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[54] **SUBSTITUTE SMOKING MATERIAL EMPLOYING DEGRADED CELLULOSE**

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[56] **References Cited**

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[57] **ABSTRACT**

A substitute tobacco material obtained from degraded cellulose having a D.P. (degree of polymerization) distribution such that the average D.P. does not exceed 95 and the portion with D.P. in excess of 110 desirably accounts for not more than 10 percent of the whole composition. The said degraded cellulose can be obtained by converting cellulose into alkali cellulose through alkali treatment, adjusting the alkali cellulose to an average D.P. below 95, neutralizing the alkali cellulose, washing with water and drying. This degraded cellulose may be utilized in its unmodified state as a substitute tobacco material or as extender or carrier for tobacco products. A mixture of this degraded cellulose and at least one member selected from the group consisting of sodium cellulose glycolate, hydroxyethyl cellulose, methyl cellulose and ethyl cellulose provides a substitute tobacco material possessed of excellent film-forming property and proves useful for the manufacture of various tobacco products when the mixture contains the degraded cellulose by 75 percent or over. Tobacco products using such a substitute tobacco material enjoy an advantage that the contents of tars and nicotine liable to do harm to the human system at the time of smoking are lowered.

4 Claims, No Drawings

SUBSTITUTE SMOKING MATERIAL EMPLOYING DEGRADED CELLULOSE

This invention relates to a substitute tobacco material suitable for use instead of naturally produced leaf tobacco as extender for tobacco products or directly as tobacco products and to a method for the manufacture thereof. The substitute tobacco material comprises a white, finely comminuted degraded cellulose produced, on a commercial scale, by decomposing cellulose at a temperature not exceeding 100°C under mild conditions to an extent such that the average D.P. as calculated according to the formula described hereinafter will not exceed 95 and the portion with D.P. in excess of 110 will desirably account for not more than 10 percent of the whole composition. It is manufactured by steeping cellulose in an alkali solution thereby converting it into alkali cellulose, adjusting the alkali cellulose to an extent such that the average D.P. will not exceed 95 and the portion with D.P. in excess of 110 will desirably account for not more than 10 percent of the whole composition, thereafter neutralizing the alkali cellulose, and washing with water and drying.

There have been marketed a variety of tobacco products including cigars, cigarettes, pipe tobacco and the like. The materials therefor are invariably obtained by cultivating naturally produced leaf tobacco. It is universally known that materials for such variety of tobacco products as mentioned above are derived from subjecting the natural leaf tobacco to different kinds of processing. In recent years, the harm the smoking is doing to the human system has come to arouse public criticism and the cause for this harm is being elucidated. One possible cause is that the natural leaf tobacco contains many chemical components responsible for the harm. At the time a given tobacco product is burnt, such components are entrained by the smoke which is inhaled into the lungs, with a result that harmful components manifest an adverse effect upon the human system. To preclude this harm, it is necessary either to deprive the natural leaf tobacco of such harmful components or to obtain a harmless tobacco material which can take place of natural tobacco. Removal of harmful components from the natural leaf tobacco proves disadvantageous from the viewpoint of equipment and economy.

A few methods have been suggested concerning tobacco materials designed for use instead of natural leaf tobacco. The first of these conventional methods proposes to obtain a substitute tobacco material by drying and processing leafy vegetable, such as lettuce and cabbage. The second method causes pulp to be beaten until its freeness reaches a fixed value and uses the resultant beaten cellulose pulp as substitute for tobacco. The third method oxidizes cellulose with nitrogen oxide so as to obtain a polymer of glucuronic anhydride having a carboxyl group substitute for a methylol group at the C_6 position and uses this polymer as substitute for tobacco (U.S. Pat. No. 3,478,751). The fourth method decomposes a carbohydrate at 100°C or higher temperature in the presence of a catalyst until the weight thereof decreases to below 90% of the original weight and uses the resultant modified carbohydrate as substitute for tobacco (British Pat. No. 1,113,979).

In the case of the product by the first method, since the leafy vegetable as the raw material is a natural product obtained by cultivation, it is liable to provide highly heterogeneous chemical components. It is, therefore, difficult for this method to ensure uniform quality of products. Considering that such leafy vegetable contains many detrimental chemical components similarly to the natural leaf tobacco, one cannot expect this method to afford a guarantee that the substitute tobacco thereof will produce only harmless smoke on burning. The product of the second method is merely an outcome of physical processing of cellulose. It has therefore a disadvantage that the smoke and taste generated by this substitute tobacco retains the pungency of the nature produced when cellulose is burnt. In the third method, when the methylol group at the C_6 position of cellulose is desired to be converted selectively to the carboxyl group, the reaction is liable to entail secondary reactions such as, for example, conversion of the methylol group to other than the carboxyl group and oxidation of the carbon at other than the C_6 position. Thus, this method has a disadvantage that the product aimed at cannot be obtained in high yields. The fourth method involves a treatment which is performed at a temperature of about 200°C and, therefore, proves disadvantageous in terms of operational efficiency as well as heat economy. Moreover, the product of this method is obtained in a weight which is not more than 90 percent, preferably 50 to 75 percent, of the original weight of the raw material. Because of this heavy loss during treatment in the course of manufacture, this method is highly uneconomical.

As a substitute for natural leaf tobacco free from such drawbacks as mentioned above, the inventors have discovered a substitute tobacco material which may be used as extender for tobacco products or which may be utilized directly as tobacco products. To be specific, they have made a discovery that a white, degraded cellulose derived by decomposing cellulose at a temperature below 100°C under mild conditions until there is obtained a D.P. distribution such that the average D.P. calculated according to the formula described hereinafter does not exceed 95 and the portion with D.P. in excess of 110 desirably accounts for not more than 10 percent of the whole composition and the tobacco material using this degraded cellulose as the principal ingredient generate smoke which has extremely low pungence and is substantially free from taste and odor.

The present invention provides a substitute tobacco material comprising a white degraded cellulose possessed of a D.P. distribution such that the average D.P. represented by the following formula is not more than 95 and the portion with D.P. in excess of 110 desirably accounts for not more than 10 percent of the whole composition.

$$\text{Average D.P., } \overline{DP} = \frac{(\sqrt[4]{\eta_{rel}} - 1) \times 8 \times 10^3}{C}$$

wherein, η_{rel} denotes the relative viscosity determined by dissolving 50 mg of degraded cellulose under discussion in 10 cc of ammonia solution of copper oxide having a copper concentration of 11 g/lit and an aqueous ammonia solution concentration of 210 g/lit, measuring the flow time, t_1 (in seconds), required by the degraded cellulose solution and the flow time, t_o (in seconds), required by the ammonia solution of copper oxide respectively in accordance with the capillary method (using Oken's modified viscosimeter), and cal-

culating the quotient of the division of T_1 by T_0 and C denotes the concentration of the degraded cellulose, in g/lit, in the ammonia solution of copper oxide).

It is another object of the present invention to provide a method for the manufacture of a substitute tobacco material of the type described above.

It is also another object of this invention to provide a mixed composition which comprises the abovedefined degraded cellulose possessed of a D.P. distribution such that the average D.P. does not exceed 95 and the portion with D.P. in excess of 110 desirably accounts for not more than 10 percent of the whole composition and one or more members selected from the group consisting of sodium cellulose glycolate, hydroxyethyl cellulose, methyl cellulose and ethyl cellulose.

It is still another object of the present invention to provide a method for the manufacture of the aforementioned mixed composition which may be uniformly mixed at any desired proportion with natural tobacco.

It is yet another object of this invention to provide a tobacco of such nature that the smoke generated at the time of consumption thereof contains at lowered concentrations tars, nicotine and other harmful components to the human system.

The other objects of the present invention will become apparent from the following description.

Degraded cellulose

The white, powdery or fibrous, degraded cellulose obtained by the method of this invention and possessed of a D.P. distribution such that the average D.P. does not exceed 95 and the portion with D.P. in excess of 110 desirably accounts for not more than 10% of the whole composition does not, at the time of combustion, produce the yellow flame and the pungent smoke which are inherent to cellulose. Instead, it burns in much the same way as the natural leaf tobacco. The smoke generated at the time of combustion possesses a flavor virtually free from taste and odor. A degraded cellulose having an average D.P. exceeding 95 is not desirable, because it is liable, at the time of combustion, to generate the yellow flame and pungent smoke inherent to cellulose as mentioned above. A degraded cellulose having a D.P. distribution such that the average D.P. is not more than 95 but the portion with D.P. in excess of 110 accounts for more than 10 percent by weight is less satisfactory than the degraded cellulose of this invention whose D.P. distribution is such that the portion with D.P. in excess of 110 accounts for not more than 10 percent. Table 1 shows the results of one panel test performed on man-made tobacco prepared by using degraded celluloses of varying values of D.P. The data indicate that pungence in smoke and flavor was perceived by quite many panel members where the average D.P. of degraded celluloses was in excess of 102, by a very few panel members where the average D.P. was 95, and by no panel members where the average D.P. was 87. In a separate panel test, a tobacco prepared by using a degraded cellulose possessed of a D.P. distribution such that the average D.P. is 95 and the portion with D.P. in excess of 110 accounts for 37 percent of the whole composition was trially smoked by 50 panel members. In this case, pungence was perceived by 46 panel members (92 percent) and it was not perceived by 4 panel members (8 percent). As is evident from the results just mentioned, the substitute tobacco material of the present invention comprises a degraded cellulose which is obtained by decomposing cellulose and is possessed of a D.P. distribution such that the average D.P. does not exceed 95 and the portion with D.P. in excess of 110 accounts for not more than 10 percent.

