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Briskin et al.

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[54] **SMOKABLE TOBACCO SUBSTITUTE MATERIAL AND METHOD**

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[52] **U.S. Cl.** ..... **131/2,** 131/17 R

[51] **Int. Cl.** **A24b 15/00**

[58] **Field of Search** 131/2, 15, 17, 140–144

[56] **References Cited**

UNITED STATES PATENTS

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Primary Examiner—Melvin D. Rein

[57] **ABSTRACT**

A smokable material formed by heating a cellulosic material, containing 3–15 percent by weight calcium and/or magnesium oxalate, at a temperature within the range of 275°–375°C for a time sufficient to effect a weight loss of the cellulosic material at least 60 percent.

39 Claims, No Drawings

SMOKABLE TOBACCO SUBSTITUTE MATERIAL AND METHOD

FIELD OF INVENTION

This invention relates to a smokable material to be used as a filler in cigars, cigarettes and pipes and/or as a wrapper for cigars and cigarettes, and it relates more particularly to a material which can be used as a tobacco substitute, alone or in admixture with tobacco in cigars, cigarettes, pipes and the like.

DESCRIPTION OF THE PRIOR ART

Considerable effort has been, and is being, expended in the search for material that can be used as a substitute for tobacco but in which undesirable characteristics of tobacco are materially minimized, if not completely eliminated.

Attempts have been made to replace tobacco with vegetable components, in the form of leaves, stems, tassels and the like, which are treated in somewhat the same way as tobacco leaves. Such attempts have found little acceptance by reason of the undesirable taste as well as the high levels of tar, polynuclear aromatics, and carbonyls in the smoke.

Another direction that has been taken by research was to produce a reconstituted smoking product which made use of a combustible organic filler in admixture with a binder, which was formed into a sheet or product intended for use as a filler in cigars, cigarettes or pipes. The cellulosic esters or ethers, alginates and natural gum, which have been used in large quantities for binders, burn with an undesirable strong taste and odor.

More recently, as a product of the research which preceded this invention, a smokable material was produced by slow, controlled and selective oxidation of cellulosic materials in the presence of liquid and/or gaseous nitrogen dioxide, as described in U.S. Pat. Nos. 3,478,752; 3,482,578; 3,491,766; 3,516,416; 3,556,109; 3,556,110; 3,559,655; 3,575,177; 3,577,994; 3,608,559; 3,608,560 and 3,643,668. While a smokable material having desirable characteristics for use as a tobacco substitute can be produced in accordance with the processes described in the aforementioned patents, the product is comparatively expensive by reason of the time required to achieve the desired oxidation and the high cost of labor and materials that are required.

Unless cheaper methods are found to stabilize oxycellulose, and further improvements are made to cut down acridity, the existing smokable mixtures based on oxycellulose do not offer much hope for an easily acceptable synthetic smoking material.

OBJECT OF THE INVENTION

It is an object of this invention to produce and to provide a method for producing a smokable material for use as a filler and/or wrapper in cigarettes, cigars, pipes and the like; in which readily available and inexpensive raw materials are employed; in which the process for preparation is simple and without the need for expensive labor; in which the process for preparation can be carried out as a continuous operation for mass production at low cost and in large volume; in which the smoke generated from the product is relatively free from tar and polycyclics, and does not form metal carbonyls or other undesirable components. The product

of the present invention has the appearance, burning and smoking characteristics of properly cured tobacco, and it can be formulated to provide a desirable smoke generation, taste, aroma and ashing characteristics. It can also be mixed with tobacco to produce a smoking product with less tars and controlled nicotine.

An important objective of this invention relates to utilization of extensive tobacco wastes in the form of tough near-root portions of stems, broken leaves and tobacco dust. The present day methods employed by the industry invariably require expensive sheet forming equipment and the product is of inferior smoking quality as it is low in tobacco aroma but very high in non-aromatic components such as cellulose, lignin, and pectins which burn with noxious odors. Our invention gives lead to the tobacco industry to usefully extract the soluble constituents from the tobacco scrap and incorporate them with our completely inocuous smoking mixture in place of natural tobacco residues left after extraction. Since the smoking mixture of our invention can directly be obtained in the form of sheets, on which the tobacco extracts can be applied from solutions, we further do away with binders which are required for reconstituting sheets. The most salient feature of this method is the control on nicotine and aroma content in the smoking mixtures to meet the varied consumer needs.

