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METHOD OF MAKING A COMPOSITE TOBACCO SHEET

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This application is a continuation-in-part of application Ser. No. 241,122, which was filed on November 30, 1962, now abandoned.

This invention relates, generally, to the method of making a composite tobacco sheet. More particularly, the invention relates to methods of making improved reconstituted tobacco compositions. In addition, the invention relates to methods for the production of adhesive compositions containing enzyme mixtures and to smoking compositions made therefrom.

During the production and processing of tobacco products, including aging, blending, sheet forming, cutting, drying, cooling, screening, shaping and packaging considerable amounts of tobacco fines and tobacco dust are produced. The tobacco industry has also long been interested in methods which would make it possible to effectively employ the midribs and stalks of the tobacco plant. It is known that tobacco fines, dust and midribs can be combined with a binder to form a coherent sheet, which resembles leaf tobacco and which is commonly referred to as reconstituted tobacco. One method for making reconstituted tobacco of this general character is disclosed in U.S. Patent 2,734,510, wherein the tobacco fines and dust are applied to the binder made of carboxymethyl cellulose, carboxymethyl hydroxyethyl cellulose or a suitable salt thereof. The binder, in such compositions, ranges from about 5% to about 50% of the weight of the tobacco employed. U.S. Patent 2,708,175 describes a binder for reconstituted tobacco which consists of plant gums, principally galactomannan. U.S. Patent 2,592,554 to Frankenburg describes, as binders for reconstituted tobacco, various water soluble polysaccharides, such as alginic and pectinic acids and their sodium and potassium salts. However, the additional cellulosic binders further increase the amount of cellulosic material in the products and tend to create an acrid and bitter smoke when the product is used to make cigarettes. The natural hydrophilic colloid gums such as guar, locust bean, algin and other commonly used materials such as Irish moss have additional disadvantages. These materials contain proteins and other materials not found in tobacco which add distinctive flavors of their own to tobacco products during smoking. Thus, Frankenburg in describing the use of various water soluble polysaccharides from plants other than tobacco teaches that care should be exercised that they must be in a state of refinement. Frankenburg teaches that these materials should be free of extraneous matter containing compounds of nitrogen, particularly proteins, and compounds of sulfur, phosphorous, the halogens, and other compounds giving undesirable products of combustion and dry distillation. Such refining is often a very tedious and difficult operation.

Sometimes the tobacco fines or stems have been chemically treated to give the desired coherence to the sheet which is formed therefrom. Some of the methods employed to make a sheet use a sandwich-type construction in which upper and lower layers of tobacco fines are adhered to a middle portion which serves as a binder for the tobacco particles. These methods of producing reconstituted tobacco sheets are costly. The processing involved is lengthy and adhesive materials must be added to the tobacco fines or the tobacco must be chemically treated to provide an adhesive quality. Most of the additives are

foreign to the tobacco and cause undesirable taste and odor notes in the smoke from the tobacco product. The quality in the sheet is not comparable to that of the natural tobacco leaf because the physical properties such as sheet thickness, tensile strength and elongation are inferior to those of the leaf.

In all of the above-described methods humectants and plasticizers are generally required in order to provide a sheet of satisfactory mechanical properties.

The present invention makes possible the production of improved reconstituted tobacco by a method which is more effective and more economical than the methods previously employed.

In accordance with the present method, no costly additives are employed and no chemical treatment of the tobacco fines or stems is necessary. Since substantially no foreign matter is introduced during the sheet formation, the flavor and aroma for that tobacco product or the smoke from the product are not adversely affected. By the present method the use of plasticizers is no longer required. The present method further makes possible the production of very thin reconstituted tobacco sheets which can be made with a thickness comparable to that of the natural tobacco leaf. The tensile strength and elongation of sheets made in accordance with the present invention are greatly improved.

In accordance with the present invention, reconstituted tobacco sheets of excellent quality are obtained by mixing dry ground tobacco stems or other tobacco plant parts with water and a catalytic amount of a specific enzyme system. The enzyme system should be such that it at least partially degrades three materials in the tobacco: the cellulose, the hemi-celluloses, and the pectin.

Suitable enzyme systems must exhibit cellulase activity, hemi-cellulase activity and pectinase activity, i.e. they must include a cellulase, a hemi-cellulase and a pectinase.