Table 1

Average D.P. of degraded cellulose and result of panel test

Smoke & taste Average D.P.	(%) Portion with D.P. in excess of 110 (%)	(A) No pungence perceived	(B) Pungence perceived slightly	(C) Pungence perceived
610	—	0	0	100
315	—	0	0	100
167	—	7	17	76
117	65	35	34	31
110	56	62	28	10
102	37	81	18	1
95	10.5	97	3	0
87	5.0	100	0	0
73	1.5	100	0	0
55	0.5	100	0	0

(Note) o Average D.P.: Calculated according to the formula described above.
o Smoke and flavor (%): The percentage represents the ratio of the number of panel members whose perception of pungence was as given in (A), (B) or (C) to the total of 50 panel members who participated in the panel test on smoke and on flavor respectively.

The degraded cellulose to be formed according to this invention is desired to have a D.P. distribution such that the portion with D.P. in excess of 110 accounts for not more than 10 percent of the whole composition. If a degraded cellulose having an average D.P. of 60 is mixed with 10 percent of a degraded cellulose having an average D.P. of 300, then the resultant mixture will have an arithmetic mean D.P. of 84. Yet, this mixture gives forth a pungent odor similar to that which is generated when a common cellulose is burnt. If a degraded cellulose having an average D.P. of 30 is mixed with 15 percent of a degraded cellulose having an average D.P. of 400, the resultant mixture will have an arithmetic mean D.P. of 85.5. Similarly to the former case, this mixture generates a pungent odor. Accordingly, the degraded cellulose satisfactory for the purpose of this invention is not simply required to have a D.P. distribution such that it will give an arithmetic mean D.P. of not more than 95. Also for this reason, one cannot be satisfied only because a given degraded cellulose has an average D.P. of not more than 95. Rather one must decompose common cellulose having higher D.P. so that the average D.P. as calculated according to the aforementioned formula will have a value of not more than 95.

The procedure to be followed for controlling the D.P. distribution of the degraded cellulose will be described in detail hereinafter.

Method for Manufacture of Degraded Cellulose

Cellulose such as, for example, a pulp prepared by a known method for the production of viscose rayon must be degraded so that it will acquire an average D.P. of not more than 95. This degradation is accomplished by steeping the cellulose in the aqueous solution of an alkali to obtain alkali cellulose, oxidatively degrading the alkali cellulose at a temperature not exceeding 100°C, thereafter neutralizing the degraded cellulose, washing with water and drying. The purpose of controlling the portion with D.P. in excess of 110 to within 10 percent of the whole composition is achieved by crushing the alkali cellulose as finely as possible in the course of the said oxidative degradation and allowing the cel-

lulose to come into uniform contact with the oxidizing atmosphere at a fixed temperature.

When the direct oxidative degradation of cellulose is attempted by means of a catalyst or heat instead of the said steeping in the alkali, it becomes difficult to control the average D.P. of the resultant degraded cellulose to a desired value. Further, such treatment is liable to entail uncalled-for secondary reactions, such as reaction of C_6 methylol group in the glucose unit of which the cellulose is composed or reaction resulting in the cleavage of pyranose ring composed of glucose units. Bacause of the formation of by-products, the resultant degraded cellulose may involve the danger of generating a smoke which is harmful to the human system. To facilitate the control of the average D.P. of the degraded cellulose and preclude possible secondary reactions, therefore, it is desirable that the oxidative degradation should be carried out after the cellulose has been steeped in an alkali to become alkali cellulose.

Raw Material

Cellulose of any origin may be used as the starting material for the product of the present invention. Examples are natural celluloses originating in wood pulp, cotton linter, ramie fiber and various kinds of paper and regenerated celluloses such as of viscose process and cuprammonium process.

Among these various kinds of celluloses, particularly desirable is a linear regenerated cellulose. The final product obtained from the cellulose is intended for the same use as tobacco. In this respect, therefore, the cellulose is required to have a shape which resembles that of shredded natural tobacco leaves and which permits ready packing particularly for the production of cigarettes. This requirement can be thoroughly fulfilled when the regenerated cellulose is a linear product comprising monofilaments each having a fineness of between 250 and 10,000 deniers.

If the monofilaments are coarser than 10,000 deniers, then the end product will become coarser and rigider than is required and, as such, will fail to suit the intended use as tobacco. If the monofilaments are finer than 250 deniers, the end product will tend to undergo further size reduction to the extent that the work of packing for the production of cigarettes will become difficult. Thus, monofilaments of the said linear regenerated cellulose may have a circular, flat, amorphous or any other shape of cross section so far as they have a fineness in the range of between 250 and 10,000 deniers. The linear regenerated cellulose may comprise one or any larger number of monofilaments. The monofilaments of the cellulose may be of a tubular shape, with bubbles contained therein. The liner cellulose may be used in its long, unbroken shape or in a shape cut to a suitable length. This linear product may be a regenerated cellulose produced by either the viscose process or the cuprammonium process.

Steeping in Alakli Solution (hereinafter referred to briefly as "steeping")

The cellulose as the starting material is converted to alkali cellulose by steeping. For the purpose of this steeping, there may be used either sodium hydroxide or potassium hydroxide. Economically it is more advantageous to use sodium hydroxide where the steeping is carried out on a commercial scale. The conditions such as the concentration of the aqueous solution of alkali to be used for this steeping, the temperature at which the steeping is made and the length of steeping time may be freely selected within the range in which the required conversion of cellulose to alkali cellulose can be accomplished. For example, the concentration of sodium hydroxide is from 8 to 24 percent, the temperature is variable between 0° and 80°C depending on the said concentration, and the steeping time is more than five minutes. As regards the manner of steeping, the given cellulose may be steeped in its original form of pulp, paper or sheet in the alkali solution. Otherwise, it may be agitated in the aqueous solution of the alkali so as to be treated in the form of slurry. Thus, the manner in which the steeping is performed is not specifically limited.

Oxidative Decomposition (or Aging)

In consequence of the preceding steeping, there is obtained an alkali cellulose which is impregnated with the aqueous solution of alkali and which is in the form of sheet, fiber or slurry. This alkali cellulose is squeezed until the weight is lowered to about three times the original weight. It is then subjected to oxidative decomposition preferably subsequent to being finely comminuted. The alkali cellulose may otherwise be subjected in its unaltered form of sheet or slurry to oxidative decomposition without being squeezed.

The conditions under which the oxidative decomposition of alkali cellulose is carried out may be suitably selected by taking into due consideration such factors as device to be used, kind of starting material, D.P. of cellulose, form of alkali cellulose, etc.

To accomplish this oxidative decomposition or aging on a commercial scale while controlling the D.P. distribution as mentioned above, it is necessary that the alkali cellulose should be finely comminuted and that a prescribed amount of alkali cellulose should be maintained at a substantially uniform temperature throughout and allowed to be held in substantially uniform contact with oxygen and/or air. When pulp or paper in the form of sheet is used as the starting material, converted to alkali cellulose by steeping and, in its original form without being finely comminuted, subjected to the aging, then the temperature differs and the condition of contact with oxygen and/or air varies between the middle portion and the peripheral portion of the sheetformed alkali cellulose or between the inner zone and the surface zone in one same portion of the sheet. If this occurs at all, there is a possibility that it will become difficult to acquire a degraded cellulose whose D.P. distribution is such that the portion with D.P. in excess of 110 accounts for not more than 10 percent of the whole composition. The said alkali cellulose, therefore, is desired to be finely comminuted as far as permissible. Fine comminution of the alkali cellulose may be accomplished by using Werner system crusher, Eirich crusher and other similar machines which are employed in the production of viscose rayon, for example. Preferably, the degree of crushing may be fixed on the order of 50 to 160 g/lit, for example.

The mass of alkali cellulose thus comminuted finely is maintained at a substantially uniform temperature and, at the same time, held in substantially uniform contact with oxygen and/or air. This temperature may be selected from the range not exceeding 100°C, preferably from the range between normal room temperature and 70°C. For the purpose of contact of the mass of alkali cellulose with oxygen and/or air, the oxygen may be used in a form diluted with an inert gas such as nitrogen or mixed with air. Instead of using oxygen in

a form mixed with air, there may be used air in its original form. In any case, it is essential that the said mass of alkali cellulose should be maintained at a temperature as uniform as permissible and brought into a uniform contact with the oxygen-containing gas as far as practicable. This requirement can be fulfilled by employing a vertical-column type aging device, a tunnel type aging device, a Werner crusher type aging device, a rotary kiln or some other suitable device which is used in the production of viscose rayon. It is not desirable to use a closed aging car or aging can which is employed in the production of viscose rayon, for such device fails to maintain the said mass of alkali cellulose at a uniform temperature or in uniform contact with oxygen and/or air.

A finely comminuted alkali cellulose obtained from a wood pulp having an average D.P. 610 was subjected to aging for 96 hours at 42°C within a column type aging device filled with a gas having 40 percent oxygen concentration. The product of this aging was then neutralized with an acid. The degraded cellulose thus obtained was found to have a D.P. distribution such that the average D.P. was 94 and the portion with D.P. in excess of 110 accounted for 6.3 percent by weight of the whole composition. The same finely comminuted alkali cellulose as mentioned above was placed in an aging can for use in the production of rayon yarn and, with a cover secured to the can, left to stand for 20 days in an aging room maintained at 30°C and relative humidity of 80 percent so as to undergo aging. The product of this aging was then neutralized with an acid. The degraded cellulose thus obtained was found to have a D.P. distribution such that the average D.P. was 95 and the portion with D.P. in excess of 110 accounted for as much as 37 percent of the whole composition.

The length of contact time should be selected suitably to suit such factors as D.P. of starting material, device to be used, temperature at which the oxidation is performed, and concentration of oxygen. In the case of an alkali cellulose produced by using as its starting material a rayon yarn grade pulp having an average D.P. of 600 to 800, for example, the alkali cellulose is prepared in a form containing the aqueous solution of alkali of a weight about twice as large as the original weight of cellulose and sent through an Eirich crusher so as to be crushed to the level of about 150 g/lit. Then, the finely crushed alkali cellulose is subjected to aging at 40° to 45°C within a column type aging device designed for the production of rayon yarn, with the oxygen concentration maintained in the range of 38 to 45 percent inside the device. After this aging is continued for more than 95 hours under the conditions mentioned above, there is obtained a degraded cellulose whose D.P. distribution is such that the average D.P. does not exceed 95 and the portion with D.P. in excess of 110 accounts for not more than 10 percent by weight of the whole composition.

