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PROCESS FOR THE MANUFACTURE OF RECONSTITUTED TOBACCO FOIL

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This invention relates to the manufacture of coherent tobacco foil containing a high percentage of tobacco and suitable as a binder for cigars, cigarillos, and similar tobacco products. More particularly it relates to a self-supporting reconstituted tobacco sheet of ground tobacco and an alkali-soluble methyl cellulose.

Numerous methods have been suggested for preparing reconstituted tobacco foils from finely divided or pulverized tobacco particles unsuitable for direct use in tobacco products. Such reconstituted foils must meet the general test of acceptability in taste and burning characteristics as well as having adequate physical properties such as tensile strength, dimensional stability and moisture retention. Furthermore, it is most desirable commercially to avoid use of an organic solvent system to minimize fire, explosion and toxicity hazards.

In United States Patent 3,025,860 Grossteinbeck and Holtz describe a method for preparing tobacco foils by intimately mixing finely divided tobacco with a liquid binder and thereafter extruding the mixture into a precipitating bath to solidify the binder and form the tobacco foil. Such cellulose derivatives as methyl cellulose, acetyl cellulose, carboxymethyl cellulose and ethyl cellulose are disclosed as particularly suitable binders when dissolved in a suitable solvent such as water, methylene chloride, acetone or a lower alcohol. With organic solvents water is recommended as the precipitating agent, while for aqueous binder solutions water-miscible precipitants such as sulfuric acid, glycerol, glycol or sorbitol are recommended.

The present invention concerns a process for preparing a reconstituted tobacco foil using alkali-soluble methyl cellulose as the binder. The process comprises (A) preparing a tobacco slurry by intermixing finely divided tobacco with an alkaline solution of alkali-soluble methyl cellulose, (B) forming a thin stream of the tobacco slurry and treating it with an aqueous inorganic acid-salt solution to coagulate the binder and form a self-supporting film, and (C) drying the film.

In the process the composition of the coagulation bath is an essential feature. The inorganic acid neutralizes the tobacco slurry while the inorganic salt causes coagulation and dehydration of the resulting insoluble methyl cellulose binder. The coagulated tobacco film has sufficient wet strength so that it is easily transferred through the process baths.

When dried the reconstituted tobacco foil or sheet is flexible over a wide range of temperature and humidity. Because of its appreciable wet strength, it has reduced sensitivity to humidity changes. As a further advantage of the present process, it has been noted that nicotine can be extracted from the tobacco during the processing and that with appropriate reagent concentrations and contact times the residual nicotine content of the reconstituted tobacco foil can be controlled.

In the practice of the present invention an alkali-soluble methyl cellulose is employed as the tobacco binder. These methyl cellulose ethers are insoluble in water and organic solvents but soluble in 2 to 8 percent aqueous sodium or potassium hydroxide. They are lightly substituted ethers having a methoxyl content of about 5 to 13 weight percent corresponding to a degree of substitution (D.S.) ranging from about 0.2 to 0.7. Particularly suitable is the alkali-soluble methyl cellulose described by Maasberg in United States Patent 2,408,326.

To obtain adequate blending it is necessary that the final slurry of alkali-soluble methyl cellulose and tobacco have a maximum viscosity of about 5,000 cps. at 20° C. Conveniently a 5 to 8 percent solution of an alkali-soluble methyl cellulose having a 2 percent viscosity of about 15–100 cps. at 20° C. is used. For optimum results it is usually desirable to filter the alkaline methyl cellulose solution to remove insoluble gels prior to addition of the tobacco.

Any finely divided tobacco may be employed. Tobacco chips, stems, clippings, and dust as well as other tobacco remains from normal manufacturing processes can be pulverized or ground into a suitable form. Preferably the tobacco should be fine enough to pass through a standard 20 mesh screen. A finer grind gives a more homogeneous and smoother appearance to the reconstituted foil.

To obtain a self-supporting reconstituted tobacco foil it is necessary to use from about 15 to 50 parts of alkali-soluble methyl cellulose per 100 parts of tobacco. While greater amounts of binder can be used, better smoking quality is obtained, of course, with a minimum of binder. If desired, other conventional additives such as plasticizers, combustion catalysts or retardants, humectants, can be added with the tobacco or methyl cellulose or directly to the tobacco-methyl cellulose slurry. Thorough blending is accomplished with conventional equipment. Heating the slurry above about 30° C. should be avoided to minimize discoloration of the tobacco.

