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PROCESS FOR THE PREPARATION OF RECONSTITUTED TOBACCO IN SHEET FORM AND PRODUCT OBTAINED THEREBY

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ABSTRACT OF THE DISCLOSURE

A process is provided for preparing reconstituted tobacco in sheet form from viscous, aqueous pasty dispersions of finely divided tobacco, from 1 to 30% of a water-soluble cellulose derivative binder having a flocculation temperature below about 80° C., and water-soluble polyalkylene glycol having a molecular weight of from 200 to 6000 to increase the flocculation temperature of the cellulose derivative to above 80° C. A reconstituted tobacco material of the above composition is also provided.

The present invention relates to a process for the preparation of reconstituted tobacco and to the product thereby obtained.

According to a known process for the preparation of sheet tobacco one prepares a paste form mixture of finely divided tobacco material and a water-soluble cellulose derivative as a binder whereupon the paste form mixture is formed to leaves, sheets or other coherent structures, which are subjected to drying. This method gives satisfactory results as long as the drying is carried out relatively slowly at temperatures below the flocculation point of about 70–80° C. which is the case when using older types of film-forming tobacco machines. When using modern machines with more rapid drying at higher temperatures it has, however, up till now not been possible to use the inexpensive and from many other points of view advantageous water-soluble cellulose ethers as binders because of the flocculation and some other disadvantages. The modern machines use higher temperatures than 70° C. which results in the cellulose derivatives precipitating from the binder solution in the form of milk-white flocks and the binder solution thereby completely losing its viscous consistency and binding capacity.

It has now surprisingly been found possible to obtain a substantial raise of the flocculation point and simultaneously obtaining a great increase in viscosity by means of a simple addition to these cellulose derivatives according to the invention. The addition enables reconstituted tobacco leaves to be produced in modern machines having rapid drying course without any obstacles. The product obtained is flexible and cohesive, due to the greater uniformity and homogeneous distribution of the binder.

According to the process of the invention reconstituted tobacco in sheet form is prepared from viscous, aqueous dispersions of finely divided tobacco and from 1 to 30%, preferably from 5 to 15% b.w. of the tobacco of a binder comprising a water-soluble cellulose derivative having a flocculation temperature below about 80° C., the characterizing feature being completely dissolving the cellulose derivative in water and mixing into the solution obtained a polyalkylene glycol having a molecular weight of from 200 to 6000, preferably from 300 to 600, and a vapor pressure of at least 10^{-3} mm. Hg at 20° C. in an amount sufficient to increase the flocculation temperature of the cellulose derivative to above 80° C., and thereafter drying the dispersion to form reconstituted tobacco in any desired form, such as sheets, foils and the like, which can be comminuted, ground or shredded, as desired. Preferably the binder solution contains at least 2.5 parts by weight of polyalkylene glycol per part by weight of cellulose derivative, with no critical upper limit except as the taste of the tobacco may be affected. Usually an amount as high as 15 parts per part of cellulose derivative has no effect on taste, and even more than this can be used if desired.

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Suitable water-soluble cellulose derivatives that can be used according to the invention are non-ionic cellulose ethers having a flocculation temperature of below 80° C. such as methylcellulose, ethylcellulose, hydroxyethylcellulose, methylhydroxyethylcellulose, ethylhydroxyethylcellulose, hydroxypropylcellulose and ethylhydroxypropylcellulose. Other water-soluble cellulose derivatives that can be used according to the invention are ionic cellulose ethers, such as carboxymethylcellulose and methylcarboxymethylcellulose, and alkali- and ammonium salts thereof. The viscosity of the water-soluble cellulose derivative should be from 45 to 3500 cp., preferably from 700 to 3000 cp. in 2 percent aqueous solution at 20° C. Particularly preferred is ethylhydroxyethylcellulose having a viscosity of from 2000 to 3000 cp., which is preferably used in an amount of from 1 to 4 parts by weight per 500 to 1000 parts by weight of water.

