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PATENT SPECIFICATION

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Inventors: JAMES FORRESTER MORMAN and JAMES BROWN PARKER

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COMPLETE SPECIFICATION

Modified Carbohydrate Material for Smoking Mixtures

We IMPERIAL CHEMICAL INDUSTRIES LIMITED of Imperial Chemical House, Millbank, London S.W.1., a British Company do hereby declare the invention, 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 modified carbohydrate material suitable as an ingredient of a smoking mixture for cigarettes, cigars, pipe and the like and to the preparation of such material. The invention also includes smoking mixtures comprising said carbohydrate material.

Smoking mixtures normally consist 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. This has been attributed to the presence in tobacco smoke of carcinogenic compounds such as 3,4-benzpyrene, but experiments in which tobacco smoke 'tar' has been painted on mice suggest that the 'tar' contains other harmful constituents. It is considered that reducing the 'tar' content correspondingly reduces the health hazard from smoking.

It has hitherto been proposed to replace at least in part the tobacco in smoking mixtures by woodpulp cellulose to obtain mixtures giving smoke containing less carcinogenic material. Such mixtures have not, however, been widely acceptable to smokers because of their objectionable aroma and taste.

It is an object of this invention to provide an improved carbohydrate ingredient which may be used as a total or partial replacement for tobacco in smoking mixtures. A further

object is to provide a smoking mixture which produces a smoke containing a lower content of harmful constituents than the smoke from tobacco.

The modified carbohydrate material of the invention is provided by subjecting carbohydrate to a catalysed degradation process at a temperature of 100—250° C until the weight of the degraded material is less than 90% of the weight of the original carbohydrate. Preferably the degradation treatment is continued until the weight of the modified carbohydrate is between 50 and 75% of the weight of the original carbohydrate.

The process may be catalysed by any of several reagents which accelerate the thermal degradation or lower the thermal degradation temperature of carbohydrates. Preferred catalysts include strong mineral acids and salts of such strong acids with weak bases. These catalysts include, for example, sulphamic acid, ammonium sulphamate, phosphoric acid, diammonium hydrogen phosphate, ammonium dihydrogen phosphate, sulphuric acid, ammonium sulphate, hydrochloric acid and ferric chloride. An alkali hydroxide, such as sodium hydroxide, or an alkali salt of a weak acid, such as sodium carbonate, may also be used as the degradation catalyst provided the degraded carbohydrate is washed substantially free of these catalysts before it is compounded in a smoking mixture. Preferably the catalyst is present in an amount ranging from 1 to 20% by weight of the carbohydrate.

Especially useful carbohydrate materials include, for example, α -cellulose, cellulose derivatives such as methyl cellulose, mono- and polysaccharides such as glucose and sucrose, starches such as rice, potato and maize

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starch, alginates, pectin, natural gums such as gum tragacanth, gum arabic and locust bean gum.

Smoking mixtures comprising the heat-treated carbohydrate material of the invention are superior in smoke quality and taste to the corresponding smoking materials made from untreated carbohydrates and they give a smoother, less irritating smoke. The smoke contains less 'tar' than tobacco smoke. The smoking mixtures may also be varied more widely in composition, and consequently in smoke properties, than mixtures containing untreated carbohydrates, and even than mixtures containing only tobacco. For example, by the incorporation of tasting and flavouring additives, the taste and flavour of the smoke can be varied to suit a wider range of consumer preferences. The proportion of nicotine, which is a desirable constituent of smoking mixtures, may also be adjusted widely as desired.

In preparing the carbohydrate material of the invention carbohydrate may be treated in any convenient form such as powder or sheet form. When the carbohydrate is in sheet form, it may be conveniently impregnated with a solution of the catalyst applied by immersion or spraying, and dried. Powdered carbohydrate may be mixed with a solution of the catalyst, and dried. The mixture of carbohydrate and catalyst is preferably heat-treated at 100 to 250° C for periods ranging from 1 minute to 4 hours, depending on the treatment temperature, the concentration of the catalyst and the weight loss desired.

The catalysed degradation process changes the colour of the carbohydrate to black which may be undesirable in cigarette filling formulations. The colour may be altered to a brown colour resembling tobacco cigarette filling by treatment of the modified carbohydrate with nitric acid, hydrogen peroxide or ammonia, preferably at elevated temperature.