Neutralization

The alkali cellulose having an average D.P. not exceeding 95 which has been obtained through the preceding processes is now neutralized with an acid. It may otherwise be xanthated with carbon disulfide and dissolved in the aqueous solution of alkali and thereafter subjected to neutralization with an acid.

For the purpose of the neutralization by the former method, there may be used an inorganic mineral acid such as, for example, sulfuric acid, hydrochloric acid or phosphoric acid. The degraded cellulose to be obtained after neutralization is desired to be washed thoroughly with water to ensure complete elimination of residual acid before it is dried. The product obtained at this stage is a white powdery or fibrous cellulose having a fiber length of 0.1 to 0.5 mm. Desirably, the molding agent for this product may be used afterward.

In the neutralization by the latter method, the alkali cellulose is xanthated and dissolved by using carbon disulfide and the aqueous solution of alkali. The said alkali cellulose may be xanthated with a required amount of carbon disulfide and thereafter dissolved in the aqueous solution of alkali. Otherwise, xanthation and dissolution of the said alkali cellulose may be accomplished simultaneously by allowing the said alkali cellulose to form a three-component system with carbon disulfide and the aqueous solution of alkali from the beginning. In either of the two procedures mentioned above, the amount of carbon sulfide to be used and the condition of xanthation may freely be selected within the range in which the said alkali cellulose is xanthated to such extent as to permit sufficient dissolution by the aqueous solution of alkali. The vessel to be used for the xanthation may be of any of the known shapes. In either of the two procedures mentioned above, the aqueous solution of alkali to be used for the purpose of dissolution of the alkali cellulose may have any concentration and temperature so far as the alkali is identical with the alkali combined to cellulose and the alkali solution is capable of dissolving the xanthation product completely. The vessel to be employed for this dissolution may be of any of the known shapes. The concentration of the dissolved solution may be selected freely within the range in which the xanthation product can be dissolved completely. For example, the concentration may be selected so that the solution obtained after dissolution will have a degraded cellulose concentration of 5 to 25 percent by weight. The dissolved solution is regenerated with an acid. This regeneration may be carried out by using any one of the known acids such as, for example, sulfuric acid, hydrochloric acid and phosphoric acid. The acid concentration and other regeneration conditions may be selected from among those already known to the art. In the said acid, a salt having the same acid radical as that acid may suitably be admixed. The product obtained after the process of regeneration may be refined and dried in accordance with an ordinary method. In the latter method, a flake-shaped product is obtained without having to use any special molding agent.

Method of Molding of Degraded Cellulose

The degraded cellulose obtained through the preceding processes has a D.P. distribution such that the average D.P. does not exceed 95 and the portion with D.P. in excess of 110 desirably accounts for not more than 10% of the whole composition. As a substitute tobacco material which generates a smoke free from pungence, this degraded cellulose may be used by itself in an unmodified form. Otherwise, it may be used as an extender or a carrier for tobacco products. The product obtained has an average D.P. not exceeding 95 and is composed of relatively short fibers having a length of 0.1–0.5 mm. It is, therefore, not easy to have this product molded in any desired form such as that of flakes or films. No molding is specifically required, however, in case the starting material happens to be a fibrous

substance comprising monofilaments having a fineness in the range between 250 and 10,000 deniers.

In order that the degraded cellulose may be molded in any given form suitable for the intended purpose of the end product without impairing its outstanding property of generating neither smoke nor odor contributing to the irritation of the mucous membranes of the nose and the throat, it is essential that a degraded cellulose having a D.P. distribution such that the average D.P. does not exceed 95 and the portion with D.P. in excess of 110 accounts for not more than 10 percent of the whole composition should be contained in an amount of 75 percent or more by weight and that, as a specific molding agent, there must be used a cellulose molding material which is composed of one member or a mixture of two or more members selected from the group consisting of sodium cellulose gylcolate, hydroxyethyl cellulose, methyl cellulose and ethyl cellulose. The resultant mixture may be molded in any desired shape to suit the intended use.

According to the present invention, the molded product of cellulose is required to contain 75 percent or more by weight of degraded cellulose which has an average D.P. below 95. If the content of degraded cellulose is smaller than 75 percent by weight, then the molded product has a disadvantage that it will generate an objectionable odor peculiar to the molding agent incorporated therein. Although this content is only required to exceed 75 percent by weight, it is nonetheless desired to fall in the range of between 75 and 99 percent in order that the molded product may enjoy good results of molding.

The substances which are generally used as molding agents are numerous. They include such cellulose derivatives as sodium cellulose xanthogenate, nitrocellulose, cellulose acetate and cellulose propionate, such synthetic high molecular compounds as polyvinyl alcohol, polyvinyl methyl ether, polyacrylamide, and polyalkylene glycols, and such natural compounds as starch, pectin, gum arabic, tragacanth gum, caraja gum, dextrine, alginates, casein and gelatin. Sodium cellulose xanthogenate, nitrocellulose, cellulose acetate and cellulose propionate not merely generate an objectionable odor upon combustion. They also has a possibility of generating a smoke containing sulfur, nitrogen and decomposition products of acetic acid and other organic acid which are harmful to the human system. They are, therefore, unsuitable as molding agents for the molded product of cellulose according to the present invention. In addition, polyvinyl alcohol, polyvinyl methyl ether, polyacrylamide and polyalkylene glycols are similarly unsuitable because they not only generate a highly irritating smoke upon combustion but also produce, in certain cases, gases which are noxious to the human system. Such natural products as starch, pectin, gum arabic, tragacanth gum, caraja gum, dextrin, alginates, casein and gelatin can hardly accomplish the object, because the molded product of the said degraded cellulose generates a highly irritating smoke and an objectionable odor when it incorporates such naturally occurring molding agent even in a very small amount.

Sodium cellulose glycolate to be used for the present purpose is desired to be such that the viscosity of 1 percent solution thereof at 25°C falls in the range of between 80 and 1800 centipoises and the D.S. (degree of substitution) of carboxymethoxyl group thereof falls in the range of between 0.6 and 1.0. Similarly, hydroxyethyl cellulose is desired to be such that the viscosity of 2 percent aqueous solution thereof at 20°C falls in the range of between 70 and 4,000 centipoises. Methyl cellulose is desired to be such that the viscosity of 2 percent aqueous solution thereof at 20°C falls in the range of between 500 and 6,000 centipoises and the D.S. (of methoxyl group) thereof falls in the range of between 1.6 and 2.0. Ethyl cellulose is desired to be such that the D.S. (of ethoxyl group) thereof falls in the range of between 0.6 and 1.5. When the said viscosity or D.S. is smaller than the lower limit of the said range, the degraded cellulose cannot be molded as required. If it is greater than the upper limit of the said range, then the molded product of degraded cellulose becomes insoluble in water and difficult of burning. Moreover, the smoke generated upon combustion acquires increased pungence.

The degraded cellulose is thoroughly dispersed in water. To the resultant dispersed system, the aforementioned cellulose derivative is added at a prescribed proportion and then mixed to form a homogeneous dispersion. This homogeneous dispersion is indispensable for obtaining a homogeneous molded product to be obtained thereafter. An attempt to place the cellulose derivative first in water and add the degraded cellulose to the resultant solution proves ineffective, because the aqueous solution of cellulose derivative has too high a viscosity to afford a homogeneously dispersed system of degraded cellulose.

The dispersed system comprising the said degraded cellulose, sodium cellulose glycolate and water is so prepared that the ratio of the combined weight of the first two compounds to the weight of water falls in the range of between 1:12 and 1:6. In case where hydroxyethyl cellulose, methyl cellulose or ethyl cellulose is used in place of sodium cellulose glycolate, the said mixing ratio is changed to the range of between 1:14 and 1:7, between 1:35 and 1:6 and between 1:35 and 1:6 respectively. Where two or more cellulose derivatives are put to use in a mixed form, the said mixing ratio may be suitably corrected by taking into account the mixing ratio assigned to each cellulose derivative. When the mixing ratio falls outside the specified range, it will become difficult to obtain a uniform product in a desired shape.

The dispersed system thus prepared is molded in the shape of a thread, plate, film or some other desired shape to suit the intended use according to the ordinary method and thereafter dried. The molded product may be mixed with natural leaf tobacco to prepare a tobacco product having a small micotine content and a light taste. The molded product obtained in the shape of thread or film may be shredded and, with necessary flavor and other additives incorporated, used to make a man-made smoking material. The molded product may be obtained in the shape of a very thin film so as to be used in place of a tubular paper enclosure wound around a cigarette.

METHOD for Manufacture of Tobacco Mixtures

Method I

The dispersed system obtained through the preceding processes is now molded in the shape of a sheet having a desired thickness on the order of 0.5 mm, for example. While the formed sheet is still in a wet condition, tobacco dust is caused to adhere as uniformly to the

sheet as possible by a known method. The sheet which now has tobacco dust deposited thereon in a uniform thickness is dried by an ordinary method. Consequently, there is obtained a laminar sheet-shaped tobacco about 0.1 mm in thickness, having tobacco dust deposited evenly on a sheet formed with the said degraded cellulose and sodium cellulose glycolate. The tobacco dust to be used for this purpose may be derived from scraps of leaf tobacco, scraps of shredding, midribs of tobacco leaves, stems of tobacco plants and whatever sources capable of providing tobacco dust. The laminar sheet-shaped product may be obtained in the construction of a sandwich having a layer of tobacco dust deposited on each side of the sheet by spreading the aforementioned tobacco dust in advance on a flat surface, allowing the dispersed system in the form of slurry to be molded in the shape of a sheet over the layer of tobacco dust and depositing tobacco dust on the upper surface of the molded sheet. The tobacco dust deposited on the surface of the molded sheet may be pressed fast such as by means of rollers to ensure thorough adhesion. At this time, compression of the tobacco dust and drying of the laminar sheet-shaped product can be accomplished simultaneously by heating the compression rollers.