The cellulases, hemi-cellulases and pectinases which can be employed include fungal or bacterial preparations derived from *Aspergillus niger* or *Aspergillus oryzae*. All three enzymes may be present in a single preparation or it may be necessary to combine more than one such preparation to provide the desired combination of the three enzymes. Such enzyme systems, in general, are derived by the treatment of plant extracts and extracts from animal organs and from fungal and bacterial cultures. Several commercial cellulase preparations and the organisms from which they are prepared are listed in a paper by C. S. Walseth (TAPPI 35: 228, 1952). Specific commercial materials which have been found to include cellulase, hemi-cellulases and pectinase are Cellulase No. 35 (Rohm and Haas Co.), Cellase 1000 (Wallerstein Co.), Cellulase 4000 (Miles Laboratories), Lipase B (Rohm and Haas Co.), and Pectinase (Nutritional Biochemical Corp.). These preparations contain varying amounts of other enzymes, such as protease, amylase, lipase and catalase. These additional enzymes are not necessary for the purposes of the present invention but it has been found that they have no undesirable effects on the products of the present invention.

One method which can be employed to determine whether or not an enzyme system exhibits cellulase activity is to place ten grams of cellophane in a solution containing one gram of the enzyme system and maintained at a pH of 4.5 and a temperature of 45° C. for a period of 24 hours. After 24 hours, the remaining cellophane is removed from the solution by filtration, is washed with water and dried in a forced air circulation oven at a temperature of 45° C. until constant weight is attained. A control is run, using cellophane treated under exactly the same condition, but in the absence of any enzyme. Cellulase activity, as it is used in this application, is exhibited when the cellophane shows a weight decrease after the

treatment of 0.5% or more, as compared with the control which contained no added enzymes.

One method which can be employed to determine whether or not an enzyme system exhibits hemi-cellulase activity is to make the following tests: 5 grams of guar gum and 5 grams of xylan are separately admixed with 500 ml. portions of an acetic acid-acetate buffer solution having a pH of 4.5 and a molarity of 0.04. In the case of the guar gum, this material is completely dissolved. However, in the case of the xylan, this material only partially dissolves and the remainder of the material is present in suspension. Then one portion of 0.05 gram of the enzyme system is added to each mixture, the mixtures being maintained at 45° C. for a period of 24 hours. The resulting mixtures are then each placed in 1000 ml. of acetone, whereby precipitates form which comprise undegraded or little degraded guar gum and xylan, respectively. These precipitates are separately filtered, dried in a similar manner as described above, to constant weight. Controls are also run for each of the materials, under identical conditions, except that no enzyme is present. Hemi-cellulase activity, as it is used in this application, is exhibited when each of the materials, i.e., the guar gum and the xylan, shows a weight decrease after this treatment of 10% or more, as compared with a control which contained no added enzymes.

One method which can be employed to determine whether or not an enzyme system exhibits pectinase activity is to dissolve 5 grams of pectin in an acetic acid-acetate buffer solution having a pH of 4.5 and a molarity of 0.04. Then 0.05 grams of the enzyme system is added to the solution, which is maintained at 45° C. for a period of 24 hours. The resulting solution is then placed in 1000 ml. of acetone, whereby a precipitate forms which comprises undegraded or little-degraded pectin. This precipitate is filtered, dried as described above until it is brought to a constant weight. A control is also run under identical conditions, except that no enzyme is present. Pectinase activity, as it is used in this specification, is exhibited when the pectin shows a weight decrease after this treatment of 10% or more, as compared with the control which contained no added enzymes.

Obviously, other assay methods which will determine the existence of the three enzymes may also be employed.

While we do not wish to be bound by any particular theory, we believe that the outstanding results obtained in accordance with this invention are due to the fact that the present enzyme systems split the high-molecular weight, water insoluble pectic, cellulolytic and hemi-cellulolytic components of tobacco into either lower-molecular weight, water-soluble materials, or slightly higher molecular weight substances which swell in water. The resulting materials are the adhesives which cause the tobacco particles to adhere together to result in the superior reconstituted tobacco sheets of the present invention.