It will be understood that the smoking mixtures of the invention may, in addition to this carbohydrate material, comprise other materials which are normal constituents of smoking mixtures, such as, for example, tobacco, untreated carbohydrate or other smoke-producing organic material and, as desired, any of the other modifying agents commonly used in such mixtures. For example, the mixtures may comprise glow-promoting catalysts, materials to improve ash coherence and colour, nicotine, flavourants or medicaments.

As glow-promoting catalysts for ensuring "fire-holding capacity", alkali metal compounds are preferred.

As materials to improve the ash, salts of ammonia, alkali metals or alkaline earth metals can be used and of these, salts of magnesium, calcium or ammonium are preferred.

Numerous beneficial flavouring materials

may be included in the mixture. These include tobacco extracts, organic esters, essential oils, menthol, tonka bean or vanillin.

Glycerol and glycols, such as, for example, ethylene glycol and di-, tri- and tetra-ethylene glycol are convenient humectants.

Other materials, such as carbonates or porous inert fillers, may be included in the smoking mixture to facilitate combustion, imparting a more open texture to the mixture, thereby facilitating access of oxygen.

It is convenient for smoking mixtures to simulate tobacco and the carbohydrate material is therefore preferably prepared in sheet form and, when required as a cigarette or pipe filling, shredded into strips. To prepare the sheeted smoking mixture, the carbohydrate, if already in sheet form, may be merely treated with the desired additives and shredded. Carbohydrate in other forms or heat-treated carbohydrate sheet which is too weak to shred properly may be comminuted to powder, mixed with a solution of film-forming agent such as, for example, a solution of a water-soluble cellulose derivative, polyvinyl alcohol, starch, pectin, gum or mucilage, formed as a film and dried. Water-soluble methyl cellulose or sodium carboxymethyl cellulose, preferably in grades such that a 2% aqueous solution has a viscosity of at least 1500 centipoises at 25° C, may advantageously be used as the film-forming agent. When an acid catalyst, such as sulphuric acid, has been used, it is advantageous to neutralise any residual acidity in the degraded carbohydrate by adding the requisite quantity of ammonia at this stage. This step leads to an improvement in the smoke quality of the final product. This improvement is also obtained when ammonia is used to improve the colour as previously described. The remaining constituents of the smoking mixture may be incorporated with the carbohydrate material into the film-forming agent. Soluble additives may, if desired, be sprayed on to the sheeted smoking mixture.

For use in cigarettes or pipes the shred mixture is preferably conditioned in a humid atmosphere to a moisture content of 5 to 15% by weight.

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

EXAMPLE 1

1.0 part of α -cellulose paper was immersed in a 6.0% aqueous solution of ammonium sulphamate and the paper compressed between blotting pads to leave 1.0 part of solution in the paper. The paper was air-dried at 55° C and then heated at 200° C for 4 hours until the weight of the degraded material was 57.6% of the original weight of α -cellulose and ammonium sulphamate. During the treatment the colour of the paper was changed

to black. The paper was allowed to cool and then sprayed with an aqueous solution containing 2% potassium carbonate and 4% glycerol and again compressed between blotting pads to leave 1.0 part of the solution in the paper. The paper was air-dried at room temperature. 1.0 part of a 6% methylene chloride extract of tobacco, to act as flavourant, was sprayed on to the sheet. The sheet was shredded to simulate the form of a cigarette filling which was then conditioned at 86% relative humidity at 25° C to give a moisture content of approximately 7%. The shred was made into cigarettes using standard cigarette paper wrappings. The average weight of the cigarettes was approximately 1.0 gram.

The sidestream and mainstream smokes produced from the cigarettes were milder and less irritating to the eyes and throat than those from an ordinary tobacco cigarette.

EXAMPLE 2

1.0 part of the shredded cigarette filling, as prepared in Example 1, was admixed with 1.0 part of cigarette tobacco shred and the mixture made into cigarettes. The average weight of the cigarettes was approximately 1.0 gram.

The sidestream and mainstream smokes produced from the cigarettes were very mild and less irritating to the eyes and throat than those of an ordinary tobacco cigarette.