SUMMARY OF THE INVENTION

It has been found that a smokable material can be produced, in accordance with the practice of this invention, by heating a cellulosic material, 3–3–15 percent by weight calcium and/or magnesium oxalate, at a temperature within the range of 275°–375°C in a non-combustible (preferably non-oxidizing atmosphere, until the weight loss of cellulosic material is at least 60 percent and preferably within the range of 60–70 percent. Our experience has indicated that if this weight loss is below 60 percent some acridity is generated on smoking.

The resulting product remains sufficiently pliable for sheeting or, if already in the form of a sheet, for processing to the desired form for use as a cigarette, cigar or pipe filler or wrapper and which yields practically no smoke or tar and is relatively free of acridity, polycyclics or other irritants that give bad taste or odor.

The product can be advantageously modified by the addition of humectants, smoke generating materials, ashing components, coloring agents, and additives for improving taste, aroma, strength and storability, without noticeable increase in undesirable components in the smoke generated therefrom.

DESCRIPTION OF PREFERRED EMBODIMENT

EXAMPLE 1

In the preferred practice of this invention, a sheet of alpha-cellulose, containing 7 percent by weight calcium oxalate, uniformly distributed with the fibers of the alpha-cellulose, is advanced continuously through an enclosed space heated to a temperature of 320°C for about 60 minutes, to achieve weight loss of 70 percent. 400 mg of a sheet portion was cut out and treated with 30 mg of gum tragacanth to impart additional strength. After air drying, 1.2 ml of an aqueous solution containing 60 mg potassium citrate, 12 mg sodium citrate, 12 mg disodium hydrogen phosphate, 18 mg sodium bicarbonate, 30 mg inositol and 35 mg glycerine was incor-

porated in the sheets and air dried. 250 mg of color formed by the reaction of triquinonyl with calcium carbonate was thoroughly mixed with the solution of 40 mg of polyvinylene carbonate in 8 to 10 ml of acetone. This color slurry, along with 2.5 mg of Primetine JMT was brushed on the above sheet to cover the entire surface area. The colored sheets were sprayed with water to acquire 30 to 40 mg of moisture and shredded. The smoking mixture thus produced when rolled into a cigarette and smoked was extremely mild with very little taste and odor. It exhibited good burning characteristics such as white flickable ash, bright glow in the tip region, and burning rate commensurate with the cigarette paper.

Instead of making use of alpha-cellulose mentioned in Example 1, use can be made of other forms of cellulosic materials, such as wood or paper pulp, straw, flax, bamboo, cotton, hemp, rice fibers, and other vegetative fibers, and the like filaments of carbohydrate materials, hereinafter referred to generally as cellulosic materials. Use can be made of such cellulosic materials in fibrous form other than in sheets but it is preferred to make use of sheets formed of such cellulosic materials for carrying out the prescribed treatment in a continuous, low cost, high speed, mass production process which yields a more uniform product at less cost.

The calcium oxalate can be incorporated in the desired amount in various ways. For most intimate and uniform distribution with the cellulosic material, the calcium oxalate can be formed in situ in the cellulosic material by first treating the cellulosic material to load the cellulosic material with 3–15 percent by weight of calcium carbonate, calcium oxide or other calcium salt followed by treatment, as by one or more impregnations, with a dilute solution containing stoichiometric quantities of oxalic acid whereupon the oxalic acid reacts with the calcium salt to form an insoluble calcium oxalate in situ in the cellulosic material.

Instead, the calcium oxalate can be introduced into the header or otherwise admixed with the cellulosic fibers for inclusion within the pulp that is run over the Fourdrinier screen of a paper making machine. The calcium oxalate can be incorporated in less intimate association with the cellulosic material, as by impregnation of the cellulosic material with an aqueous medium containing the calcium oxalate in suspension in the desired concentration. When use is made of cellulosic material other than in sheet form, the calcium oxalate can be admixed with the cellulosic material in the form of a dry powder or aqueous suspension, and then sheeted or dried for subsequent thermal treatment in the manner described.

It is desirable to make use of at least 3 percent by weight but not more than 15 percent by weight of the calcium oxalate and preferably an amount within the range of 7–10 percent by weight of the cellulosic material. Instead of calcium oxalate, use can be made of magnesium oxalate or other alkaline earth metal salt of oxalic, citric, or pivalic acids or mixtures thereof in amounts equivalent to the calcium oxalate.

Thermal treatment of the cellulosic material should be carried out in an atmosphere which does not support combustion of the cellulosic material at the temperatures to which the cellulosic material is exposed. It is preferred to make use of a non-oxidizing atmosphere and preferably an inert or reducing atmosphere. For this purpose, it is desirable to make use of an enclosed heating chamber without air circulation therethrough, although air may enter in small amounts through the inlet and outlet openings at which the cellulosic material enters and leaves the chamber or a nonoxidizing or inert gas, such as nitrogen, carbon dioxide and the like can be circulated through the enclosed heated space for control of atmospheric conditions.