Enzyme concentrations which may be employed should be in the range of 0.01 to 40%, based on the weight of the tobacco material, but are preferably in the range of 0.03 to 10%. The preferred upper limit of 10% is based on the economics involved in using an excess of the enzyme system. The upper limit of 40% is based on the necessity of keeping the concentration low enough to prevent the formation of combustion products in the smoke from the pyrolysis of the enzyme itself, and by the fact that high concentrations cause too great an increase in the sugar content of the tobacco. This high content of sugars makes the paste excessively gluey and gives an undesirable flavor to the smoke of the final tobacco product. The most preferred concentration of the enzyme system is 0.1%–1.0%.

Tobacco stems and fines may be employed in various forms. The fines or ground stems may be of various sizes, varying from particles which will pass through a 10 mesh screen to particles which will be retained by a 400 mesh screen.

It is preferred during the treatment of the tobacco with the enzyme system that water be present in an amount corresponding to about 9 parts by weight per part of tobacco, in order to form a slurry of the tobacco parts. However, from about 5 to about 20 parts, by weight, of water per part of tobacco will be satisfactory, depending on the particular method which is used.

The treatment time is dependent upon the mesh size of the tobacco material, the enzyme concentration, the degree of agitation, the temperature, and the pH of the slurry. Generally this will vary between about 30 minutes and 48 hours, but is preferably between about 2 and 12 hours.

The pH range of the tobacco composition may be from about 3.5 to about 6.0 but is preferably between about 4.0 and 5.0.

The treatment temperature can be between about 20° and about 80° C., with the preferred temperature being in the lower portions of this range, for example from about 25° C. to about 60° C.

During the treatment, the tobacco-enzyme system must be agitated. This may be accomplished by continuous stirring or the like or by periodic agitation.

The invention may be illustrated by the following examples:

Example 1

Forty grams of ground bright stems (midribs), which passed through a 325 mesh screen and which were retained on a 400 mesh screen, were suspended in 300 ml. of water. Cellulase 35 concentrate (0.04 g., or 0.1% based on the weight of the tobacco stem material) was added to the suspension. This material (produced by Rohm and Haas Company) is a mixture of polysaccharases including cellulase, pectinase and hemi-cellulase. The slurry was kept at 50° C. in a Waring Blendor and stirred occasionally.

After 24 hours, the contents of the Blendor were stirred vigorously again, and then cast with a casting knife on a glass plate covered with a trace of silicone grease to facilitate removal of the sheet. After drying at room temperature, a thin sheet of very good mechanical quality was easily peeled off the glass plate. Measurements obtained with an Instron Tester showed that the tensile strength was 1.1 kg. (per inch width) and the elongation 1.5%. The sheet was 0.20 mm. in thickness.

A second lot of 40 grams of ground bright stems of the same mesh size as that given above was made into a suspension in 300 ml. of water. No enzyme was added. Processing procedures identical to those used above were carried out. It was observed that the suspension was not as viscous as that of the one to which the enzyme had been added. The dried sheet had little cohesion between the particles and crumbled readily. It was not possible to shred it in the conventional manner for use as a cigarette filler. However, cigarettes were prepared from it to use as a control in smoking tests.

The cast control sample and the sheet made from the enzyme-treated tobacco were both analyzed by standard methods of analyses for cellulose, hemicellulose, and pectin. The analyses showed that the three carbohydrates had decreased and that the water solubles had increased in the enzyme-treated sheet. By paper chromatography, it was determined that mono- and oligo-saccharides had increased markedly in the treated product, indicating that the enzyme mixture had acted upon the high molecular-weight carbohydrates to break the chain linkages. Thus, the high molecular-weight cellulose, hemicellulose, and pectin were broken down into lower molecular-weight materials giving an adhesive quality to the slurry and making it possible to cast a sheet with excellent physical characteristics.

The sheet made by the enzyme treatment of tobacco was shredded into cigarette filler and cigarettes were prepared from it. Subjective evaluation of the smoke from

the control cigarette and from the treated tobacco showed that the treatment did not adversely affect the flavor, aroma, burning qualities, or ash strength.

Five additional tobacco sheets were also made by the above technique employing, respectively, in place of the 0.04 g. of Cellulase 35 concentrate the following: 0.4 g. of Lipase B (Rohm and Haas Co.), 0.04 g. Cellase 1000 (Wallerstein Co.), 0.4 g. Pectinol 10 M (Rohm and Haas Co.), 0.4 g. Pectinase S.C. (Miles Laboratories), and 0.4 g. Pectinase (Nutritional Biochemical Co.). These sheets were similar in nature to the sheet prepared using Cellulase 35.