EXAMPLE 3

Shredded cigarette filling was prepared as described in Example 1 except that a 2.5% aqueous solution of ammonium sulphamate was used instead of the 6.0% solution described in Example 1 and the impregnated paper was heated at 246° C for 2 minutes instead of 200° C for 4 hours. The weight of the degraded material was 58.6% of the original weight of α -cellulose and ammonium sulphamate. The shredded filling was made into cigarettes of average weight approximately 1.0 gram.

The sidestream and mainstream smokes were very similar to those described in Example 1.

EXAMPLE 4

Shredded cigarette filling was prepared as described in Example 1 except that a 5.0% aqueous solution of sulphamic acid was used instead of the aqueous solution of ammonium sulphamate. The weight of the degraded material was 62% of the original weight of the α -cellulose and sulphamic acid. The filling was made into cigarettes of average weight approximately 1.0 gram.

The sidestream and mainstream smokes produced from the cigarettes were very mild and less irritating to the eyes and throat than those of an ordinary tobacco cigarette.

EXAMPLE 5

Shredded cigarette filling was prepared as described in Example 1 except that an 8.6% aqueous solution of ammonium sulphate was used instead of the aqueous solution of ammonium sulphamate and the heat treatment carried out at 175° C instead of 200° C. The weight of the degraded material was 68% of the original weight of α -cellulose and ammonium sulphate. The filling was made into cigarettes of average weight approximately 1.0 gram.

The sidestream and mainstream smokes were very similar to those described in Example 1.

EXAMPLE 6

1.5 parts of cellulose (sulphite process wood-pulp containing approximately 92% α -cellulose) slurried in 100 parts of water were beaten in a conventional manner. 0.38 parts of calcium carbonate and 400 parts of water were then added and the slurried material formed into a sheet of paper in a conventional manner and dried between blotting pads at room temperature. The sheet, which contained approximately 10% by weight calcium carbonate, was immersed in a 5.6% aqueous solution of ammonium sulphamate and compressed between blotting pads to leave 1.5 parts of solution in the sheet. The sheet was then dried at room temperature, heated at 200° C for 2 hours to give a weight of degraded material which was 61% of the original weight of the cellulose, calcium carbonate and ammonium sulphamate. The heat-treated material was further processed as described in Example 1 except that a 10% aqueous solution of glycerol was used instead of 4%. The resulting shredded filling was made into cigarettes of average weight approximately 1.0 gram.

The sidestream and mainstream smokes produced from the cigarettes were milder and less irritating to the eyes and throat than those from an ordinary tobacco cigarette.

EXAMPLE 7

Shredded cigarette filling was prepared as described in Example 6 except that the sheet contained 12.8% magnesium carbonate instead of calcium carbonate and also 0.008 parts of cationic starch added to the slurry in order to improve retention of the magnesium carbonate. The weight of degraded material after heat treatment was 63% of the original weight of cellulose, magnesium carbonate and ammonium sulphamate.

The sidestream and mainstream smokes from cigarettes prepared from the shredded filling were much milder and less irritating to the eyes and throat than those of an ordinary tobacco cigarette.

EXAMPLE 8

Shredded cigarette filling was prepared as

described in Example 1 except that a 5% aqueous solution of ammonium dihydrogen phosphate was used instead of aqueous ammonium sulphamate solution and 10% glycerol was used instead of 4%. In this example the weight of the degraded material was 75% of the original weight of α -cellulose and ammonium dihydrogen phosphate.

The sidestream and mainstream smokes from cigarettes prepared from the shredded filling were milder and less irritating to the eyes and throat than those of an ordinary tobacco cigarette.

EXAMPLE 9

Shredded cigarette filling was prepared as described in Example 8 except that a 5% aqueous solution of diammonium hydrogen phosphate was used instead of aqueous ammonium dihydrogen phosphate solution. The weight of the degraded material was 74% of the original weight of α -cellulose and diammonium hydrogen phosphate.

The sidestream and mainstream smokes from cigarettes prepared using the cigarette filling were milder and less irritating to the eyes and throat than those from an ordinary tobacco cigarette.

EXAMPLE 10

Shredded cigarette filling was prepared as described in Example 3 except that a 5.6% aqueous solution of sulphuric acid was used instead of an aqueous ammonium sulphamate solution. The weight of the degraded material was 53% of the original weight of α -cellulose and sulphuric acid. The shredded filling was made into cigarettes of average weight approximately 1.0 gram.