Instead of 60 minutes exposure at 320°C, as described in Example 1, the cellulosic material can be exposed to a temperature within the range of 275°–375°C, for a time depending somewhat upon the temperature employed but, under no conditions, less than a time and temperature sufficient to bring about at least a 60 percent but not more than about 75 percent weight reduction of the cellulosic material. In a continuous operation, which makes use of cellulosic material in sheet form, the time of exposure can be controlled by variation in the rate of travel of the sheet through the heated space and/or the length of the path of travel of the cellulosic material therethrough. The sheet can be advanced through the heated space for travel back and forth over rollers or it can be supported during travel through the heated space on endless wire belts or the like.

In a continuous operation, which makes use of cellulosic material other than in sheet form, such as in the form of separated fibers and the like, such cellulosic material can be supported on a foraminous screen or wire belt during travel through the heated space. In batch operation, the cellulosic material is merely exposed to the desired temperature for a time sufficient to effect the desired degradation, preferably in an arrangement that will provide for relatively uniform exposure of the cellulosic material to the elevated treating temperature conditions existing.

The product obtained from the described thermal treatment is grayish-black in color. If processed in sheet form, the product issuing from the heat treating chamber usually is still in the form of a sheet having sufficient mass integrity and flexibility to enable incorporation of additives, and processing into the final product. The incorporation of additives is desirable to improve strength, taste, aroma, ashing characteristics, glow or burning properties and color.

To improve mass integrity and strength, addition can be made of a binder in the amount of 1–5 percent by weight of the treated cellulosic material and preferably in an amount within the range of 2–4 percent by weight. For this purpose, it is preferred to make use of a gum, such as guar gum, gum tragacanth, and the like natural gums, a cellulosic derivative such as methyl cellulose, carboxymethyl cellulose, hydroxyethyl cellulose, and the like, preferably water soluble cellulosic resins.

The mass integrity and strength is further enhanced on incorporation of a polyalkylene carbonate such as polyvinylene carbonate, as described in the copending application Ser. No 252,003. This resin has been preferably selected for various reasons such as:

1. polyvinylene carbonate forms a very strong and flexible sheet which burns cleanly without contributing undesirable odor or taste;
2. the reconstituted sheet or sheet treated with the polyvinylene carbonate is water resistant thereby to militate against leaching of water soluble additives;

3. it serves as a vehicle for coating the color on the sheets;
4. it contributes to the resiliency of the product and includes the filling power of the smokable material when employed as a filler for cigarettes, cigars and pipes;
5. it improves the stability of the smokable material during storage and helps maintain the moisture balance in the smoking product.

It is also desirable to incorporate a humectant which is effective to keep the smokable material moist and pliable and to enhance the packing characteristics as well as the burning characteristics of the product. For this purpose, use can be made of a polyhydric alcohol, such as glycerol or glycol such as ethylene glycol, propylene glycol, and the like, inositol, butane diol and the like hydroscopic materials and mixtures thereof. These materials can be applied by spraying from water solution onto the sheet or by admixture with the treated cellulosic material. Addition is made in amounts up to 10 percent by weight and preferably in an amount within the range of 1–4 percent by weight.

The glow and ashing characteristics of the smoking material of this invention can be improved by the addition of a water soluble alkali metal salt, such as sodium or potassium salt of a low molecular weight hydroxy acid such as oxalic, citric, maleic, pivalic and the like organic acid, or carbonates, bicarbonates, or phosphates, such as potassium citrate, sodium citrate, potassium bicarbonate, potassium maleate and dihydrogen sodium phosphate, and the like, and mixtures thereof. Such mineralizing agents or ashing ingredients can be incorporated in amounts up to 30 percent by weight of the smokable material and preferably in an amount within the range of 5–10 percent by weight.

From the standpoint of appearance, it is desirable that the smokable material have the dark brownish color of rich cured tobacco. The dark gray cellulosic material resulting from the described thermal treatment of this invention cannot be colored by conventional dyestuffs unless employed in enormous quantities.

It has been found that novel use, as a coloring material, can be made of triquinonyl $C_6O_6.8H_2O$ (copending application Ser. No. 252,003) which produces a strong orange to brown color with the calcium of magnesium in the cellulosic product to provide a non-leachable coloring agent that is effective to convert the dark gray product to a rich brown color which cannot be leached from the product.

