in 96 parts hot distilled water. 6.72 parts of magnesium carbonate (magnesite), 3.86 parts of calcium carbonate, 0.38 part of the protein casein (extracted for 48 hours with petroleum ether at 30°C) and 5.0 parts of α -cellulose were added to the above mixture and the whole stirred until homogeneous (about 1 hour).

The slurry was cast on glass plates to give a film 0.006 in. thick which was dried, removed from the plates, cut and shredded. The shred was sprayed with a solution of 0.0002 part of phenyl acetic acid and 0.002 part of 2-phenyl ethanol in 50 parts ether and the solvent allowed to evaporate. When the treated shred was made into cigarettes and smoked in comparison with a similar cigarette not containing the two sprayed-on additives, there was a marked preference for the cigarette with the additives because of its improved tobacco character.

Example 12

1.8 parts of glycerol dissolved in 4 parts of distilled water were mixed with 2.4 parts sodium carboxymethyl cellulose dissolved in 96 parts distilled water. 5.98 parts of magnesium carbonate (magnesite), 3.44 parts of calcium carbonate, 0.3 parts of zein protein, 1 part bentonite and 5.08 parts of a material made by heating α -cellulose with ammonium sulphamate were added to the above mixture and the whole stirred until homogeneous (about 1 hour).

The slurry was cast on glass plates and dried to give a film 0.006 in. thick. The film was removed, shredded and the shred was sprayed with a solution of 0.001 part of phenyl acetic acid and 0.002 part of 2-phenyl ethanol, in 50 parts ether and the solvent allowed to evaporate. When the treated shred was made into cigarettes and smoked, in comparison with a similar cigarette not containing the two sprayed-on

additives, there was a marked preference for the cigarettes with the additive because of improved tobacco character.

Example 13

1.8 parts of glycerol dissolved in 4 parts of distilled water were mixed with 2.4 parts sodium carboxymethyl cellulose dissolved in 96 parts distilled water. 5.98 parts of magnesium carbonate (magnesite), 3.44 parts of calcium carbonate, 2.69 parts of casein protein and 1 part of bentonite and 2.69 parts of a material made by heating α -cellulose with ammonium sulphamate were added to the above mixture and the whole stirred until homogeneous (about 1 hour).

The slurry was cast on glass plates and dried to give a film 0.006 in. thick. The film was removed shredded and the shred was sprayed with a solution of 0.001 part of phenyl acetic acid and 0.002 part of 2-phenyl ethanol in 50 parts ether and the solvent allowed to evaporate. When the treated shred was made into cigarettes and smoked in comparison with a similar cigarette not containing the two sprayed-on additives, there was a marked preference for the cigarettes with the additive because of improved tobacco character.

Example 14

1.8 parts of glycerol dissolved in 4 parts of distilled water were mixed with 2.4 parts sodium carboxymethyl cellulose dissolved in 96 parts distilled water. 5.98 parts of magnesium carbonate (magnesite), 3.44 parts of calcium carbonate, 0.088 parts of casein protein, 1 part bentonite and 5.21 parts of a material made by heating α -cellulose with ammonium sulphamate were added to the above mixture and the whole stirred until homogeneous (about 1 hour).

The slurry was cast on glass plates and dried to give a film 0.006 in. thick. The film was removed, shredded and the shred was sprayed with a solution of 0.001 part of phenyl acetic acid and 0.002 part of 2-phenyl ethanol in 50 part ether and the solvent allowed to evaporate. When the treated shred was made into cigarettes and smoked in comparison with a similar cigarette not containing the two sprayed-on additives, there was a marked preference for the cigarettes with the additive because of improved tobacco character.

Example 15

1.8 parts of glycerol dissolved in 4 parts of distilled water were mixed with 2.4 parts sodium carboxymethyl cellulose dissolved in 96 parts distilled water. 5.08 parts of magnesium carbonate (magnesite), 2.92 parts of calcium carbonate, 1.4 parts of casein protein and 1 part bentonite and

5.38 parts of a material made by heating α -cellulose with ammonium sulphamate were added to the above mixture and the whole stirred until homogeneous (about 1 hour).