Method II

Tobacco dust may be mixed with the slurry formed of the degraded cellulose and the molding agent and the resultant slurry or paste may be molded in the shape of a sheet. This mixture in the form of slurry or paste may be obtained by adding the tobacco dust to the slurry of degraded cellulose plus molding agent and blending them into slurry or paste. It may otherwise be obtained by first wetting the tobacco dust, blending the wetted tobacco dust into paste, adding the paste into the said slurry of degraded cellulose plus molding agent and blending them into slurry or paste. Such tobacco mixture may also be obtained by first allowing the said degraded cellulose and tobacco dust to be dispersed in water and blended into paste, adding the said cellulose derivative to the paste under the aforementioned conditions and blending them until there is formed slurry or paste. This mixture in the form of slurry or paste is molded to the shape of a film by a known method of extrusion using a slit and an extruder and then dried by an ordinary method.

In any of the methods mentioned above, the tobacco dust content may be freely varied by adjusting the thickness of the sheet or by adjusting the amount of tobacco dust to be deposited on the sheet.

The laminar product thus obtained may be shredded so as to be used for the production of cigarettes or pipe tobacco. It may also be used as a cigar when it is rolled up and bound, with the whole encased or not encased in a natural tobacco leaf.

In any case, tars and nicotine which are contained in the smoke of tobacco and which are harmful to the human system can be lessened by substituting the laminar product for as much natural tobacco leaf as possible. Incorporation of the laminar product also can render the flavor and taste of tobacco light and mild without impairing the flavor and taste inherent to tobacco.

In the course of the manufacture of sheet or in the course of the deposition of tobacco dust to the sheet, known flavors and other additives such as combustion promoter, ash improver, humectant and methanol may be added to the slurry or paste formed of degraded cellulose and molding agent.

The present invention is described in further detail with reference to preferred embodiments to be cited hereinafter.

EXAMPLE 1:

A. In a tank provided with an agitation means and containing 17.5 percent aqueous solution of sodium hydroxide, 1 kg of wood pulp having an average D.P. of 610 and an α-cellulose content of 93.5 percent was agitated in the state of 4 percent slurry for 16 minutes. By squeezing the treated pulp with a device using a vacuum filter and a press roll and subsequently crushing the squeezed pulp with an Eirich type crusher, there was obtained 2.6 kg of alkali cellulose having a cellulose concentration of 32.5 percent and a sodium hydroxide concentration of 15.6 percent. The alkali cellulose was exposed to air containing 40 percent of oxygen at 42°C for 96 hours and then weakly acidified with an excess amount of 20 percent sulfuric acid. Thereafter, it was thoroughly neutralized by being washed in flowing water for one hour. The neutralized cellulose was dried in an electric drier at 60°C. Consequently, there was obtained 890 g of white degraded cellulose in the form of short fibers about 0.3 mm in length. This degraded cellulose was found to have a D.P. distribution such that the average D.P. was 93 and the portion with D.P. in excess of 110 accounted for 6.3% of the whole composition.

B. Man-made cigarettes each weighing about 0.9 g were prepared by forming a part of the degraded cellulose in the shape of rolls and enclosing the rolls in roll paper used for the production of ordinary cigarettes. These cigarettes were trially smoked by about 50 smokers. Of these many smokers, 97 percent gave a rating that both main stream and side stream of smoke from these cigarettes were free from pungence and were nearly tasteless and odorless.

The said degraded cellulose and natural leaf tobacco were mixed at a weight ratio of 33:67. Cigarettes each weighing 0.85 g were prepared by forming the mixture in the shape of rolls and enclosing these rolls in roll paper of the kind used generally for the production of ordinary cigarettes. When the cigarettes were subjected to the similar smoking test, it was found that both main stream and side stream of the smoke were considerably milder and the taste of smoking was milder than ordinary cigarettes formed solely of finely cut natural leaf tobacco.

When the same smoking test was performed on cigarettes prepared by using a degraded cellulose having an average D.P. of 162 which had been obtained by repeating the aforesaid method, excepting that the exposure to air containing 40 percent of oxygen was made at 42°C for 24 hours, only 7 percent of the smokers answered that both main stream of smoke were free from pungence and were nearly tasteless and odorless. An answer that the taste of smoking was rather unpleasant, though main stream and side stream of smoke were considerably alleviated of pungence was given by 17 percent of the smokers. The remaining 76 percent of the smokers answered that the taste of smoking as well as main stream and side stream of smoke was objectionable.

EXAMPLE 2:

A. The amount 940 g of a white degraded cellulose of the form of short fibers having a D.P. distribution such that the average D.P. was 88 and the portion with D.P. in excess of 110 accounted for 4.7 percent of the whole composition was obtained by repeating the procedure of Example 1, with the following exceptions:

Raw material: 1 kg of refined cotton linter having an average D.P. of 820 and an α-cellulose content of 98 percent.

Alkali cellulose obtained consequently: Cellulose concentration 32.2 percent and sodium hydroxide concentration 15.5 percent.

Oxidation: Oxidation carried out in a tower type aging device for 103 hours instead of 96 hours.

Neutralizing agent: 13% hydrochloric acid.

B. Cigarettes each weighing about 0.9 g were prepared by forming a part of the resultant degraded cellulose in the shape of rolls and enclosing the rolls in roll paper of the kind generally used for the production of ordinary cigarettes. When these cigarettes were trially smoked by about 50 smokers, it was learnt that both smoke and taste were free from pungence and were nearly tasteless and odorless.

EXAMPLE 3:

A. In a slurry steeper, 600 g of wood pulp having an average D.P. of 620 and an α-cellulose content of 91.5% were steeped for about 20 minutes in 17.5% aqueous solution of sodium hydroxide at 53°C. The steeped pulp was squeezed to a weight 2.8 times the original weight and then crushed. Consequently there was obtained 1.6 kg of alkali cellulose having a cellulose concentration of 32.2 percent and a sodium hydroxide concentration of 15.5 percent. This alkali cellulose was subjected to aging by a tower type aging process at 40°C for 120 hours, with oxygen concentration fixed at 38 percent. The aging gave the cellulose a D.P. distribution such that the average D.P. was about 95 and the portion with D.P. in excess of 110 accounted for 6.0 percent of the whole composition. The alkali cellulose is then transferred into a small churn. The churn was vacuumized and 140 cc of carbon disulfide was received in the churn, causing the alkali cellulose to be xanthated at 28°C for 90 minutes. Consequently, there was obtained about 1.8 kg of a yellowish orange product. This product was added to 3.8 kg of 2.63 percent aqueous solution of sodium hydroxide and mixed therewith by agitation at 18°C for 1 hour. Consequently, there was obtained a yellowish orange, slightly viscous aqueous solution having a degraded cellulose concentration of 8.7 percent and a sodium hydroxide concentration of 6.2 percent. The said aqueous solution was formed in a layer having a thickness of about 1 mm by using an applicator. The layer was immersed in 20 percent sulfuric acid at 20°C for about five minutes, allowing the layer to be thoroughly regenerated and converted into a film. The film was thoroughly washed with water and dried at 105°C for 1 hour. Analysis showed this film to be a degraded cellulose having an average D.P. of 82 and containing a β-glucoside linkage. It was white and was relatively easily broken into small strips.

B. Cigarettes each weighing about 0.9 g were prepared by forming small strips of the said degraded cellulose in the shape of rolls and enclosing the rolls in roll paper of the kind generally used for the production of ordinary cigarettes. These cigarettes were trially smoked by about 50 smokers. The results of the test led to a conclusion that they burnt smoothly without producing a yellow flame of the kind frequently observed in the combustion of cellulose and that both main stream and side stream of the smoke were free from pungence and were nearly tasteless and odorless.

The procedure mentioned above was repeated, except that the aging was carried out for 24 hours. The degraded cellulose thus obtained was found to have an average D.P. of 175. Cigarettes prepared of this degraded cellulose by repeating the procedure of Example 1 were subjected to the same smoking test as in Example 1. Only 5 percent of the smokers answered that both main stream and side stream of smoke were free from pungence and were nearly tasteless and odorless. The answer by 19 percent of the smokers was that the taste of smoking was rather objectionable, though both main stream and side stream of the smoke were considerably alleviated of pungence. The remaining 76 percent of the smokers gave the answer that the taste of smoking as well as main stream and side stream of smoke was unpleasant.

EXAMPLE 4:

A film of degraded cellulose which had a D.P. distribution such that the average D.P. was 76 and the portion with D.P. in excess of 110 accounted for 0.8 percent of the whole composition and which was white and was relatively easily broken into small strips was obtained by repeating the procedure of Example 3-A, with the following exceptions:

Raw material: 600 g of refined cotton linter having an average D.P. of 815 and an α-cellulose content of 97.3%.

Alkali cellulose obtained consequently: Cellulose concentration of 32.5 percent and sodium hydroxide concentration of 15.5 percent.

Aging: The aging was carried out for 200 hours instead of 120 hours to give the cellulose an average D.P. of about 89.

Sulfurization: The sulfurization was carried out with 180 cc of carbon disulfide at 30°C for 90 minutes.

Fine strips of the said degraded cellulose were mixed in an amount of 23 percent by weight ratio to shredded tobacco available on the market. Cigarettes each weighing about 0.85 g were prepared by forming the resultant mixture in the shape of rolls and enclosing the rolls in roll paper of the kind generally used for the production of ordinary cigarettes. When these cigarettes were trially smoked in the same manner as in Example 3B, none of the smokers felt pungence in the smoke and the taste of smoking.