Instead of forming the coloring component in situ by reaction of the triquinonyl with the calcium or magnesium present in the cellulosic product, the desired results can be achieved by forming the coloring component separate and apart from the cellulosic material and then incorporating the separately formed coloring agent with the cellulosic material with the aid of a binder, such as polyvinylene carbonate, to provide a colored binder which is added in the manner described in Example 1.

Flavor and aroma can be improved by the addition of flavoring agents such as nicotine, menthol, chlorogenic, caffeic and quinic acids, essential oils, tobacco extracts and the like.

EXAMPLE 2

The procedure of Example 1 was repeated in full, except for incorporation of alcohol and water extracts of a tobacco stem material before applying the color with polyvinylene carbonate on the sheets. Thus 120 mg of water extract was brushed on the sheets from 1 ml solution and after air drying, 60 mg of alcohol extract contained in 1 ml of alcoholic solution were brushed on and again air dried. The sheets had rich tobacco color, texture and aroma and when fabricated into a cigarette produced a very mild smoke rich in tobacco aroma and good burning characteristics.

EXAMPLE 3

Same as in Example 2 but 5 mg of menthol was incorporated along with color and polyvinylene carbonate. The cigarette fabricated from this material had all the essential qualities of Example 2 and a pleasant menthol flavor.

EXAMPLE 4

A smoking mixture was prepared according to the procedure of Example 1. 750 mg of the shreds were admixed with 250 mg of natural tobacco strands and fabricated into a regular cigarette and smoked. The cigarette had all the essential qualities of Examples 1 and 2 and produced very mild smoke in comparison to a regular cigarette based on 100 percent natural tobacco of the same brand.

EXAMPLE 5

Same as in Example 4 but the ratio of tobacco to our non-tobacco smoking mixture was reversed. The cigarette produced more smoke and flavor but was still considerably milder thn a regular cigarette based on 100 percent natural tobacco of the same brand.

EXAMPLE 6

As in Example 1 but potassium bicarbonate was eliminated. The ash was somewhat flaky with no other material difference.

EXAMPLE 7

As in Example 1 but disodium hydrogen phosphate was eliminated. The ash was not easily flickable and the mixture burned faster and unevenly with respect to the wrapper.

It will be apparent from the foregoing that there is provided a new and improved smokable material which is relatively free of the objectionable characteristics of cured tobacco but which can be used instead of or in combination with cured tobacco in the manufacture of a smokable material such as fillers for cigarettes, cigars and pipes or wrappers for cigarettes and cigars. The smokable material described may be prepared by a method which is capable of continuous mass production operations to produce an inexpensive product which is readily processed into the form desired for use as a smokable material.

It will be understood that changes may be made in the details of formulation and operation without departing from the spirit of the invention, especially as defined in the following claims.

We claim:

1. A smokable material comprising the product of the thermal reaction of a cellulosic material in the form of a fibrous carbohydrate containing 3–15 percent by weight alkaline earth metal salt of a carboxylic acid, at a temperature within the range of 275°–375°C in a non-

oxidizing atmosphere until the weight loss of cellulosic material is at least 60 percent.

2. A smokable material as claimed in claim 1 in which the thermal reaction of the cellulosic material is carried out until the weight loss of cellulosic material is within the range of 60–75 percent.

3. A smokable material as claimed in claim 1 in which the alkaline earth metal salt is selected from the group consisting of calcium and magnesium oxalate.

4. A smokable material as claimed in claim 3 in which the calcium and magnesium oxalate are present in an amount within the range of 3–15 percent by weight.

5. A smokable material as claimed in claim 1 which includes a binder present in an amount within the range of 1–5 percent by weight.

6. A smokable material as claimed in claim 5 in which the binder is a water soluble binder selected from the group consisting of cellulose derivatives and water soluble gums.

7. A smokable material as claimed in claim 1 which includes a humectant in an amount up to 10 percent by weight.

8. A smokable material as claimed in claim 7 in which the humectant is present in an amount within the range of 1–10 percent by weight of the smokable material.

9. A smokable material as claimed in claim 7 in which the humectant is a polyhydric compound selected from the group consisting of glycerol, glycol, and polyhydric alcohol.

10. A smokable material as claimed in claim 1 which includes a mineralizing and ashing ingredient in an amount up to 10 percent by weight of the smokable material.

11. A smokable material as claimed in claim 10 in which the mineralizing and ashing ingredient is present in an amount within the range of 3–15 percent by weight.

12. A smokable material as claimed in claim 10 in which the mineralizing agent and ashing ingredient is selected from the group consisting of an alkali metal salt of a lower carboxylic acid, carbonate, bicarbonate and phosphate.