Example 2

A sheet was prepared as in Example 1 except that 4 g. of Cellulase 35 (10% based on the weight of the tobacco stem material) was added to the suspension of ground tobacco stems and water. The sheet was darker and somewhat stickier than that obtained by using 0.1% of the enzyme system. It was very plastic and tore, rather than broke, on the Instron Tester. Its thickness was 0.07 mm. Its tensile strength 0.22 kg. (per inch width), and it had an elongation of 3.4%.

Analysis for water solubles in a cast control sample made only with ground tobacco stems and water and in the sheet made from the enzyme treated stems showed a 25% increase in water solubles in the treated material. Standard analyses of the control and treated sample showed a 29% decrease in cellulose content, a 32% decrease in hemicellulose, and a 54% decrease in pectin for the treated tobacco. By paper chromatographic analyses, it was found that the mono- and oligo-saccharide content of the treated sheet was high.

Cigarettes made from a control and from the shredded, treated sheet showed that the treatment had not adversely altered the smoking flavor and smoking characteristics.

Example 3

Ground bright stems which were retained on a 50 mesh screen were treated with 0.1% of Cellulase 35 as in Example 1 and cast into a sheet. The sheet had a coarser texture than the sheets made from stems ground to a finer mesh. Its thickness was 0.25 mm. It had a tensile strength by Instron testing of 1.0 kg. (per inch width) and an elongation of 3.9%. The sheet when shredded and made into cigarettes had smoking qualities similar to those of the sheet prepared as in Example 1.

Example 4

Ground bright stems which were retained on an 80 mesh screen were treated as in Example 1 without the addition of the enzyme system. The suspension after treatment at 50° C. in a Waring Blendor could not be spread smoothly into a thin layer. The dried sheet crumbled easily, had no continuous texture, and only very little binding power. This material was used as in the control for a sheet prepared in an identical manner except that 0.3% Cellulase 35 was added. The cast and dried sheet from the enzyme-treated tobacco showed excellent adhesive and mechanical properties similar to those of the sheet prepared in Example 1.

Example 5

Bright stems, ground in a Wiley Mill and not sieved to specific mesh sizes were treated as in Example 1 with 2.5% Cellulase 35. The cast and dried sheet had good adhesive and mechanical properties. Its smoking characteristics were similar to those of the treated sheets prepared in Examples 1–5 above.

Example 6

A mixture containing a variety of mesh sizes and a variety of tobacco types (50% ground bright stem, 25% ground burley stem, and 25% tobacco leaf fines) was treated as in Example 1 with 2.5% Cellulase 35. The slurry was cast on a silicon-treated glass plate and dried at room temperature. The resulting sheet had a good texture, good mechanical properties, and good smoking characteristics.

Example 7

Forty grams of bright tobacco leaf, both aged and unaged, were ground and put through a 50-mesh sieve. Each lot of fines was mixed with 400 ml. of water and treated as in Example 1 with 0.1% Cellulase 35 (based on the weight of the tobacco fines). The slurries were cast on glass plates, dried at room temperature, and removed as sheets. The sheets showed very good tensile strength and burned with good burning characteristics.

Cigarette filler was made from a control and from the sheet made with the treated batch lots.

Smoking evaluation of cigarettes made from the sheet prepared with treated leaf fines showed that the smoke from the treated sample had an added sweetness and a slight floral note.

Example 8

A sheet was formed using the same substances, same amounts, and same procedures as those given above in Example 7 except that, before casting, 2 g. of crystalline magnesium nitrate was added (based on 40 grams of tobacco) to alter the burning qualities of the filler to be prepared from the sheet. A sheet was obtained which had a 0.09 mm. thickness, a 0.42 kg. tensile strength, and a 2.0% elongation. The addition of the metallic salt increased the tensile strength and modified the burning characteristics.

A similar addition of 2 g. of crystalline aluminum chloride (based on 40 grams of tobacco) to a sheet prepared as in Example 7 markedly increased the tensile strength and decreased the elasticity.