EXAMPLE 5:

A skein of 4000 denier viscose rayon monofilaments having a flat cross section was steeped in an excess amount of 17.5 percent aqueous solution of sodium hydroxide at 20°C. The steeped skein was squeezed by a mangle to obtain a skein of alkali cellulose monofilaments having a weight about three times the original weight. This skein was exposed to air having an oxygen concentration of 40 percent at 42°C for 85 hours and then weakly acidified with an excess amount of 20 percent sulfuric acid, thoroughly neutralized by being

washed in flowing water for 30 minutes and finally dried by an ordinary method.

The product thus obtained was found to have a D.P. distribution such that the average D.P. was 96 and the portion with D.P. in excess of 110 accounted for 7.5 percent of the whole composition. Cigarettes each weighing about 0.8 g were prepared by cutting the filaments of the product to a length of about 5 cm, forming the cut filaments in the shape of rolls and enclosing the rolls in roll paper of the kind generally used for the production of ordinary cigarettes. When these cigarettes were trially smoked by about 50 smokers, the results of the test led to a conclusion that both main stream and side stream of smoke were free from pungence and were nearly tasteless and odorless.

EXAMPLE 6:

A degraded cellulose having a D.P. distribution such that the average D.P. was 90 and the portion with D.P. in excess of 110 accounted for 4.9 percent of the whole composition was obtained by repeating the procedure of Example 5, with the following exceptions:

Raw material: A skein of hollow viscose rayon yarns composed of 15 unit monofilaments and having an overall fiber size of 6,000 deniers.

The filaments of the treated skein were cut to a length of about 3 cm. By the use of smoking pipes, the cut filaments were trially smoked by about 50 smokers in the same manner as in Example 5. The results of this test led to a conclusion that both main stream and side stream of smoke were free from pungence and were nearly tasteless and odorless.

When the same procedure was followed except that the length of time of the exposure to air containing oxygen was decreased to 16 hours, the product was found to have an average D.P. of 173. The resultant filaments of the skein were trially smoked in the same manner as mentioned above, only 8 percent of the smokers answered that both main stream and side stream of smoke were free from pungence and were nearly tasteless and odorless. The answer by 22 percent of the smokers was that the taste of smoking was rather objectionable, although main stream and side stream of smoke were considerably alleviated of pungence. The remaining 70 percent of the smokers gave the answer that the taste of smoking as well as main stream and side stream of smoke was objectionable.

EXAMPLE 7:

A. A white degraded cellulose of short fibers about 0.2 mm in length having a D.P. distribution such that the average D.P. was 74 and the portion with D.P. in excess of 110 accounted for 0.6 percent of the whole composition was obtained by repeating the procedure of Example 1A, with the following exceptions:

Raw material: 1 kg of tissue paper having an average D.P. of 780 and an α-cellulose content of 85 percent.

Steeping: The raw material was steeped for 20 minutes in the state of 4 percent slurry at 48°C in 18% aqueous solution of potassium hydroxide.

Alkali cellulose obtained consequently: Cellulose concentration of 32.5 percent and potassium hydroxide concentration of 21.5 percent.

Aging: The aging was carried out at 40°C for 200 hours, with oxygen concentration fixed at 38%. Neutralization: 20% sulfuric acid.

B. In a household electric mixer, 90 g of the said degraded celllulose and 930 cc of distilled water were agitated at the rate of 5,900 rpm. While the mixture was being thus agitated, 15 g of powdered sodium cellulose glycolate which had a D.S. of 0.9 and whose 1 percent aqueous solution manifested a viscosity of 1200 poises at 25°C was added gradually to the mixture. Subsequently, 20 g of powdered potassium citrate as combustion promoter, 20 g of powdered calcium carbonate as ash improver and 10 g of glycerin as humidity regulator were added. By continuing the agitation at about 65°C for 25 minutes, there was obtained about 1 kg of a white, viscous mashy slurry.

C. This slurry was formed in the shape of a film having a thickness of 1 mm by using an applicator. The film was dried for 60 minutes in an electric drier at 105°C. Consequently, there was obtained a white dry film having a thickness of 0.08 mm.

D. This film was shredded to strips having a width of about 1 mm. Cigarettes each weighing about 0.9 g were prepared by forming the shredded strips in the shape of rolls and enclosing the rolls in roll paper of the kind generally used for the production of ordinary cigarettes. When these cigarettes were subjected to the same smoking test as described in Example 1, it was learnt that both main stream and side stream of smoke were free from pungence and were nearly tasteless and odorless.

EXAMPLE 8:

A. About 900 g of white degraded cellulose of the shape of short filaments having a D.P. distribution such that the average D.P. was 78 and the portion with D.P. in excess of 110 accounted for 2.0 percent of the whole composition was obtained by repeating the procedure of Example 8A, with the following exceptions:

Raw material: 1 kg of wood pulp having an average D.P. of 636 and an α-cellulose content of 93.7 percent.

Steeping: The raw material was steeped in the state of 3.8 percent slurry for 21 minutes in 17.8 percent aqueous solution of sodium hydroxide at 50°C.

Compression and comminution: The steeped pulp was treated on a compression unit using a vacuum filter and a press roll and then crushed by a garnet wire type crusher to afford 2.5 kg of alkali cellulose. This alkali cellulose was found to have a cellulose concentration of 32.8 percent and a total alkali concentration of 15.6 percent.

Aging: The aging was carried out for 185 hours in the atmosphere having an oxygen concentration of 44 percent at 40°C.

Neutralization: The neutralization was effected by using 10% hydrochloric acid.

B. About 1.5 kg of white, mashy slurry was obtained by treating the resultant degraded cellulose by following the procedure of Example 7B, with the following exceptions: Raw material: 140 g of degraded cellulose and 1,350 cc of deionized water.

Mixer: The agitation was carried out at the rate of 6,500 rpm.

Additive used: 10 g of powdered sodium cellulose glycolate having a D.S. of 1.0, whose 1 percent aqueous solution at 25°C exhibited a viscosity of 1,700 centipoises.

Operating conditions after incorporation of additive: The mixture was agitated for 30 minutes at 65°C.

C. Treatment of the said slurry: The treatment of Example 7C was repeated, with the exception that the degraded cellulose was mixed with the shredded tobacco available on the market at a ratio of 1:3.

Cigarettes were prepared by forming the mixture in the shape of rolls and enclosing the rolls in roll paper of the kind generally used for the production of ordinary cigarettes. When they were trially smoked by about 50 smokers, practically all the smokers gave a rating that the taste became lighter. None of the smokers felt pungence in the smoke and the odor.

EXAMPLE 9:

About 1.5 kg of white, mashy slurry was obtained by repeating the procedure of Example 7B, with the following exceptions:

Raw material: 100 g of degraded cellulose obtained in Example 8A and 1,440 cc of deionized water.

Velocity of agitation: 6,500 rpm.

Additive used: 20 g of powdered sodium cellulose glycolate with D.S. 0.6 whose 1 percent aqueous solution at 25°C manifested a viscosity of 300 centipoises.

Operational conditions after incorporation of additive: The mixture was agitated for 20 minutes at 60°C.

A white thin film having a thickness of about 35 microns was obtained by treating the said slurry by following the procedure of Example 7C, with the exception that a film having a thickness of about 0.4 mm was dried at 70°C for 20 hours.

This film was shredded to strips of a suitable size. In place of roll paper generally used for the production of ordinary cigarettes, the strips of the film were wrapped around rolls of shredded tobacco available on the market to prepare cigarettes. When these cigarettes were trially smoked by about 50 smokers, 95 percent of the smokers gave a rating that the smoke had less pungence and the taste was lighter than cigarettes wrapped in roll paper.

EXAMPLE 10:

A. About 850 g of white degraded cellulose of the shape of short fibers about 0.2 mm in length having a D.P. distribution such that the average D.P. was 72 and the portion with D.P. in excess of 110 accounted for 0.4 percent of the whole composition was obtained by repeating the procedure of Example 8A, with the following exceptions:

Steeping: The raw material was steeped and agitated in the state of 4.0 percent slurry for 18 minutes in 17.5 percent aqueous solution of sodium hydroxide.

Alkali cellulose obtained consequently: The alkali cellulose obtained in the amount of 2.8 kg had a cellulose concentration of 32.5 percent and a total alkali concentration of 15.5 percent.

Aging: The aging was carried out for 200 hours in the atmosphere having an oxygen concentration of 42 percent instead of 44%.

Neutralization: The neutralization was effected by using 20% sulfuric acid.

B. About 1.1 kg of white, mashy slurry was obtained by repeating the procedure of Example 8B, with the following exceptions:

Raw material: 100 g of degraded cellulose and 1,040 cc of deionized water.

Additive used: 30 g of powdered sodium celuulose glycolate with D.S. 0.6 whose 1 percent aqueous solution at 25°C manifested a viscosity of 90 centipoises.

Operating conditions after incorporation of additive: The agitation was carried out at 50°C for 30 minutes.

C. The slurry mentioned above was formed in the shape of a film having a thickness of about 0.6 mm on a glass sheet by using an applicator. Tobacco dust passed through a sieve of 100-mesh size was immediately sprinkled on the film still in its wet state to form thereon a layer of tobacco dust of a substantially uniform thickness. The deposited tobacco dust was pressed down lightly with a rubber roller. Then, the wet film carrying therein the layer of tobacco dust was dried at 55°C in an electric drier by an ordinary method. The laminar sheet tobacco thus prepared was found to be composed of degraded cellulose, sodium cellulose glycolate and tobacco dust at an approximate weight ratio of 10:3:17. The sheet tobacco had a thickness of about 0.1 mm.

Cigarettes were prepared by shredding the laminar sheet tobacco into strips of a width of about 0.5–1 mm, forming the strips in the shape of rolls and enclosing the rolls in roll paper of the kind generally used for the production of ordinary cigarettes. When these cigarettes were trially smoked by about 50 smokers, it was learnt as a conclusion that they burnt as smoothly as ordinary cigarettes and the taste of smoking was very light and mild.