Suitable polyalkylene glycols include polyethylene glycol, polypropylene glycol, or mixed polyethylenepropylene glycols or mixtures thereof having a molecular weight of from 200 to 6000, preferably from 300 to 600 and having a vapor pressure less than 10^{-3} mm. Hg at 20° C. and having a viscosity at 100° C. of from 3 to 1000 centistokes, preferably from 5.5 to 10.0 centistokes. Particularly preferred is polyethylene glycol having a molecular weight of about 400, and a vapor pressure of 10^{-8} mm. Hg at 25° C., and a viscosity of about 7.3 centistokes at 100° C. The amount of polyalkylene glycol is at least 2.5 to about 15 times the amount of cellulose derivative used.

A particularly preferred combination according to the invention is a binder solution containing about 3.3 percent by weight of ethylhydroxyethylcellulose with the viscosity 2000–3000 cp. in 2 percent aqueous solution at 20° C. (Modocoll E. 600®, Mo och Domsjö Aktiebolag) and 17.0 percent polyethylene glycol with the molecular weight 400 and a vapor pressure of 10^{-8} mm. Hg at 25° C. (Modopeg 400®, Mo och Domsjö Aktiebolag), the remainder being water.

The polyalkylene glycol can be mixed into the cellulose derivative solution in pure form but it is, of course, also possible to dissolve it in water before this and thereafter mix it into the cellulose derivative solution in the desired amount. Incorporation of the polyalkylene glycol must be carried out first after having dissolved the cellulose derivative completely in water to the formation of a clear, homogeneous solution free from lumps.

The ratio of cellulose derivative to polyalkylene glycol is depending on the increase in flocculation temperature and thickening effect that are desired in each case, and the molecular weight of both components. Although the proportions given above are suitable in most cases it is, of course, possible to vary them within wide limits.

In order to obtain the desired effect according to the present invention it is absolutely necessary that the mixing of the cellulose derivative and the polyalkylene glycol is carried out in such a way that the polyalkylene glycol is added to a water solution of the cellulose derivative after the latter has been completely dissolved, and not vice versa. If the cellulose derivative or a solution thereof is added to a polyalkylene glycol solution no positive effect at all is obtained. It is believed that this phenomenon is quite new and surprising, and it will be further illustrated in the experiments below.

Examples A to C

The effect on flocculation temperatures of water solutions of ethylhydroxyethylcellulose of different viscosities at different concentrations is shown by the following data. The cellulose derivative was completely dissolved in water, and then, with stirring, different amounts of polyethylene glycol having the molecular weight 400 and a vapor pressure of 10^{-8} at 25° C. were added. The viscosity of the solution obtained was then measured at 20° C. in a Brookfield viscosimeter. The flocculation temperature of the solutions was determined at 1 percent concentration of the cellulose derivative by immersing the samples in a water bath, the temperature of which gradually was raised until flocculation occurred. The results obtained are shown in Table I below.

TABLE I

A. Ethylhydroxyethylcellulose, 250–400 cp. Percent B.W.		Polyethylene glycol, M.W. 400 Percent B.W.	Viscosity, cp.	Flocculation temperature, ° C.
1	2	0	369	72
2	2	2	387	76
3	2	5	458	79
4	2	10	643	86
5	2	20	1,710	90
6	3	0	2,540	------
7	3	5	4,800	------
8	3	10	11,300	------
B. Ethylhydroxyethylcellulose 700–1100 cp. percent B.W.				
1	2	0	1,050	73
2	2	2	1,080	------
3	2	5	1,130	78
4	2	10	1,580	------
5	2	20	3,320	95
C. Ethylhydroxyethylcellulose 2,000–3,000 cp. percent B.W.				
1	1	0	------	79
2	1	5	------	85
3	1	10	------	91
4	1	20	------	95
5	3.3	0	17,700	------
6	3.3	17	161,000	------
Control [1]	3.3	17	2,750	79

[1] In this example the polyethylene glycol was first dissolved in water after which the cellulose derivative was added to the polyethylene glycol solution.

Examples A to C show the remarkable increase in the flocculation temperature that occurs when the polyalkylene glycol in accordance with the present invention is added to the completely dissolved cellulose derivative. Simultaneously a desirable viscosity increase is obtained. Example C control shows that with an inverted order of addition, i.e. when the cellulose derivative is added to a solution of the polyalkylene glycol, the effect is totally absent.