- 5 The slurry was cast on glass plates and dried to give a film 0.006 in. thick. The film was removed, shredded and the shred was sprayed with a solution of 0.004 part of phenyl acetic acid and 0.04 part of 2-phenyl ethanol in 50 parts ether and the solvent allowed to evaporate. When the treated shred was made into cigarettes and smoked in comparison with a similar cigarette not containing the two sprayed-on additives, there was a marked preference for the cigarettes with the additive because of improved tobacco character.

Example 16

- 20 1.8 parts of glycerol dissolved in 4 parts of distilled water were mixed with 2.4 parts sodium carboxymethyl cellulose dissolved in 96 parts distilled water. 5.98 parts of magnesium carbonate (magnesite), 3.44 parts of calcium carbonate, 0.3 parts casein and 1 part bentonite and 5.08 parts of a condensate of succinaldehyde (see Patent Application No. 22270/69) (Serial No. 1298354) were added to the above mixture and the whole stirred until homogeneous (about 1 hour).

- 35 The slurry was cast on glass plates and dried to give a film 0.006 in. thick. The film was removed, shredded and the shred was sprayed with a solution of 0.001 part of phenyl acetic acid and 0.002 part of 2-phenyl ethanol in 50 parts ether and the solvent allowed to evaporate. When the treated shred was made into cigarettes and smoked in comparison with a similar cigarette not containing the two sprayed-on additives, there was a marked preference for the cigarettes with the additive because of improved tobacco character.

Example 17

- 50 1.8 parts of glycerol dissolved in 4 parts of distilled water were mixed with 2.4 parts sodium carboxymethyl cellulose dissolved in 96 parts distilled water. 5.98 parts of magnesium carbonate (magnesite), 3.44 parts of calcium carbonate, 0.3 parts of soya protein, 1 part bentonite and 5.08 parts of a material made by heating α -cellulose with ammonium sulphamate were added to the above mixture and the whole stirred until homogeneous (about 1 hour).

- 60 The slurry was cast on glass plates and dried to give a film 0.006 in. thick. The film was removed, shredded and the shred was sprayed with a solution of 0.001 part of phenyl acetic acid and 0.002 part of 2-phenyl ethanol in 50 parts ether and the solvent allowed to evaporate. When the treated shred was made into cigarettes and

smoked in comparison with a similar cigarette not containing the two sprayed-on additives, there was a marked preference for the cigarettes with the additive because of improved tobacco character.

Example 18

0.34 parts glycerol were dissolved in 4 parts distilled water and mixed with 6.6 parts sodium carboxymethyl cellulose dissolved in 96 parts water. 6.2 parts dolomite (Dolodust—Registered Trade Mark) 5.6 parts diatomaceous earth (celite), 0.22 part charcoal, 0.34 part octanol and 0.4 part casein protein were added to the above mixture and the whole stirred until homogeneous (about 1 hour).

The slurry was cast on glass plates and dried to give a film 0.006 in. thick. The film was removed, shredded and sprayed with a solution of 0.0004 parts phenyl acetic acid and 0.0008 parts 2-phenyl ethanol in 50 parts ether and the solvent allowed to evaporate. When the treated shred was made into cigarettes and smoked in comparison with similar cigarettes not containing the two sprayed on additives there was a marked preference for the cigarettes with the two sprayed on additives because of improved taste and after-taste which had a cigar/pipe tobacco nature.

We are aware of the Customs and Excise Act 1952, and we make no claim to the use of the invention in contravention of the law.

WHAT WE CLAIM IS:

1. A smoking mixture comprising an organic combustible material as smoke-producing fuel, protein and phenyl acetic acid and/or 2-phenyl ethanol.

2. A smoking mixture as claimed in Claim 1 wherein the amount of protein to smoke-producing fuel is in the range 1:1 to 1:60.

3. A smoking mixture as claimed in Claim 1 or Claim 2 wherein the amount of protein to smoke-producing fuel is in the range 1:8 to 1:30.