The procedure just mentioned was repeated, with the exception that the length of time of aging was shortened to 24 hours and the degraded cellulose used in this case had an average D.P. of 165. Cigarettes were prepared in the same manner as mentioned above and were trially smoked similarly. In this test, about 20 percent of the smokers reported to have perceived pungence in the smoke and in the taste of smoking. Five smokers had a fit of coughing.

EXAMPLE 11:

A. About 900 g of white degraded cellulose of the shape of short fibers about 0.3 mm in length having a D.P. distribution such that the average D.P. was 91 and the portion with D.P. in excess of 110 accounted for 5.3 percent of the whole composition was obtained by repeating the procedure of Example 2A, with the following exceptions:

Raw material: The average D.P. of the raw material was 830.

Steeping: The steeping was carried out in the state of 3 percent slurry at 30°C for 45 minutes.

Compression and comminution: The slurry was passed through a heat-exchanger for 15 minutes so as to be lowered to 51°C. It was then squeezed and subsequently crushed with a garnet wire type crusher.

Alkali cellulose obtained consequently: 2.9 kg of alkali cellulose having a cellulose concentration of 32.8 percent and a total alkali concentration of 17.7 percent.

Aging: The aging was carried out for 110 hours at 40°C in the atmosphere having an oxygen concentration of 43 percent.

Neutralization: The neutralization was effected by using 10 percent hydrochloric acid.

B. About 1.2 kg of white, mashy slurry was obtained by treating 140 g of the said degraded cellulose by repeating the procedure of Example 8B, with the following exceptions:

Amount of deionized water used: 1,050 cc.

Additives used: 20 g of potassium citrate as combustion promoter, 10 g of glycerin as humidity regulator.

Operating conditions after incorporation of additives: The mixture was agitated continuously for 40 minutes at 63°C.

C. A laminar sheet tobacco composed of degraded cellulose, sodium cellulose glycolate and tobacco dust at an approximate weight ratio of 14:1:12 was obtained by repeating the procedure of Example 10C, with the exception that the said slurry was first formed to the shape of a sheet having a thickness of about 0.4 mm.

Cigarettes were prepared by treating this laminar sheet tobacco by repeating the procedure of Example 10C. The cigarettes were trially smoked in the same manner as in Example 10C. The results of this test led to a conclusion that the condition of combustion was indistinguishably similar to that of ordinary cigarettes and the taste of smoking was very light and mild.

Another laminar sheet tobacco was obtained by repeating the same procedure as mentioned above, except that the amount of degraded cellulose used was decreased to 25 g. This sheet tobacco was composed of degraded cellulose, sodium cellulose glycolate and tobacco dust at an approximate weight ratio of 2.5:1:3 and had a thickness of about 0.09 mm. Cigarettes were prepared of this laminar sheet tobacco by following the same procedure as mentioned above. These cigarettes were subjected to smoking test by the same method. Consequently, 48 percent of the smokers perceived objectional odor in the smoke and complained of unpleasant sensation.

EXAMPLE 12:

A. About 830 g of white degraded cellulose of the shape of short fibers about 0.15 mm in length having a D.P. distribution such that the average D.P. was 61 and the portion with D.P. in excess of 110 accounted for a barely detectable fraction of the whole composition was obtained by repeating the procedure of Example 1A, with the following exceptions:

Raw material: Wood pulp of rayon grade having an average D.P. of 630.

Steeping: The steeping was carried out at 50°C.

Comminution: A garnet wire type crusher was used.

Alkali cellulose obtained consequently: Cellulose concentration 32.6 percent and total alkali concentration 15.7 percent.

Aging: The aging was carried out for 285 hours at 40°C in the atmosphere having an oxygen concentration of 42 percent.

Neutralization: The neutralization was effected with 10 percent hydrochloric acid.

B. One hundred (100) g of degraded cellulose and 130 g of tobacco dust passed through a 100-mesh sieve were placed in conjunction with 2,600 cc of deionized water in a general-purpose mixer and agitated at the rate of 4,500 rpm. While the agitation was in process, 30 g of powdered sodium cellulose glycolate having a D.S. of 0.6 whole 1% aqueous solution had a viscosity of 90 centipoises at 25°C was added gradually to the mixture being agitated. When this agitation was continued at 60°C for 50 minutes, there was obtained about 2.8 kg of yellowish brown, mashy slurry.

C. The slurry was formed in the shape of a sheet having a thickness of about 0.6 mm on a glass sheet by using an applicator. The sheet was dried at 60°C in an electric drier. Consequently, there was obtained a yellowish brown, dry sheet tobacco mixture having a thickness of about 0.09 mm.

This sheet tobacco mixture was shredded into strips of a width of about 0.5 to 1 mm. Cigarettes were prepared by forming the strips in the shape of rolls and enclosing the rolls in roll paper of the kind generally used for the production of ordinary cigarettes. These cigarettes were subjected to smoking test employing about 50 smokers. The conclusion drawn from the test was that the condition of combustion was practically the same as that of ordinary cigarettes and the taste of smoking was very light and mild.

Cigarettes were prepared by repeating the method just mentioned, with the exception that the length of time of the exposure of the said alkali cellulose to the oxygencontaining atmosphere was lessened to 40 hours and the degraded cellulose had an average D.P. of 136. When these cigarettes were trially smoked by the same method as mentioned above, about 18 percent of the smokers reported to have perceived pungence on the mucous membrane of the nose and the throat.

EXAMPLE 13:

A. A white degraded cellulose (I) of the shape of short fibers about 0.3 mm in length having a D.P. distribution such that the average D.P. was 92 and the portion with D.P. in excess of 110 accounted for 8.3 percent of the whole composition was obtained by repeating the procedure of Example 11A, with the following exceptions:

Steeping: The raw material was steeped in the state of 2.5 percent slurry in 18.1 percent aqueous solution of sodium hydroxide.

Temperature after passage through the heat-exchanger: 52°C.

Comminution: Crushing was effected by using an Eirich crusher.

Alkali cellulose obtained consequently: About 2.9 kg of alkali cellulose having a cellulose concentration of 32.5% and a total alkali concentration of 15.8 percent was obtained.

Aging: The aging was carried out in the atmosphere having an oxygen concentration of 41 percent.

Neutralization: The neutralization was effected by using 20% sulfuric acid.

B. A white mashy slurry was obtained by treating 140 g of the said degraded cellulose in accordance with the procedure of Example 7, with the following exceptions:

Raw water: Deionized water.

Rate of agitation: 6,500 rpm.

Additives used: 10 g of powdered sodium cellulose glycolate which had a D.S. value of 1.0 and whose 1 percent aqueous solution at 25°C had a viscosity of 1,700 centipoises, 15 g of potassium citrate and 10 g of citric acid each as combustion promoter, 10 g of sodium carbonate as ash improver and 10 g of glycerin as moisture regulator.

Agitation after incorporation of additives: The mixture was agitated at 55°C for 20 minutes.

Separately, 100 g of tobacco dust having a particle size smaller than 100 mesh was added to 160 cc of deionized water in a separate container and the mixture was agitated to produce a tobacco dust paste (II).

The tobacco dust paste was added to the aforesaid white mashy slurry. The resultant mixture was agitated at 55°C for 40 minutes. Consequently, there was obtained about 2.6 kg of yellowish brown slurry. This slurry was formed in the shape of a sheet with the aid of an applicator by following the procedure of Example

12. By drying this sheet, there was obtained a sheet tobacco mixture having a thickness of about 0.1 mm.

This sheet tobacco mixture was shredded into strips. Cigarettes were prepared of the strips by following the procedure of Example 12. They were then subjected to the same smoking test as mentioned in Example 12. The conclusion drawn from this test was that the condition of combustion was undistinguishably similar to that of ordinary cigarettes and the taste of smoking was very light and mild.

Another sheet tobacco mixture was prepared by repeating the procedure just mentioned, with the exception that the white mashy slurry was produced by using the degraded cellulose in a lessened amount of 25 g. When cigarettes prepared of this sheet tobacco mixture by the same procedure as mentioned above were subjected to the same smoking test, it was learnt that the smoke emitted unpleasant odor.

EXAMPLE 14:

A. About 930 g of white powdery degraded cellulose having a D.P. distribution such that the average D.P. was 60 and the portion with D.P. in excess of 110 accounted for barely detectable fraction of the whole composition was obtained by repeating the procedure of Example 13A, with the following exceptions:
Raw material: The average D.P. of the cotton linter was 825.
Steeping: The steeping was carried out for 60 minutes.
Heat exchange prior to squeezing: Omitted.
Crusher: A garnet wire type crusher was used.
Alkali cellulose obtained consequently: 2.7 kg of alkali cellulose having a cellulose concentration of 32.8 percent and a total alkali concentration of 15.5 percent.
Aging: The aging was carried out for 250 hours in the atmosphere having an oxygen concentration of 42 percent.
Neutralization: The neutralization was effected by using 10 percent hydrochloric acid.

B. About 1.1 kg of white mashy slurry was obtained by treating 90 g of the said degraded cellulose in accordance with the procedure of Example 7B, with the following exceptions:
Raw water: 990 cc of deionized water.
Rate of agitation: 6,500 rpm.
Additive used: 20 g of powdered hydroxyethyl cellulose whose 2% aqueous solution at 20°C had a viscosity of 4,000 centipoises.
Length of heating time subsequent to incorporation of additives: 30 minutes.

C. A dry white film having a thickness of about 0.09 mm was obtained by treating the white mashy slurry in accordance with the procedure of Example 7C.