The sidestream and mainstream smokes were very similar to those described in Example 1.

EXAMPLE 11

Shredded cigarette filling was prepared as described in Example 3 except that a 6.5% aqueous solution of phosphoric acid was used instead of an aqueous ammonium sulphamate solution. In this example heat treatment was carried out at 225° C for 4 minutes instead of 246° C for 2 minutes. The weight of the degraded material was 73% of the original weight of α -cellulose and phosphoric acid. The shredded filling was made up into cigarettes of average weight approximately 1.0 gram.

The sidestream and mainstream smokes were much less acrid and less irritating to the eyes and throat than those of an ordinary tobacco cigarette.

EXAMPLE 12

1.0 part of sheeted α -cellulose pulp was immersed in a 10.0% aqueous solution of sulphuric acid and the pulp compressed between rolls to leave 1.0 part of solution in the pulp. The pulp was air-dried at 45° C and then heated at 225° C for 4 minutes which made the pulp black. The weight of the

degraded material was 75% of the original weight of α -cellulose and sulphuric acid. The black pulp sheet was allowed to cool and then ground to a powder passing a 120 B.S.S. sieve. 11.4 parts of the black powder were intimately mixed with 2.0 parts of calcium carbonate and added to 100 parts of a stirred aqueous solution containing 2.0 parts of sodium carboxymethyl cellulose, 2.8 parts of glycerol, 0.8 parts of citric acid and 1.0 part of potassium citrate. The resulting slurry was spread to give a film 0.015 inches thick on a moving endless steel band conveyor and dried at 130° C. The dried film was sprayed with water to give approximately 25% moisture content in the film, removed from the steel band and conditioned at 70% relative humidity at 20° C to give a moisture content of approximately 13%. The film was shredded to simulate the form of cigarette filling and the shred made into cigarettes using standard cigarette paper wrappings. The average weight of the cigarettes was approximately 1.0 gram.

The sidestream and mainstream smokes produced from the cigarettes were milder and less irritating to the eyes and throat than those from an ordinary tobacco cigarette.

EXAMPLE 13

1.0 part of sheeted α -cellulose pulp was immersed in a 10.0% aqueous solution of sulphuric acid and the pulp compressed between rolls to leave 1.0 part of solution in the pulp. The pulp was air-dried at 45° C and then heat-treated at 225° C for 4 minutes which made the pulp black. The weight of the degraded material was 75% of the original weight of α -cellulose and sulphuric acid. The black pulp sheet was allowed to cool and then ground to a powder passing a 120 B.S.S. sieve. 11.4 parts of the black powder were added to 30 parts of water and the resulting slurry was neutralised with 1.6 ml. of 0.88N aqueous ammonium hydroxide solution. This black slurry was added to a solution containing 70 parts of water, 2.0 parts of sodium carboxymethyl cellulose, 2.8 parts of glycerol, 0.8 parts of citric acid, 1.0 part of potassium citrate, and 2.0 parts of calcium carbonate. The resulting slurry was processed as in Example 12. Cigarettes of average weight 1.0 gram was prepared.

The sidestream and mainstream smokes produced from the cigarettes were milder and less irritating to the eyes and throat than those from an ordinary tobacco cigarette and the taste quality was superior to that of a cigarette made from the material produced without the addition of ammonia at the slurry stage.

EXAMPLE 14

A black heat-treated sheeted pulp was prepared by treating α -cellulose with sulphuric acid as described in Example 12. 20 parts of this black pulp were immersed in 20 parts

of a 35% aqueous nitric acid solution at 90° C for approximately 30 seconds. The product obtained was washed free from nitric acid with water and dried at 50° C to give 15 parts of sheet having an orange-brown colour resembling that of cigarette tobacco.

The sheet was further processed as described in Example 12 to give cigarettes which on smoking produced sidestream and mainstream smokes which were milder and less irritating to the eyes and throat than those from an ordinary cigarette.