Example 9

To 150 gallons of water at 40° C. was added 125 pounds of ground bright stems which passed through a 50 mesh screen and 250 grams (0.5% by weight) of Cellulase No. 35. Intermittent agitation by a Cowles dissolver was employed and a small immersion heater was used to maintain the temperature at 45° C. A sample of the slurry was taken 3 hours from the start of the operation and was cast into a test sheet. This test sheet showed good tensile properties.

The slurry thickened rapidly and stayed then at about constant viscosity until the end of the enzyme treatment which lasted for 23 hours. Additional heating was not necessary for the last 12 hours. Intermittent stirring maintained the temperature at 45° C. The pH of the slurry was 4.85. A sandwich sheet with underdust and overdust as well as a binder-only type sheet were produced.

The sheets showed the following properties:

	Binder-Only Sheet (Sprayed film)	Sandwich-Type Sheet (Sprayed onto tobacco dust and covered with tobacco dust)
Basis weight, gms./ft.^{2}	11.7	13.1
Moisture at room conditions, percent	12.5	11.0
Tensile, kg./in	0.97	0.77
Elongation, percent	8.5	1.6
Work, gm. cm./in.^{2}	162.9	17.8
Tear, gms		6.4

Example 10

To 150 gallons of water at 25° C. was added 180 pounds of whole bright stems and 350 grams of Rohm and Haas Cellulase 35 to form a slurry. Ten hours after the start of the operation, agitation by a Cowles dissolver was started. In less than two hours, after agitation was started, the viscosity of the slurry rose to 57,000 centipoises (as measured by a Brookfield Viscometer model

RVF, Spindle 6, at 10 r.p.m.). Agitation was continued for another two hours and a sandwich sheet was produced in a manner similar to that described in Example 9. The sheet showed the following properties:

Basis weight, gms./ft.^{2}	15.1
Moisture at room conditions, percent	13.3
Tensile, kg./in.	0.36
Elongation, percent	5.9
Work gm. cm./in.^{2}	37.6
Folded tensile, kg./in.	0.35

Example 11

Two pounds whole bright stems were soaked in water overnight. The next morning the stems and the liquid were poured into a Valley beater, additional water was added and the temperature raised to 45° C. The valley beater divided the stems into fine particles by a crushing action. 30.0 g. Cellulase 35 concentrate was added. Water was added to a consistency of 1.2 and a freeness of 100. Two hours after the addition of the enzyme the product was cast into sheets which, after drying, showed good mechanical properties.

As used herein, the terms "cellulase," "hemicellulase" and "pectinase" are used interchangeably with, respectively, "cellulases," "hemicellulases" and "pectinases."

We claim:

A method for producing a coherent tobacco sheet comprising the steps of (1) admixing for a period of from about 30 minutes to about 48 hours and at a temperature of from about 20° C. to about 80° C. tobacco plant parts with water and with from 0.1 to 40%, based on the weight of the tobacco plant parts, of a mixture of enzymes comprising cellulase, hemi-cellulases and pectinase, to form a mixture having a pH of from about 3.5 to about 6.0, (2) forming a pulp from said mixture, (3) forming said pulp into a wet sheet and (4) drying said wet sheet, sufficient

cellulase being present in said mixture of enzymes to cause a weight loss of at least 0.5% in cellophane when 10 parts of said cellophane are placed in an aqueous solution containing one part of said mixture of enzymes, the mixture of enzymes and cellophane being maintained at a pH of 4.5 and a temperature of 45° C. for a period of 24 hours, sufficient hemi-cellulase being present in said mixture of enzymes to cause a weight loss of at least 10% in a mixture of equal parts of guar gum and xylan contained in an acidic acid buffer solution having a pH of 4.5 and a molarity of 0.04, when 200 parts of said mixture of guar gum and xylan are admixed with one part of said mixture of enzymes and the mixture of enzymes and the mixture of guar gum and xylan are maintained at a temperature of 45° C. for a period of 24 hours, sufficient pectinase being present in said mixture of enzymes to cause a weight loss of at least 10% in pectin which has been dissolved in an acetic acid buffer solution having a pH of 4.5 and a molarity of 0.04, when one part of said mixture of enzymes is combined with 100 parts of said pectin and the mixture of enzymes and pectin is maintained at a temperature of 45° C. for a period of 24 hours.

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