The addition of the finely divided tobacco material, which can consist of ground tobacco leaf, tobacco stems, tobacco scrap, tobacco dust etc., can be made in different ways with consideration of the above mentioned critical conditions at the addition of the polyalkylene glycol. Thus one can mix the finely divided tobacco material with dry, powdered cellulose derivative and thereafter add water in the desired quantity with continued stirring. When the cellulose derivative is completely dissolved one can add polyalkylene glycol or polyalkylene glycol solution in desired quantity, and thereafter form the paste form mixture obtained to sheets or other coherent bodies and dry. Another possibility is to first dissolve the cellulose derivative in desired quantity of water and then mix the finely ground tobacco herewith, after which polyalkylene glycol is added and shaping and drying is carried out as mentioned above. A third especially suitable method is first to prepare a water solution of the cellulose deriva-

tive, and after complete dissolution of the cellulose derivative to add polyalkylene glycol, and then mix the finely divided tobacco material into the binder solution obtained, and shape and dry the mixture.

In the above mixing procedures it should be avoided that air comes into the binder solution in order to avoid undesirable frothing (foaming).

In addition to the binder other substances used in similar compositions may be added to the tobacco material, such as fire improving agents, humectants, softeners, fillers, reinforcing agents, pH-regulators etc. Examples of such substances are activated montmorillonite, organic and inorganic acids such as maleic, citric, tartaric, phosphoric, and boric acid, glycerol and lower glycols, sorbitol, mineral and cellulose fibres, and diatomaceous earth.

It is also possible according to the present invention to substitute part of the water used as solvent with more volatile organic solvents, such as ethylalcohol, acetone, isopropylalcohol etc. as well as to substitute part of the water-soluble cellulose derivative with a cellulose derivative soluble in organic solvents in order to improve the water resistance of the film.

If a low water solubility of the tobacco sheet material prepared is desired, it is suitable to add a treatment with an agent which produces cross-linking in the cellulose material in connection with the drying. Suitable such agents are for example tannin, and glyoxal, Such treatment with glyoxal may be carried out before, during or after the paste form mixture is carried through the drying oven. Particularly suitable is to apply glyoxal immediately before the sheet enters the oven and to have the cross-linking to take place at the elevated temperature in the oven. The invention is further illustrated by the following examples.

Example D

Dry tobacco leaf was ground in a mill to such a particle size that 100% passed a 80 mesh sieve. In a vessel provided with a stirrer were added 3.3 kilograms of ethylhydroxyethylcellulose having the viscosity 2000–3000 cp. in 2 percent aqueous solution at 20° C. (Modocoll E 600 ®) after which 1000 kilograms of water were added with stirring. Stirring was continued at room temperature for 30 minutes till the cellulose derivative had completely dissolved and formed a homogeneous, clear solution without lumps. Then 20 kilograms of polyethylene glycol with the molecular weight 400 and a vapor pressure of 10^{-8} mm. Hg. at 25° C., 6 kilograms of burning regulator (colloidal silica), and 6 kilograms of diatomaceous earth were added with stirring. When a homogeneous mixture had been obtained 100 kilograms of powdered tobacco scrap was mixed in until a homogeneous paste was obtained. The paste was pumped to a storage tank from which it then was supplied to a sheet-forming tobacco machine, where it was laid down on a 16.5 meter drying belt running through an electrically heated oven with a drying temperature of 85 to 90° C. A strong sheet with the thickness 0.1 mm. was obtained which showed an extraordinarily smooth surface and good flexibility.

Example E

To the same mixer as in Example D were added 100 kilograms of powdered tobacco scrap after which 10 kilograms of dry powdered ethylhydroxyethylcellulose with the viscosity 250–400 cp. in 2 percent aqueous solution at 20° C. (Modocoll E 100 ®) were mixed in. When a homogeneous mixture had been obtained 500 kilograms of water were added with continued stirring. After about 30 minutes of mixing 20 kilograms of polyethylene glycol with the molecular weight 600 and a vapor pressure of 10^{-9} mm. Hg at 25° C., dissolved in 200 kilograms of water, were added. From the paste form mixture obtained a reconstituted tobacco sheet of the thickness 0.1 mm. was prepared in the same way as in

Example D. The sheet obtained was strong and had an extraordinarily good surface eveness and flexibility.