EXAMPLE 15

A black heat-treated sheeted pulp was prepared by treating α -cellulose with sulphuric acid as described in Example 12. 20 parts of this black pulp were immersed in 40 parts of a 15% aqueous hydrogen peroxide solution for 6 hours. The product obtained was washed free from hydrogen peroxide with water and dried at 40° C to give 16 parts of sheet having an orange colour resembling that of cigarette tobacco.

The sheet was further processed as described in Example 12 to give cigarettes which on smoking produced sidestream and mainstream smokes which were milder and less irritating to the eyes and throat than those from an ordinary tobacco cigarette.

EXAMPLE 16

20 parts of a black heat-treated sheeted pulp, prepared by treating α -cellulose with sulphuric acid as described in Example 12, were exposed to gaseous ammonia at atmospheric pressure and ambient temperature for 60 minutes. Excess ammonia was removed under reduced pressure to give 21.4 parts of a dark brown sheeted pulp material.

The pulp material was further processed as described in Example 12 to give cigarettes which on smoking produced sidestream and mainstream smokes which were milder and less irritating to the eyes and throat than those of an ordinary tobacco cigarette.

EXAMPLE 17

19.0 parts of gum tragacanth were mixed with 20.0 parts of a 10% aqueous solution of ammonium sulphamate. The resulting wet paste was air-dried at 60° C and heated at 225° C for 18 minutes to give a black powder which weighed 60% of the original weight of gum tragacanth and ammonium sulphamate. The black powder was stirred with 35% aqueous nitric acid for 3 minutes at 90° C, filtered, washed free from acid with water and dried at 50° C to give a powder having an orange-brown colour.

The powder was ground to pass a 120 B.S.S. sieve. 11.4 parts of the powder were intimately mixed with 2.0 parts of calcium carbonate and added into 100 parts of a stirred aqueous solution containing 2.0 parts

of sodium carboxymethyl cellulose, 2.8 parts of glycerol, 0.8 parts of citric acid and 1.0 part of potassium citrate. The resulting slurry was processed as described in Example 12 to give cigarettes which on smoking produced sidestream and mainstream smokes which were milder and less irritating to the eyes and throat than those of an ordinary tobacco cigarette.

EXAMPLE 18

Cigarettes were prepared as described in Example 17 except that locust bean gum was used instead of gum tragacanth and the duration of heat-treatment at 225° C was 60 minutes. In this example the weight of black powder produced was 70% of the original weight of locust bean gum and ammonium sulphamate.

The sidestream and mainstream smokes produced from the cigarettes were very mild and less irritating to the eyes and throat than those from an ordinary tobacco cigarette.

EXAMPLE 19

Cigarettes were prepared as described in Example 17 except that gum arabic was used instead of gum tragacanth and the duration of heat treatment at 225° C was 30 minutes. The weight of black powder produced was 74% of the original weight of gum arabic and ammonium sulphamate.

The sidestream and mainstream smokes from the cigarettes were very mild and less irritating to the eyes and throat than those from an ordinary tobacco cigarette.

EXAMPLE 20

Cigarettes were prepared as described in Example 17 except that D-glucose was used instead of gum tragacanth and the duration of heat treatment at 225° C was 12 minutes. The weight of black powder produced was 63% of the original weight of D-glucose and ammonium sulphamate.

The sidestream and mainstream smokes from the cigarettes were very mild and less irritating to the eyes and throat than those from an ordinary tobacco cigarette.

EXAMPLE 21

Cigarettes were prepared as described in Example 17 except that methyl cellulose was used instead of gum tragacanth and the duration of heat treatment was 10 minutes at 225° C. The weight of black powder produced was 63% of the original weight of methyl cellulose and ammonium sulphamate.

The sidestream and mainstream smokes from the cigarettes were very mild and less irritating to the eyes and throat than those from an ordinary tobacco cigarette.

EXAMPLE 22

Cigarettes were prepared as described in Example 17 except that rice starch was used

instead of gum tragacanth and the duration of heat treatment was 35 minutes at 225° C. The weight of black powder produced was 66% of the original weight of rice starch and ammonium sulphamate.

5 The sidestream and mainstream smokes from the cigarettes were very mild and less irritating to the eyes and throat than those from an ordinary tobacco cigarette.

10 EXAMPLE 23

Cigarettes were prepared as described in Example 17 except that pectin was used instead of gum tragacanth and the duration of heat treatment was 20 minutes at 225° C. The weight of black powder produced was 60% of the original weight of pectin and ammonium sulphamate.