This film was shredded to strips having a width of about 1 mm. The strips were mixed with shredded tobacco available on the market at the rate of 1:3. Cigarettes were prepared by forming the resulting mixture in the shape of rolls and enclosing the rolls with roll paper of the kind generally used for the production of ordinary cigarettes. These cigarettes were trially smoked by about 50 smokers. Consequently, nearly 100 percent of the smokers gave a rating that the taste became lighter. None of the smokers perceived pungence in the smoke and in the odor.

EXAMPLE 15:

About 1.7 kg of white, mashy slurry was obtained by repeating the procedure of Example 14, with the following exceptions:
Raw water: 1,680 cc of deionized water was used.
Additive used: A similar compound whose 2 percent aqueous solution at 20°C had a viscosity of 700 centipoises.
Treatment subsequent to incorporation of the additive: The mixture was agitated at 60°C for 20 minutes.

The said slurry was formed in the shape of a sheet having a thickness of about 0.4 mm on a glass sheet by using an applicator. Thereafter, the sheet was dried at 70°C for 20 hours according to an ordinary method. Consequently, there was obtained a white thin film having a thickness of about 38 microns.

This film was cut to pieces of a suitable size. Cigarettes were prepared by forming shredded tobacco in the shape of rolls and enclosing the rolls in the said cut pieces of film instead of using roll paper of the ordinary kind. When these cigarettes were trially smoked by about 50 smokers, as much as 96 percent of the smokers gave a rating that the smoke was less pungent and the taste was lighter than cigarettes enclosed in roll paper of the ordinary kind.

EXAMPLE 16:

About 1.25 kg of white, mashy slurry was obtained by repeating the procedure of Example 10A and B, except that 1,170 cc of deionized water and 30 g of powdered hydroxyethyl cellulose whose 2 percent aqueous solution at 20°C had a viscosity of 80 centipoises were gradually added into the mixture being agitated and the agitation was continued at 45°C for 30 minutes.

A laminar sheet tobacco composed of degraded cellulose, hydroxyethyl cellulose and tobacco dust at an approximate weight ratio of 10:3:16 was obtained by repeating the procedure of Example 10C, except that the drying was made at 60°C.

Cigarettes prepared from this sheet tobacco and those similarly prepared from comparative sheet tobacco were found to have the same taste as those of Example 10.

EXAMPLE 17:

About 1.7 kg of white, mashy slurry was obtained by repeating the procedure of Example 11, except that in Step B, 150 g of degraded cellulose was used in combination with 1,600 cc of deionized water, powdered hydroxyethyl cellulose whose 32 percent aqueous solution at 20°C had a viscosity of 3,600 centipoises was used in place of sodium cellulose glycolate, and the agitation was continued at 65°C for 50 minutes. The laminate sheet tobacco thus obtained was composed of degraded cellulose, hydroxyethyl cellulose and tobacco dust at an approximate weight ratio of 15:1:13 and was about 0.1 mm thick.

Another laminar sheet tobacco was obtained by following the procedure mentioned above, except that the amount of degraded cellulose used was lessened to 25 g. This sheet tobacco was found to be composed of degraded cellulose, hydroxyethyl cellulose, and tobacco dust at an approximate weight ratio of 2.5:1:2.5 and have a thickness of about 0.8 mm. Cigarettes were prepared from this laminar sheet tobacco by the same procedure as mentioned above. When these cigarettes were subjected to smoking test by the same method as mentioned above, 61 percent of the smokers perceived

objectionable odor in the taste of smoking and complained of unpleasant sensation.

EXAMPLE 18:

A yellowish brown sheet having a thickness of about 0.08 mm was obtained by repeating the procedure of Example 12, except that in Step B, 2,340 cc of deionized water was used, hydroxyethyl cellulose whose 2 percent aqueous solution at 20°C had a viscosity of 80 centipoises was used in place of sodium cellulose glycolate, and the agitation was continued at 58°C for 60 minutes to yield about 2.5 kg of yellowish brown, mashy slurry and in Step C, the slurry was formed in the shape of a sheet having a thickness of about 0.5 mm.

Cigarettes prepared from this sheet tobacco were totally identical in taste of smoking with those prepared in the corresponding example. The comparative sheet described above gave entirely the same results.

EXAMPLE 19:

The procedure of Example 13 was repeated except that in Step B, 150 g of degraded cellulose was used in combination with 2,280 cc of deionized water, powdered hydroxyethyl cellulose whose 2 percent aqueous solution at 20°C had a viscosity of 3,800 centipoises was used in place of sodium cellulose glycolate, and the agitation was continued at 50°C for 20 minutes to produce a white mashy slurry.

A tobacco dust paste was prepared by following the procedure of the said example. This paste was added to the said slurry and the resultant mixture was agitated at 60°C for 45 minutes. Consequently, there was obtained about 2.7 kg of yellowish brown slurry. With an applicator, this slurry was formed in the shape of a sheet by repeating the procedure of Example 1. It was dried to produce a sheet smoking mixture having a thickness of about 0.1 mm.

The results of the smoking test performed on cigarettes prepared of this sheet smoking mixture and on cigarettes of the comparative product were totally identical with the results obtained of those cigarettes prepared in the corresponding example.

EXAMPLE 20:

A. About 900 g of white degraded cellulose of the shape of short fibers about 0.1 mm in length having a D.P. distribution such that the average D.P. was 54 and the portion with D.P. in excess of 110 accounted for a barely detectable fraction of the whole composition was obtained by repeating the procedure of Example 1A, with the following exceptions:

Raw material: Wood pulp having an average D.P. of 620 and an α-cellulose content of 93.5 percent.

Steeping: The raw material was steeped in 17.8 percent aqueous solution of sodium hydroxide for 18 minutes.

Crushing: A garnet wire type crusher was used.

Alkali cellulose obtained consequently: Cellulose concentration 32.5% and total alkali concentration 15.5 percent.

Aging: The aging was carried out at 40°C in the atmosphere having an oxygen concentration of 42 percent for 310 hours.

Neutralization: The neutralization was effected with 10 percent hydrochloric acid.

B. About 1.6 kg of white mashy slurry was obtained by repeating the procedure of Example 8B, with the following exceptions:

Amount of deionized water used: 1,500 cc.

Additive used: Powdered methyl cellulose which had a D.S. value of 1.8 and whose 2% aqueous solution at 20°C had a viscosity of 3,500 centipoises was used in place of sodium cellulose glycolate.

Length of treatment subsequent to incorporation of the additive: 20 minutes.

C. The procedure of Example 7C was repeated.

The film obtained consequently was shredded to strips having a width of about 1 mm. The strips were mixed with shredded tobacco available on the market at the rate of 1:3. Cigarettes were prepared by forming the resultant mixture in the shape of rolls and enclosing the rolls with roll paper of the kind generally used for the production of ordinary cigarettes. When these cigarettes were trially smoked by about 50 smokers, nearly 100% of the smokers gave a rating that the taste became lighter. None of the smokers felt pungence in the smoke and the odor.

EXAMPLE 21:

About 2.3 kg of white mashy slurry was obtained by repeating the procedure of Example 20A and B, with the following exceptions:

Raw material: 55 g instead of 140 g of degraded cellulose and 2,275 cc of deionized water.

Rate of agitation: 3,000 rpm.

Additive: Powdered methyl cellulose which had a D.S. value of 1.7 and whose 2 percent aqueous solution at 20°C had a viscosity of 1,600 centipoises.

Operating condition subsequent to incorporation of the additive: The mixture was agitated at 45°C for 25 minutes.

A white thin film having a thickness of about 30 microns was obtained by repeating the procedure of Example 7C, except that the slurry was formed in the shape of a film having a thickness of about 0.8 mm and the film was dried at 70°C for 20 hours.

This film was cut into pieces having a suitable size. Cigarettes were prepared by using the pieces and then trially smoked by about 50 smokers. Consequently, 97 percent of the smokers gave a rating that the smoke was less pungent and the taste was lighter than cigarettes enclosed in roll paper of the ordinary kind.

EXAMPLE 22:

A laminar sheet tobacco composed of degraded cellulose, methyl cellulose and tobacco dust at an approximate weight ratio of 10:3:15 was obtained by repeating the procedure of Example 16, except that, in Step B, 1,040 cc instead of 1,170 cc of deionized water and powdered methyl cellulose which had a D.S. value of 1.6 and whose 2 percent aqueous solution at 20°C had a viscosity of 550 centipoises were used and, in Step C, the slurry was formed in the shape of a sheet having a thickness of about 0.7 mm instead of 0.6 mm.

The results of the smoking test performed on cigarettes prepared of this sheet tobacco and on cigarettes prepared of the comparative product were totally identical with the results obtained of those cigarettes prepared in the corresponding example.

EXAMPLE 23:

A laminar sheet tobacco composed of degraded cellulose, methyl cellulose and tobacco dust at an approximate weight ratio of 15:1:14 was obtained by repeating the procedure of Example 17, except that in Step B,

powdered methyl cellulose which had a D.S. value of 2.0 and whose 2 percent aqueous solution at 20°C had a viscosity of 5,800 centipoises was used in place of hydroxyethyl cellulose and the agitation was continued at 60°C for 40 minutes to afford about 3.3 kg of white mashy slurry.

The results of the smoking test performed on cigarettes prepared of this laminar sheet tobacco and on cigarettes prepared of the comparative product were totally identical with the results obtained of those cigarettes prepared in the corresponding example. The laminar sheet tobacco of which the said comparative product was formed was composed of degraded cellulose, methyl cellulose and tobacco dust at an approximate weight ratio of 2.5:1:3.5. The sheet had a thickness of about 0.1 mm.

EXAMPLE 24: D.S.

A yellowish brown sheet having a thickness of about 0.1 mm was obtained by repeating the procedure of Example 18, except that 2,080 cc of deionized water and powdered methyl cellulose which had a D.S value of 1.6 and whose 2 percent aqueous solution at 20°C had a viscosity of 550 centipoises were used and the agitation was continued at 65°C for 65 minutes to afford about 2.3 kg of yellowish brown mashy slurry. The drying of the formed sheet was carried out at 65°C.