13. A smokable material as claimed in claim 12 in which the carboxylic acid is selected from the group consisting of oxalic acid, citric acid, pivalicacid and maleic acid.

14. A smokable material as claimed in claim 1 which includes a coloring agent for converting the dark gray cellulosic product to a rich brown color.

15. A smokable material as claimed in claim 14 in which the coloring agent comprises the reaction product of a calcium base with triquinonyl.

16. A smokable material as claimed in claim 1 which includes an organic amine in amount sufficient to adjust the pH of the smokable material to a pH of at least 8.

17. In the method of producing a smokable material comprising the steps of combining an alkaline earth metal salt of an organic hydroxy acid with a cellulosic material in an amount within the range of 3–15 percent by weight of the cellulosic material, exposing the cellulosic material in a non-combustible atmosphere to a temperature within the range of 275°–375°C for a time sufficient to effect a weight loss of at least 60 percent but not more than 75 percent of the cellulosic material, and then processing the thermally reacted cellulosic product to the form desired for the smokable material.

18. The method as claimed in claim 17 in which the cellulosic material is exposed to the elevated temperature in a non-oxidizing atmosphere or under vacuum.

19. The method as claimed in claim 17 in which the alkaline earth metal salt of the organic hydroxy acid is selected from the group consisting of calcium and magnesium oxalate.

20. The method as claimed in claim 19 in which the salt is present in an amount within the range of 3–15 percent by weight of the cellulosic material.

21. The method as claimed in claim 17 which includes the step of coating the product of the thermal reaction with a coloring agent that converts the dark gray color of the thermal reaction product to a rich brown.

22. The method as claimed in claim 21 in which the coloring agent is formed by adding triquinonyl to the thermal reaction product for reaction with the calcium or magnesium salts present to form a brown-orange color.

23. The method as claimed in claim 21 in which the coloring agent is formed by a reaction between triquinonyl and a calcium or magnesium compound and then coating the colored reaction product onto the thermally reacted cellulosic material.

24. The method as claimed in claim 17 which includes the step of incorporating a binder with a thermally reacted cellulosic material in an amount within the range of 1–5 percent by weight of the cellulosic material.

25. The method as claimed in claim 24 in which the binder is sprayed onto the cellulosic material.

26. The method as claimed in claim 24 in which the binder is admixed for uniform distribution with the cellulosic material.

27. The method as claimed in claim 24 in which the binder is polyvinylene carbonate.

28. The method as claimed in claim 17 which includes the step of incorporating a humectant with the thermally reacted cellulosic material in an amount up to 5 percent by weight.

29. The method as claimed in claim 28 in which the humectant is a water soluble polyhydric organic compound.

30. The method as claimed in claim 28 in which the humectant is a compound selected from the group consisting of glycerol, a glycol and a polyhydric alcohol.

31. The method as claimed in claim 17 which includes the step of adding an ashing ingredient to the thermally reacted cellulosic material in an amount up to 30 percent by weight.

32. The method as claimed in claim 31 in which the ashing ingredient is added in an amount up to 6–30 percent by weight of the thermally reacted cellulosic material.

33. The method as claimed in claim 31 in which the ashing ingredient is selected from the group consisting of an alkali metal and ammonium salt of an organic acid, carbonate, bicarbonate and phosphate.

34. The method as claimed in claim 17 which includes the step of adjusting the pH of the smoke by the addition of an organic amine, ammonium salt or urea, in an amount to adjust the pH from 7 to 8.5.

35. The smokable material as claimed in claim 1 which includes natural tobacco products, tobacco ex-

tracts or reconstituted tobacco materials, in any percentage, form and shape.

36. The smokable material as claimed in claim **1** which includes additives for casing, flavoring and aroma.

37. The smokable material as claimed in claim **36** which includes nicotinic, chlorogenic, caffeic or quinic acids or their derivatives as flavoring agents.

38. The smokable material as claimed in claim **36** which includes menthol as a flavoring agent.

39. A cigar filler, cigarette filler, cigar wrapper, cigarette wrapper and pipe filler comprising the smokable material as claimed in claim **1.**

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UNITED STATES PATENT OFFICE
CERTIFICATE OF CORRECTION

Patent No. 3,861,401 Dated January 21, 1975

Inventor(s) Theodore S. Briskin et al.

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

column 2, line 32, after "materials,"

cancel "3-" and substitute --- containing ---.

Signed and sealed this 15th day of April 1975.

(SEAL)
Attest:

RUTH C. MASON
Attesting Officer

C. MARSHALL DANN
Commissioner of Patents
and Trademarks

USCOMM-DC 60376-P69
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