15 The sidestream and mainstream smokes from the cigarettes were very mild and less irritating to the eyes and throat than those from an ordinary tobacco cigarette.

20 EXAMPLE 24

Cigarettes were prepared as described in Example 17 except that alginic acid was used instead of gum tragacanth and the duration of heat treatment was 23 minutes at 225° C. The weight of black powder produced was 60% of the original weight of alginic acid and ammonium sulphamate.

30 The sidestream and mainstream smokes from the cigarettes were very mild and less irritating to the eyes and throat than those from an ordinary tobacco cigarette.

35 EXAMPLE 25

Cellulose sheet was held above the surface of a refluxing concentrated hydrochloric acid solution (approximately 110° C) for 2 hours, then dried at 60° C. The weight of the black-brown material produced was 75% of the original weight of cellulose. The degraded material was ground to a powder passing a 120 B.S.S. sieve and made into cigarettes of average weight 1.0 gram as in Example 12.

40 The sidestream and mainstream smokes produced from the cigarettes were milder and less irritating to the eyes and throat than those from ordinary tobacco cigarettes.

45 EXAMPLE 26

Cigarettes were prepared as in Example 12 except that a 10.0% solution of ferric chloride was used instead of sulphuric acid and the duration of heat treatment at 230° C was 20 minutes. The weight of the black material produced was 75% of the original weight of α -cellulose and ferric chloride.

50 The sidestream and mainstream smokes produced by the cigarettes were milder and less irritating than those from ordinary tobacco cigarettes.

55 EXAMPLE 27

60 Cigarettes were prepared as in Example 12

except that a 10.0% solution of sodium carbonate was used instead of sulphuric acid, the duration of heat treatment at 230° C was 20 minutes and the heat-treated pulp sheet was washed substantially free of sodium carbonate. The weight of the dark brown material produced was 73% of the original weight of α -cellulose and sodium carbonate.

The sidestream and mainstream smokes produced by the cigarettes contained less 'tar' than the smokes from ordinary tobacco cigarettes but were less pleasant than the smokes from the product of Example 12.

65 EXAMPLE 28

Cigarettes were prepared as in Example 12 except that a 7.5% solution of sodium hydroxide was used instead of sulphuric acid, the duration of heat treatment at 230° C was 15 minutes and the heat-treated pulp sheet was washed substantially free of sodium hydroxide. The weight of the dark brown material produced was 72% of the original weight of α -cellulose and sodium hydroxide.

The sidestream and mainstream smokes produced by the cigarettes were similar to those of Example 27.

WHAT WE CLAIM IS:—

1. A process for the preparation of a modified carbohydrate suitable for smoking mixtures which comprises subjecting carbohydrate to a catalysed degradation process at a temperature of 100 to 250° C until the weight of the degraded carbohydrate is less than 90% of the weight of the original carbohydrate.

2. A process as claimed in Claim 1 wherein the degradation treatment is continued until the weight of the modified carbohydrate is between 50 and 75% of the weight of the original carbohydrate.

3. A process as claimed in Claim 1 or Claim 2 wherein the degradation catalyst comprises a strong mineral acid or a salt of a strong mineral acid with a weak base.

4. A process as claimed in Claim 3 wherein the degradation catalyst comprises sulphamic acid, ammonium sulphamate, phosphoric acid, diammonium hydrogen phosphate, ammonium dihydrogen phosphate, sulphuric acid, ammonium sulphate, hydrochloric acid or ferric chloride.

5. A process as claimed in Claim 1 or Claim 2 wherein the degradation catalyst comprises alkali hydroxide or an alkali salt of a weak acid and the degraded carbohydrate is washed substantially free of catalyst.

6. A process as claimed in Claim 5 wherein the degradation catalyst comprises sodium hydroxide or sodium carbonate.

7. A process as claimed in any one of Claims 1 to 6 wherein the catalyst is present in an amount ranging from 1 to 20% by weight of the carbohydrate.

8. A process as claimed in any one of



instead of gum tragacanth and the duration of heat treatment was 35 minutes at 225° C. The weight of black powder produced was 66% of the original weight of rice starch and ammonium sulphamate.