Example F

To the same mixer as in Example E were added 100 kilograms of powdered tobacco scrap, after which 12 kilograms of dry, powdered carboxymethylcellulose with the viscosity 100–150 cp. in 2 percent aqueous solution at 20° C. were mixed in. When a homogeneous dry mixture had been obtained 500 kilograms of water were added with continued stirring. After 40 minutes mixing 20 kilograms of polyethylene glycol with the molecular weight 300 and a vapor pressure of 10^{-7} at 25° C., dissolved in 200 kilograms of water, were added. From the paste form mixture obtained a reconstituted tobacco sheet of the thickness 0.1 mm. was prepared in the same way as in Example D. The sheet obtained was strong and exhibited a very good surface smoothness and flexibility, which remained even after a long time of storage.

What is claimed is:

1. In a process for the preparation of reconstituted tobacco in sheet form from viscous, aqueous pasty dispersions of finely divided tobacco and from 1 to 30% by weight of the tobacco of a binder comprising a water-soluble cellulose derivative having a flocculation temperature below about 80° C., the dispersions being formed into foils and similar structures and subjected to drying, the improvement which comprises completely dissolving the cellulose derivative in water and mixing into the solution obtained a water-soluble polyalkylene glycol having a molecular weight of from 200 to 6000 and a vapor pressure of less than 10^{-3} mm. Hg at 20° C. in an amount of at least 2.5 parts per part of cellulose derivative present and sufficient to increase the flocculation temperature of the cellulose derivative to above 80° C. and forming a dispersion of finely divided tobacco, the cellulose derivative, and the water soluble glycol before rendering the tobacco in sheet form.

2. A process according to claim **1**, in which the water-soluble cellulose derivative is a non-ionic cellulose ether selected from the group consisting of methyl cellulose, ethyl cellulose, hydroxyethyl cellulose, methyl hydroxyethyl cellulose, hydroxypropyl cellulose, ethyl hydroxyethyl cellulose and ethyl hydroxypropyl cellulose.

3. A process according to claim **1**, in which the water-soluble cellulose derivative is an ionic cellulose derivative selected from the group consisting of carboxymethyl cellulose and methyl carboxymethyl cellulose and alkali- and ammonium salts thereof.

4. A process according to claim **1**, in which the water-soluble polyalkylene glycol is polyethylene glycol.

5. A process according to claim **1**, in which the finely divided tobacco first is mixed with the cellulose derivative, said cellulose derivative being in dry, powderous form, after which the water is added to completely dissolve the cellulose derivative.

6. A process according to claim **1**, in which the finely divided tobacco is added to a water solution of the cellulose derivative, and then the water-soluble polyalkylene glycol is added.

7. A process according to claim **1**, in which the polyalkylene glycol is mixed with an aqueous solution of the cellulose derivative and then the finely divided tobacco is added to the mixture.

8. A process according to claim **7**, in which 100 parts by weight of the finely divided tobacco is mixed with a binder solution containing from 2 to 10 parts by weight of ethyl hydroxyethyl cellulose having a viscosity of from 2000 to 3000 cps. in 2 percent water solution at 20° C., and from 500 to 1000 parts by weight of water, to which binder solution has been added water-soluble polyethylene glycol having a molecular weight of from 350 to 450 in such an amount that the binder solution contains from 15 to 30 parts by weight of polyethylene glycol per part of weight of ethyl hydroxyethyl cellulose.

9. A process according to claim **1**, in which the reconstituted tobacco material is treated with a cross-linking agent which forms cross-links in the cellulose derivative during the drying process.

10. A process according to claim **9**, in which the cross-linking agent is glyoxal.

11. A reconstituted tobacco material comprising tobacco, a water-soluble cellulose derivative which in aqueous solution has a flocculation temperature below about 80° C., and a water-soluble polyalkylene glycol having a molecular weight of from 200 to 6000 and a vapor pressure of at least 10^{-3} mm. Hg at 20° C. in an amount of at least 2.5 parts per part of cellulose derivative present to increase the flocculation temperature of the cellulose derivative in aqueous solution to above 80° C., and improved cohesiveness and flexibility of the product.

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