4. A smoking mixture as claimed in any one of the preceding claims comprising a minimum of 25% by weight of the aforesaid smoke-producing fuel, up to 7% by weight of protein, up to 0.02% of phenyl acetic acid and/or 0.2% of 2-phenyl ethanol.

5. A smoking mixture as claimed in any one of the preceding claims wherein the organic combustible material is a smoke-producing carbohydrate.

6. A smoking mixture as claimed in Claim 5 wherein the smoke-producing carbohydrate comprises α -cellulose, cellulose derivatives, sugars, starch, alginate, pectin or natural gum.

5.38 parts of a material made by heating α -cellulose with ammonium sulphamate were added to the above mixture and the whole stirred until homogeneous (about 1 hour).

- 5 The slurry was cast on glass plates and dried to give a film 0.006 in. thick. The film was removed, shredded and the shred was sprayed with a solution of 0.004 part of phenyl acetic acid and 0.04 part of 2-phenyl ethanol in 50 parts ether and the solvent allowed to evaporate. When the treated shred was made into cigarettes and smoked in comparison with a similar cigarette not containing the two sprayed-on additives, there was a marked preference for the cigarettes with the additive because of improved tobacco character.

Example 16

- 20 1.8 parts of glycerol dissolved in 4 parts of distilled water were mixed with 2.4 parts sodium carboxymethyl cellulose dissolved in 96 parts distilled water. 5.98 parts of magnesium carbonate (magnesite), 3.44 parts of calcium carbonate, 0.3 parts casein and 1 part bentonite and 5.08 parts of a condensate of succinaldehyde (see Patent Application No. 22270/69) (Serial No. 1298354) were added to the above mixture and the whole stirred until homogeneous (about 1 hour).

- 35 The slurry was cast on glass plates and dried to give a film 0.006 in. thick. The film was removed, shredded and the shred was sprayed with a solution of 0.001 part of phenyl acetic acid and 0.002 part of 2-phenyl ethanol in 50 parts ether and the solvent allowed to evaporate. When the treated shred was made into cigarettes and smoked in comparison with a similar cigarette not containing the two sprayed-on additives, there was a marked preference for the cigarettes with the additive because of improved tobacco character.

Example 17

- 50 1.8 parts of glycerol dissolved in 4 parts of distilled water were mixed with 2.4 parts sodium carboxymethyl cellulose dissolved in 96 parts distilled water. 5.98 parts of magnesium carbonate (magnesite), 3.44 parts of calcium carbonate, 0.3 parts of soya protein, 1 part bentonite and 5.08 parts of a material made by heating α -cellulose with ammonium sulphamate were added to the above mixture and the whole stirred until homogeneous (about 1 hour).

- 60 The slurry was cast on glass plates and dried to give a film 0.006 in. thick. The film was removed, shredded and the shred was sprayed with a solution of 0.001 part of phenyl acetic acid and 0.002 part of 2-phenyl ethanol in 50 parts ether and the solvent allowed to evaporate. When the treated shred was made into cigarettes and

smoked in comparison with a similar cigarette not containing the two sprayed-on additives, there was a marked preference for the cigarettes with the additive because of improved tobacco character.

Example 18

0.34 parts glycerol were dissolved in 4 parts distilled water and mixed with 6.6 parts sodium carboxymethyl cellulose dissolved in 96 parts water. 6.2 parts dolomite (Dolodust—Registered Trade Mark) 5.6 parts diatomaceous earth (celite), 0.22 part charcoal, 0.34 part octanol and 0.4 part casein protein were added to the above mixture and the whole stirred until homogeneous (about 1 hour).

The slurry was cast on glass plates and dried to give a film 0.006 in. thick. The film was removed, shredded and sprayed with a solution of 0.0004 parts phenyl acetic acid and 0.0008 parts 2-phenyl ethanol in 50 parts ether and the solvent allowed to evaporate. When the treated shred was made into cigarettes and smoked in comparison with similar cigarettes not containing the two sprayed on additives there was a marked preference for the cigarettes with the two sprayed on additives because of improved taste and after-taste which had a cigar/pipe tobacco nature.