The sidestream and mainstream smokes from the cigarettes were very mild and less irritating to the eyes and throat than those from an ordinary tobacco cigarette.

EXAMPLE 23

Cigarettes were prepared as described in Example 17 except that pectin was used instead of gum tragacanth and the duration of heat treatment was 20 minutes at 225° C. The weight of black powder produced was 60% of the original weight of pectin and ammonium sulphamate.

The sidestream and mainstream smokes from the cigarettes were very mild and less irritating to the eyes and throat than those from an ordinary tobacco cigarette.

EXAMPLE 24

Cigarettes were prepared as described in Example 17 except that alginic acid was used instead of gum tragacanth and the duration of heat treatment was 23 minutes at 225° C. The weight of black powder produced was 60% of the original weight of alginic acid and ammonium sulphamate.

The sidestream and mainstream smokes from the cigarettes were very mild and less irritating to the eyes and throat than those from an ordinary tobacco cigarette.

EXAMPLE 25

Cellulose sheet was held above the surface of a refluxing concentrated hydrochloric acid solution (approximately 110° C) for 2 hours, then dried at 60° C. The weight of the black-brown material produced was 75% of the original weight of cellulose. The degraded material was ground to a powder passing a 120 B.S.S. sieve and made into cigarettes of average weight 1.0 gram as in Example 12.

The sidestream and mainstream smokes produced from the cigarettes were milder and less irritating to the eyes and throat than those from ordinary tobacco cigarettes.

EXAMPLE 26

Cigarettes were prepared as in Example 12 except that a 10.0% solution of ferric chloride was used instead of sulphuric acid and the duration of heat treatment at 230° C was 20 minutes. The weight of the black material produced was 75% of the original weight of α -cellulose and ferric chloride.

The sidestream and mainstream smokes produced by the cigarettes were milder and less irritating than those from ordinary tobacco cigarettes.

EXAMPLE 27

Cigarettes were prepared as in Example 12

except that a 10.0% solution of sodium carbonate was used instead of sulphuric acid, the duration of heat treatment at 230° C was 20 minutes and the heat-treated pulp sheet was washed substantially free of sodium carbonate. The weight of the dark brown material produced was 73% of the original weight of α -cellulose and sodium carbonate.

The sidestream and mainstream smokes produced by the cigarettes contained less 'tar' than the smokes from ordinary tobacco cigarettes but were less pleasant than the smokes from the product of Example 12.

EXAMPLE 28

Cigarettes were prepared as in Example 12 except that a 7.5% solution of sodium hydroxide was used instead of sulphuric acid, the duration of heat treatment at 230° C was 15 minutes and the heat-treated pulp sheet was washed substantially free of sodium hydroxide. The weight of the dark brown material produced was 72% of the original weight of α -cellulose and sodium hydroxide.

The sidestream and mainstream smokes produced by the cigarettes were similar to those of Example 27.

WHAT WE CLAIM IS:—

1. A process for the preparation of a modified carbohydrate suitable for smoking mixtures which comprises subjecting carbohydrate to a catalysed degradation process at a temperature of 100 to 250° C until the weight of the degraded carbohydrate is less than 90% of the weight of the original carbohydrate.

2. A process as claimed in Claim 1 wherein the degradation treatment is continued until the weight of the modified carbohydrate is between 50 and 75% of the weight of the original carbohydrate.

3. A process as claimed in Claim 1 or Claim 2 wherein the degradation catalyst comprises a strong mineral acid or a salt of a strong mineral acid with a weak base.

4. A process as claimed in Claim 3 wherein the degradation catalyst comprises sulphamic acid, ammonium sulphamate, phosphoric acid, diammonium hydrogen phosphate, ammonium dihydrogen phosphate, sulphuric acid, ammonium sulphate, hydrochloric acid or ferric chloride.

5. A process as claimed in Claim 1 or Claim 2 wherein the degradation catalyst comprises alkali hydroxide or an alkali salt of a weak acid and the degraded carbohydrate is washed substantially free of catalyst.

6. A process as claimed in Claim 5 wherein the degradation catalyst comprises sodium hydroxide or sodium carbonate.

7. A process as claimed in any one of Claims 1 to 6 wherein the catalyst is present in an amount ranging from 1 to 20% by weight of the carbohydrate.