To form the reconstituted tobacco foil, the binder slurry is preferably extruded into a coagulating bath through a die having a slit of proper dimensions to obtain a dried reconstituted foil about 1.2 to 8 mils thick and of the desired width. Alternately it may be extruded or cast onto a suitable surface such as a continuous belt and conveyed into the bath. The coagulant solution is preferably dilute sulfuric acid saturated with sodium sulfate. As the caustic in the extruded slurry is neutralized by the acid, the sodium sulfate causes immediate coagulation and gelation of the water-insoluble methyl cellulose binder thereby forming a self-supporting film which can be drawn from the bath, water washed, dried and stored for use in the manufacture of tobacco products.

The inorganic acid-salt bath used to treat the extruded film serves not only to convert the alkali-soluble methyl cellulose into a water-insoluble form but also dehydrates and "salts out" the methyl cellulose film. Since sulfates are particularly effective in "salting out" the methyl cellulose film, the sulfuric acid-sodium sulfate bath is preferred. However, baths containing hydrochloric or phosphoric acid and the corresponding salts can also be used. For rapid neutralization with the preferred sulfuric acid bath, the concentration of sulfuric acid should be at least 8 percent and preferably from about 10 to 30 percent. The sodium sulfate content should be at least 15 percent and preferably near the saturation level. By addition of make-up sulfuric acid and water and removal of spent coagulation solution, the concentration of acid and salt can be maintained within desired limits for continuous operation. Bath temperatures of from about 15 to 45° C. can be employed, but since rapid coagulation is enhanced by heat, a temperature between 30 and 45° C. is often advantageous. Higher temperatures tend to degrade the tobacco foil.

To achieve essentially complete neutralization and coagulation of a tobacco film up to about 10 mils thick, a contact time in the sulfuric acid-sodium sulfate bath of from about 10 to 60 seconds at 30° to 40° C. is usually adequate. The reconstituted tobacco foil is then taken from the coagulation bath and washed with water to re-

move the excess acid and the soluble salts. The water wash can be accomplished using a water bath, water sprays, or other means, but preferably with water preheated to between 40° and 95° C. since hot water is more effective in removing the salts and strengthening the coagulated film.

Following the water wash the reconstituted tobacco foil can be further treated as desired prior to final drying. For example, brief contact with a dilute sodium bicarbonate solution can be used to insure complete neutralization of any residual inorganic acid. Further water washing can be used to reduce the nicotine content. A foil weakened by prolonged soaking in water can be strengthened by return to the acid-salt bath. Finally the tobacco foil can be plasticized with a humectant if desired by dipping in 5 to 15 percent aqueous propylene glycol or glycerine just prior to drying.

The reconstituted tobacco foil is dried at 20 to 120° C. advantageously through contact with a heated metal roll having a surface temperature of from about 100–120° C. To minimize darkening of the tobacco foil, temperatures higher than about 120° C. should be avoided.

In this process a reconstituted tobacco foil having a dried thickness of at least 1.2 mils has adequate strength even in the coagulation bath so that it is self-supporting under normal operating conditions. However, for high speed operations, use of a continuous belt or similar means to support the foil may be advantageous.

The dried reconstituted tobacco foil containing alkali-soluble methyl cellulose as a binder is flexible and self-supporting over a wide range of temperature and relative humidity. Typically it contains by analysis less than 0.5 percent organic acid calculated as sulfuric acid and less than 0.1 percent inorganic sulfate. It is readily wound into coils which can be stored under ambient conditions although storage at a relative humidity of from about 20 to 80 percent is preferred for optimum quality. This reconstituted foil has enhanced resistance to moisture and suitable tensile strength and dimensional stability for use as a binder in the preparation of tobacco products.

This invention is further illustrated by the following representative embodiment. Unless otherwise specified, all parts and percentages are by weight.

To a solution of 90 parts of sodium hydroxide in 800 parts of water was added 53 parts of alkali-soluble methyl cellulose (6.0% —OCH_3) having a viscosity of 72 cps. as a 2% solution in 6% caustic at 20° C., 4 parts of a liquid polyoxyethylene glycol (M.W. 200) and 4 parts of a solid polyoxyethylene glycol (M.W. 4,000). The mixture was stirred thoroughly while cooling to about 0° to 5° C., filtered through a 150 mesh screen to remove insoluble particles and deaerated by centrifuging. Then a slurry of 159 parts of finely divided tobacco (100 mesh) and 610 parts of water was added to the alkaline methyl cellulose solution. The binder and tobacco slurry was thoroughly intermixed, deaired and then transferred to the reservoir of a film extrusion die having a 0.014" x 5.0" slit.