We are aware of the Customs and Excise Act 1952, and we make no claim to the use of the invention in contravention of the law.

WHAT WE CLAIM IS:

1. A smoking mixture comprising an organic combustible material as smoke-producing fuel, protein and phenyl acetic acid and/or 2-phenyl ethanol.

2. A smoking mixture as claimed in Claim 1 wherein the amount of protein to smoke-producing fuel is in the range 1:1 to 1:60.

3. A smoking mixture as claimed in Claim 1 or Claim 2 wherein the amount of protein to smoke-producing fuel is in the range 1:8 to 1:30.

4. A smoking mixture as claimed in any one of the preceding claims comprising a minimum of 25% by weight of the afore-said smoke-producing fuel, up to 7% by weight of protein, up to 0.02% of phenyl acetic acid and/or 0.2% of 2-phenyl ethanol.

5. A smoking mixture as claimed in any one of the preceding claims wherein the organic combustible material is a smoke-producing carbohydrate.

6. A smoking mixture as claimed in Claim 5 wherein the smoke-producing carbohydrate comprises α -cellulose, cellulose derivatives, sugars, starch, alginate, pectin or natural gum.



* C L M S *



7. A smoking mixture as claimed in any one of Claims 1 to 4 wherein the organic combustible material is a modified carbohydrate.
- 5 8. A smoking mixture as claimed in Claim 7 wherein the modified carbohydrate is oxidised cellulose.
- 10 9. A smoking mixture as claimed in any one of Claims 1 to 4 wherein the organic combustible material is a thermally degraded carbohydrate material of U.K. Patent No. 1,113,979 which is prepared by subjecting carbohydrate to a catalysed degradation process at a temperature of 100-15 250°C until the weight of the degraded material is less than 90% of the dry weight of the original carbohydrate.
- 20 10. A smoking mixture as claimed in Claim 9 wherein the modified carbohydrate comprises thermally degraded cellulose.
- 25 11. A smoking mixture as claimed in any one of claims 1 to 4 wherein the organic combustible material is a solid condensate produced by acid or base catalysed condensation of a compound of the formula $R^1COCH_2CH_2COR^2$ (I) (or a precursor thereof) wherein R^1 and R^2 which may be the same or different, each represents a hydrogen atom, or an alkyl, 30 hydroxyalkyl or formyl group.
- 35 12. A smoking mixture as claimed in Claim 11 wherein the organic combustible material comprises a solid condensate from succinaldehyde, acetyl acetone or a solid condensate from a precursor of (I) which is a compound containing a furan ring structure.
13. A smoking mixture as claimed in any one of the preceding claims comprising pure protein free of residual fat.
- 40 14. A smoking mixture as claimed in any one of the preceding claims comprising animal protein selected from albumin, casein, gelatine, peptone, haemoglobin or wool protein or vegetable protein selected 45 from protein from maize (zein), wheat (glutin and gliadin), soyabean or ground nut.
- 50 15. A smoking mixture as claimed in any one of the preceding claims comprising protein acid hydrolysate or protein enzyme hydrolysate.
- 55 16. A smoking mixture as claimed in Claim 15 comprising casein acid hydrolysate or casein enzyme hydrolysate.
- 60 17. A smoking mixture as claimed in any one of the preceding claims containing other ingredients such as are normally used in smoking mixtures to impart desired physical properties and burning characteristics.
- 65 18. A smoking mixture as claimed in Claim 17 comprising glow-controlling catalysts, materials to improve ash coherence and colour, nicotine, flavourants, medicaments and humectants or film-forming binding agents.
- 70 19. A smoking mixture as claimed in any one of the preceding claims blended with tobacco.
20. A smoking mixture according to claim 1 substantially as hereinbefore described with reference to the Examples.

ALEXANDER McPHERSON,

Agent for the Applicants.



* C L M S *