8. A process as claimed in any one of

Claims 1 to 7 wherein the carbohydrate comprises cellulose or α -cellulose, cellulose derivative, mono- or polysaccharide, starch, alginate, pectin or natural gum.

5 9. A process as claimed in Claim 8 wherein the carbohydrate comprises methyl cellulose, glucose, sucrose, gum tragacanth, gum arabic, locust bean gum, rice starch, potato starch or maize starch.

10 10. A process as claimed in any one of Claims 1 to 9 wherein the carbohydrate is mixed or impregnated with a solution of catalyst and dried before subjection to the degradation process.

15 11. A process as claimed in any one of Claims 1 to 10 wherein the carbohydrate and a catalyst are maintained at a temperature of 100–250° C for a period of 1 minute to 4 hours.

20 12. A process as claimed in any one of Claims 1 to 11 wherein the degraded carbohydrate is treated with nitric acid, hydrogen peroxide or ammonia to alter the colour of the degraded carbohydrate.

25 13. A process as claimed in Claim 12 wherein the treatment is carried out at elevated temperature.

30 14. A process as claimed in any one of Claims 1 to 13 wherein any residual acid in the degraded carbohydrate is neutralised with ammonia.

35 15. A process as claimed in any one of Claims 1 to 13 wherein the degraded carbohydrate is prepared in sheet form.

40 16. A process as claimed in Claim 15 wherein the degraded carbohydrate is mixed in powder form with a solution of a film-forming agent and sheeted.

45 17. A process as claimed in Claim 16 wherein the film-forming agent comprises a water-soluble cellulose derivative, polyvinyl alcohol, starch, pectin, gum or mucilage.

50 18. A process as claimed in Claim 17 wherein the film-forming agent comprises water-soluble methyl cellulose or sodium carboxymethyl cellulose.

55 19. A process as claimed in Claim 18 wherein the methyl cellulose or sodium carboxymethyl cellulose is such that a 2% aqueous solution has a viscosity of 1500 cps. at 25° C.

20. A process as claimed in any one of

Claims 16 to 19 wherein any residual acid in the mixture is neutralised by addition of ammonia to the mixture of degraded carbohydrate and solution of film-forming agent. 55

21. A process as claimed in any one of Claims 1 to 20 wherein a glow-promoting catalyst, nicotine, flavourants, medicaments or ash or colour improver is mixed with the degraded carbohydrate. 60

22. A process as claimed in Claim 21 wherein an alkali metal compound is added as glow-promoting catalyst.

23. A process as claimed in Claim 21 wherein a salt of ammonia, an alkali metal or an alkaline earth metal is added to the degraded carbohydrate as ash improver. 65

24. A process as claimed in Claim 21 wherein tobacco extract, organic ester, essential oil, menthol, tonka bean or vanillin is added to the degraded carbohydrate as flavourant. 70

25. A process as claimed in Claim 21 wherein glycerol or glycol is added to the degraded carbohydrate as a humectant. 75

26. A process as claimed in Claim 21 wherein a carbonate or porous inert filler is added to the degraded carbohydrate to facilitate combustion. 80

27. A process as claimed in any one of Claims 15 to 26 wherein the degraded carbohydrate sheet is shredded to form a cigarette or pipe filling.

28. A process as claimed in Claim 27 wherein the shred material is conditioned in a humid atmosphere to a moisture content of 5 to 15% by weight. 85

29. A process for the production of a modified carbohydrate material for smoking mixtures substantially as herein described with reference to the Examples. 90

30. A modified carbohydrate material for smoking mixtures whenever prepared by a process as claimed in any one of Claims 1 to 29. 95

31. Smoking mixtures comprising modified carbohydrate material as claimed in Claim 30.

32. A cigarette, cigar or pipe filling comprising a smoking mixture as claimed in Claim 31. 100

THOMAS J. REID
Agent for the Applicants

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ERRATA

SPECIFICATION No. 1,113,979

Page 2, line 37, for "caalysed" read "catalysed"

Page 2, line 78, for "carbonhydrate" read "carbohydrate"

Page 3, line 42, for "shreaded" read "shredded"

THE PATENT OFFICE
24th June 1968