The results of the smoking test performed on cigarettes prepared of this sheet smoking mixture and on cigarettes prepared of another sheet smoking mixture formed for the purpose of comparison in accordance with the procedure of the corresponding example were totally identical with the results obtained of the cigarettes prepared in the corresponding example.

EXAMPLE 25:

About 2.6 kg of yellowish brown slurry was obtained by repeating the procedure of Example 19, except that 140 g of degraded cellulose, 2,200 cc of deionized water and powdered methyl cellulose which had a D.S. value of 2.0 and whose 2 percent aqueous solution at 20°C had a viscosity of 5,800 centipoises were used and the agitation was continued at 60°C for 25 minutes to afford a white mashy slurry. The tobacco dust paste obtained by following the same procedure was mixed with this slurry and the resultant mixture was agitated at 65°C for 50 minutes.

The results of the smoking test performed on cigarettes prepared of this sheet smoking mixture and of cigarettes prepared for the purpose of comparison in accordance with the procedure of the corresponding example were totally identical with the results obtained of the cigarettes prepared in the corresponding example.

EXAMPLE 26:

A laminar sheet tobacco composed of decomposed cellulose, ethyl cellulose and tobacco dust at an approximate weight ratio of 10:3:14 was obtained by repeating the procedure of Example 22, except that in Step B, powdered ethyl cellulose having a D.S. value of 0.65 was used in place of sodium cellulose glycolate and, in Step C, the slurry was formed in the shape of a sheet having a thickness of 0.8 mm instead of 0.7 mm.

The results of the smoking test performed on cigarettes prepared of this laminar sheet tobacco and on cigarettes prepared for the purpose of comparison in accordance with the procedure of the corresponding example were similar to the results obtained of the cigarettes prepared in the corresponding example.

EXAMPLE 27:

About 3.1 kg of white mashy slurry was obtained by repeating the procedure of Example 23, except that 3,000 cc of deionized water was used in combination with 140 g of degraded cellulose and powdered ethyl cellulose having a D.S. value (as ethyl group) of 1.4 was used.

By following the same procedure, a laminar sheet tobacco composed of degraded cellulose, ethyl cellulose and tobacco dust at an approximate weight ratio of 14:1:12 was obtained from the said slurry.

Cigarettes were prepared from this sheet by repeating the procedure of Example 11. When these cigarettes were trially smoked, the results were similar to the results obtained of the cigarettes prepared in the corresponding example. In the case of other cigarettes prepared for the purpose of comparison by the procedure of the corresponding example, as much as 60 percent of the smokers perceived objectional odor in the taste of smoking and complained of unpleasant sensation.

EXAMPLE 28:

About 2.5 kg of yellowish brown, mashy slurry was obtained by repeating the procedure of Example 24, except that the amount of deionized water was 2,340 cc, powdered ethyl cellulose having a D.S. value (as ethyl group) of 0.65 was used in place of methyl cellulose and the agitation was continued at 63°C for 55 minutes.

The said slurry was formed in the shape of a sheet having a thickness of 0.6 mm and the sheet was dried in the same manner. Consequently, there was produced a yellowish brown sheet having a thickness of about 0.09 mm.

The results of the smoking test performed on cigarettes prepared of this sheet and of cigarettes prepared for the purpose of comparison in accordance with the corresponding example were identical with the results obtained of the cigarettes prepared in the corresponding example.

EXAMPLE 29:

About 3.0 kg of yellowish brown slurry was obtained by repeating the procedure of Example 25, except that 2,690 cc of deionized water was used in combination with 130 g of degraded cellulose, powdered ethyl cellulose having a D.S. value (as ethyl group) of 1.4 was used in place of methyl cellulose and the agitation was continued at 55°C for 20 minutes to afford a white mashy slurry. The white mashy slurry was mixed with the tobacco dust paste prepared separately and the resultant mixture was agitated at 65°C for 45 minutes.

Cigarettes were prepared of the sheet obtained from the yellowish brown slurry and other cigarettes were prepared for the purpose of comparison in accordance with the procedure of the corresponding example. The results of the smoking test performed on these cigarettes were totally identical with the results obtained of the cigarettes prepared in the corresponding example.

EXAMPLE 30:

A. The amount of 732 g of white degraded cellulose

of the shape of short fibers about 0.2 mm in length having a D.P. distribution such that the average D.P. was 72 and the portion with D.P. in excess of 110 accounted for 1.5 percent of the whole composition was obtained by repeating the procedure of Example 1A, with the following exceptions:

Raw materials: Wood pulp having an average D.P. of 656 and an α-cellulose content of 93.5 percent.

Steeping: The raw material was steeped in the state of 4.0 percent slurry in 17.5 percent aqueous solution of sodium hydroxide, with the agitation continued for 18 minutes.

Alkali cellulose obtained consequently: 2.8 kg of alkali cellulose was found to have a cellulose concentration of 32.0 percent and a total alkali concentration of 15.6 percent.

Aging: The aging was carried out for 200 hours in the atmosphere having an oxygen concentration of 44 percent instead of 42 percent.

Neutralization: The neutralization was effected by using 20 percent sulfuric acid.

B. About 1 kg of slurry was obtained by repeating the procedure of Example 8B, with the following exceptions:

Raw material: 60 g of white degraded cellulose of the shape of short fibers about 0.23 mm in length and 925 cc of distilled water.

Additives used: 5 g of powdered sodium cellulose glycolate which had a D.S. value of 0.9 and whose 1 percent aqueous solution had a viscosity of 1,200 centipoises and powdered hydroxyethyl cellulose whose 2 percent aqueous solution at 20°C had a viscosity of 4,000 centipoises.

C. The procedure of Example 8C was repeated.

EXAMPLE 31:

A. About 850 g of white degraded cellulose of the shape of short fibers about 0.2 mm in length having a D.P. distribution such that the average D.P. was 72 and the portion with D.P. in excess of 110 accounted for 0.4 percent of the whole composition was obtained by repeating the procedure of Example 10A.

B. About 1 kg of white mashy slurry was obtained by repeating the procedure of Example 8B, with the following exceptions:

Raw material: 75 g of degraded cellulose and 910 g of deionized water.

Additives: 10 g of powdered sodium cellulose glycolate which had a D.S. value of 1.0 and whose 1 percent aqueous solution at 25°C had a viscosity of 1,700 centipoises and 5 g of powdered methyl cellulose which had a D.S. value of 1.8 and whose 2% aqueous solution at 20°C had a viscosity of 3,500 centipoises.

Operating condition subsequent to incorporation of the additive: Agitation made at 60°C for 20 minutes. C. A laminar sheet tobacco composed of degraded cellulose, sodium glycolate plus methyl cellulose, and tobacco dust at an approximate weight ratio of 10:2:17 and having a thickness of about 0.1 mm was obtained by repeating the procedure of Example 10C.

D. This laminar sheet tobacco was shredded to strips having a width of about 0.5 to 1 mm. Cigarettes were prepared by forming the strips in the shape of rolls and enclosing the rolls in roll paper of the kind generally used for the production of ordinary cigarettes. On trial smoking, these cigarettes were found to have a very light and mild taste. Absolutely no pungence was perceived in the smoke and the odor.

EXAMPLE 32:

About 2.0 kg of yellowish brown, mashy slurry was obtained by repeating the procedure of Example 24, except that 2,000 cc of deionized water, 10 g of methyl cellulose which had a D.S. value of 1.6 and whose 2 percent aqueous solution at 20°C had a viscosity of 550 centipoises and 20 g of hydroxyethyl cellulose whose 2 percent aqueous solution at 20°C had a viscosity of 80 centipoises were used and the agitation was continued at 63°C for 55 minutes.

The said slurry was formed in the shape of a sheet having a thickness of 0.6 mm. This sheet was dried in the same manner as mentioned above to afford a yellowish brown sheet having a thickness of about 0.09 mm.

This sheet was shredded into strips having a width of about 0.5–1 mm. When the shredded pieces were trially smoked with smoking pipes, the taste of smoking was found to be fine and light. Neither pungence nor objectionable odor was perceived.

What we claim is:

1. A substitute tobacco material comprising a degraded cellulose having an average D.P. of up to 95 and prepared by treating a cellulose with an alkali followed by aging of the resulting alkali cellulose with an oxygen-containing gas, and sodium cellulose glycolate in which the degree of substitution of carboxymethoxy group is between 0.6 and 1.0 percent aqueous solution thereof has a viscosity of between 80 and 1,800 centipoises at 20°C., the said degraded cellulose being contained therein in an amount at least 75 percent by weight of said material.

2. A substitute tobacco material comprising a degraded cellulose having an average D.P. of up to 95 and prepared by treating a cellulose with an alkali followed by aging of the resulting alkali cellulose with an oxygen-containing gas, and hydroxyethyl cellulose, a 2 percent agueous solution of which hydroxyethyl cellulose has a viscosity of between 70 and 4,000 centipoises at 20°C., the said degraded cellulose being contained therein in an amount at least 75 percent by weight of said material.

3. A substitute tobacco material comprising a degraded cellulose having an average D.P. of up to 95 and prepared by treating a cellulose with an alkali followed by aging of the resulting alkali cellulose with an oxygen-containing gas, and methylcellulose in which the degree of substitution of methoxyl group is between 1.6 and 2.0 and a 2 percent aqueous solution thereof has a viscosity of between 500 and 6,000 centipoises at 20°C., the said degraded cellulose being contained therein in an amount at least 75 percent by weight of said material.

4. A substitute tobacco material comprising a degraded cellulose having an average D.P. of up to 95 and prepared by treating a cellulose with an alkali followed by aging of the resulting alkali cellulose with an oxygen-containing gas, and ethyl cellulose in which the degree of substitution of ethoxyl group is between 0.6 and 1.5, the said degraded cellulose being contained therein in an amount at least 75 percent by weight of said material.

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