The die was positioned at one end of a sulfuric acid-sodium sulfate bath with the slit adjusted horizontally below the surface of the coagulant solution. The solution contained by analysis 12.9% sulfuric acid and 20.3% sodium sulfate and was held in a nickel trough 4.5" wide, 48" long, with a bath depth of about 2". The bath temperature was about 28° C. By means of air pressure on the reservoir, the slurry was extruded into the bath as a stream which instantly coagulated to a continuous film. The coagulated film was withdrawn at the opposite end of the bath after a total immersion time of about 30 seconds using a 2.5" diameter plastic roll having a surface speed of 8 ft./min. It was then rinsed in a bath of flowing hot water (60–65° C.) for about 30 seconds and wound continuously on a drum driven by a slip clutch.

A portion of this reconstituted film was further treated by immersion in a 1.2% sodium bicarbonate solution for 10 seconds followed by rewashing in the hot rinse bath and finally by immersing in a solution of 10% aqueous glycerol for about 1 to 3 seconds. Several samples of the plasticized film were dried on glass plates overnight at room temperature and 50% relative humidity. The remainder was dried on an infrared heated polished metal cylinder having a surface temperature of about 110 to 120° C. with a contact time of 2 minutes. Darkening occurred at higher temperatures. The dried tobacco film had a thickness of 3 to 4 mils, was flexible, moisture resistant and easily coiled.

Film samples taken at several stages were analyzed for sulfuric acid and sodium sulfate content by repeated extraction with hot water and analysis of the aqueous extract by standard methods. Typical analyses are given in Table 1.

TABLE 1

Stage	Analyses [a]			
	Total weight	Water [b]	H_2SO_4	Na_2SO_4
1. After coagulation bath	10.30	7.45	0.555	1.30
2. After first water wash	11.11	9.10	0.26	0.65
3. Dried foil	1.75	0.75	0.003	<0.001

[a] Parts per part of dried extracted foil.
[b] Includes water-soluble plasticizers.

Using a standard Instron test machine, the dry strength of the plasticized tobacco foil having a film thickness of from 3 to 4 mils was 400–700 p.s.i. The wet strength of the foil was determined using a Scott IP–2 Serigraph and a 1" x 2" strip of foil, the load being applied one minute after wetting the foil. For foils about 3 mils thick typical wet strengths ranged from 100 to 200 g./in. width and for foil about 4 mils thick, from 200–300 g./in. width.

In other similar runs the extruded tobacco-alkali-soluble methyl cellulose slurry was neutralized and coagulated with dilute hydrochloric acid saturated with sodium chloride and with dilute phosphoric acid saturated with sodium phosphate. Satisfactory reconstituted tobacco foil was obtained.

We claim:

1. A method of producing a reconstituted tobacco foil comprising:
   (A) Preparing a tobacco slurry by intermixing 100 parts of finely divided tobacco with an alkaline solution containing from 15 to 50 parts of alkali-soluble methyl cellulose having a D.S. of about 0.2 to 0.7, said slurry having a viscosity of less than about 5,000 cps. at 20° C.;
   (B) Forming a thin stream of the tobacco slurry and treating it with an aqueous inorganic acid-salt coagulated bath selected from the group consisting of aqueous sulfuric acid and sodium sulfate, aqueous hydrochloric acid and sodium chloride, and aqueous phosphoric acid and sodium phosphate whereby it is coagulated into a self-supporing film; and
   (C) Drying the film.

2. The method of claim 1 wherein the alkali-soluble methyl cellulose has a viscosity of from about 15 to 100 cps. as a 2 percent solution in dilute caustic at 20° C.

3. The method of claim 1 wherein the tobacco slurry is extruded into the coagulating bath.

4. The method of claim 1 wherein a coagulant bath of sulfuric acid and sodium sulfate is employed.

5. The method of claim 1 wherein a coagulant bath of hydrochloric acid and sodium chloride is employed.

6. The method of claim 1 wherein a coagulant bath of phosphoric acid and sodium phosphate is employed.

7. The method of claim 4 wherein the bath con-

tains at least 8 weight percent of sulfuric acid and 15 weight percent of sodium sulfate.

8. A reconstituted tobacco foil comprising finely divided tobacco and a minor proportion of alkali-soluble methyl cellulose characterized by a degree of substitution of from about 0.2 to 0.7 and a viscosity of from about 15 to 100 cps. as a 2 percent solution in dilute caustic at 20° C.

9. The reconstituted tobacco foil of claim 8 containing from about 15 to 50 parts of alkali-soluble methyl cellulose per 100 parts of finely divided tobacco.

10. The method of claim 4 wherein the tobacco slurry is coagulated in a bath containing from 10–30 weight

percent sulfuric acid and at least 15 weight percent sodium sulfate at a temperature of 30–45° C. with a contact time of 10–60 seconds.

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