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[54] **PROCESS OF MANUFACTURING**
RECONSTITUTED TOBACCO OF LIGHT COLOR
1 Claim, No Drawings

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ABSTRACT: A process of treating tobacco in the manufacture of reconstituted tobacco is disclosed wherein the color of the finished reconstituted product is lightened by the removal of basic polyphenols present in the starting batch. The process involves washing the tobacco with water to form an aqueous extract thereof, separating the extract and treating it with absorbent agents such as activated alumina or polyamids to remove the basic polyphenols and finally reincorporating the extract minus the removed polyphenols into the fibrous tobacco mass.

PROCESS OF MANUFACTURING RECONSTITUTED TOBACCO OF LIGHT COLOR

The present invention relates to a method for the preparation of reconstituted tobacco sheet and more particularly to an improved process for preparing light colored reconstituted tobacco sheet, despite the use of heat as a processing aid at any step of the process.

The commercial method for the manufacture of reconstituted tobacco sheet requires the application of heat at some stage in the processing e.g. either in the preparative stages and/or in the drying. It is known that the color of the finished tobacco product darkens because of the heating; the level of darkening being a function of temperature and heating time. As a rule, it is desirable to produce a light colored tobacco sheet and various steps are taken to achieve this goal, for example, the addition of dyes or coloring chemicals, the addition of color masking agents ("whiteners") such as titanium dioxide or Dicalite, or control of temperature and heating time. These steps are never completely satisfactory because of the limitations under which they can be applied. In the case of the addition of dyeing, coloring and masking chemicals, the introduction of extraneous material can be undesirable, especially if excessive amounts are needed to produce the desired results. Excessive use of dyes can produce "leaching" problems while excessive use of "whiteners" can result in sheets which are greyish in appearance or lack proper physical structure. In the case of controlled temperature and heating time, undesirable limits are put upon the manufacturing procedures. The invention herein revealed makes it possible to produce a light colored, reconstituted tobacco sheet without the resort to dyes for color preservation and without any undue limitations on the temperature and heating time used in the process. This is achieved by selectively removing from the tobacco the agents responsible for the darkening of the tobacco on heating.

In the preparation of additive reconstituted tobacco, tobacco, such as stems, field scrap, cuttings, whole leaf and the like, is preferably uniformly comminuted and formed into tobacco sheet suitable for later machine processing. On the other hand, in the preparation of all-tobacco reconstituted sheet or in reconstituted sheets wherein a more efficient utilization of the natural gums of the tobacco is desired, the tobacco is usually ground to colloid-forming size particles by a wet process or refining. However, the large amount of energy and time required for the preparation of these colloid-forming size tobacco particles has limited its use for the preparation of reconstituted sheets.

To reduce the energy expended and time involved to comminute the tobacco particles in the wet process, it is desirable to cook the tobacco prior to refining. This cooking operation reduces the refining time and simultaneously improves the physical properties of the finished product. However, the cooking causes darkening of the tobacco and hence darkens the sheets made from this cooked tobacco material, thereby making such sheets unacceptable in a number of manufactured tobacco products.

Many attempts have been made heretofore to produce reconstituted tobacco sheets using heat at some stage in the processing without the adverse darkening of the tobacco, using various techniques and methods, but, in general, such attempts have met with only limited commercial success. For example, it has been known in the art, that washing the tobacco products with water prior to treatment, replacing the washings with fresh water and then cooking the washed tobacco gives a relatively acceptable product with respect to color. However, it is unsatisfactory from an economic standpoint, since the discarded washings contain a large quantity of useful tobacco product.

The present invention provides a solution to this longstanding problem, and results in a great reduction in energy and time required to obtain good quality reconstituted tobacco

sheets of acceptable color.

A further object of the present invention is to provide a method for producing a light colored reconstituted tobacco sheet despite the use of heat as a processing aid.

5 A still further object of the present invention is to remove polyphenols from the tobacco prior to exposure of the tobacco-water mixture to heat in the preparation of the reconstituted tobacco sheet without resort to dyes and color chemical additives to the tobacco.

10 Another object of the present invention is to remove from the tobacco prior to exposure of the tobacco-water mixture to heat, only the agents causing the darkening of the tobacco and leaving all other valuable material in the tobacco, without adding anything to the tobacco, so as to provide a light colored tobacco sheet.

15 Still another object of the present invention is to provide a method for accomplishing all of the above, which can be used in processing large quantities of tobacco resulting in a reconstituted sheet of tobacco having an acceptable color and to do so in an economical and easily controllable method.

20 Other objects and advantages of the present invention will be apparent from the further reading of the specification and of the appended claims.

Tobacco as defined for this invention includes any type of tobacco suitable for the manufacture of reconstituted tobacco sheet, such as stem, veins, scrap and waste tobacco, cuttings and the like, as well as whole leaf or portions thereof.

In general, the method of the present invention discloses a method of removing from the tobacco washings the agents responsible for the darkening of the tobacco when it is exposed to heat. The treated washings are then returned to the tobacco being processed, and the product prepared in a conventional manner to provide a reconstituted tobacco sheet of good physical properties without detriment to the sheet color.

30 It has long been commonly believed that the agents responsible for the darkening of the tobacco on cooking were the carbohydrates and the amino acids, which react during the heating to form dark colored compounds, the process being called the "Maillard" reaction. We have discovered that neither the carbohydrates nor the amino acids darken the tobacco significantly on heating, but that polyphenols contained in the tobacco, especially the basic-polyphenols, do cause a significant darkening of the tobacco upon exposure to heating. This phenomenon is also pH dependent.

35 A series of tests were run to isolate and determine the agents which caused tobacco darkening upon heating. The following procedure illustrates how the polyphenols were identified as the agents which cause tobacco darkening during heating. A sample of "Bright" stem dust weighing 10 g. was mixed with 90 ml. of distilled water. The slurry was filtered on a Buchner funnel using Whatman No. 1 filter paper. The filter cake and filtrate were retained. The filter cake was subsequently mixed with 90 ml. of a predetermined liquid and the slurry transferred to a vessel, such as a mason jar, which was sealed and placed in a pressure cooker, where it was heated at 250° F. at 15 p.s.i. for 2 hours. Upon cooling, three sheets having a thickness of about 0.25—0.30 mm. were cast from the slurry on 20 cm. x 20 cm. glass plates. The sheets were dried in an oven at 110—115° C. for about 15—20 minutes and the color of each sheet was read in a Gardner automatic color difference meter, Model AC-2 A, Series 200. Readings were made by placing the plate over the Gardner aperture with the tobacco side facing the aperture. A number of readings were taken at different points on each plate so as to obtain an overall average.

40 The test results in table I were obtained with various additives in the predetermined liquid and serve to illustrate that polyphenols are the agents causing the darkening of the tobacco on exposure to heat. Water-washed Bright stem dust was used in all cases, unless otherwise indicated. The weight of the washed tobacco in each sample was 7 g. dry weight and this was suspended in 90 ml. of liquid prior to heating.

TABLE 1.—COLOR OF SHEETS PREPARED FROM BRIGHT STEM DUST HEATED IN SOLVENTS CONTAINING VARIOUS ADDITIVES

Materials	pH before heating	pH after heating	Rd	a+	b+
Washed tobacco plus water (positive control).....	5.3		24.9	3.1	21.5
Unwashed tobacco plus water.....		3.8	16.3		
Washed tobacco plus washings (negative control).....		4.0	16.4		
Washed tobacco plus sucrose (2 gm.) plus water.....			25.4		
Washed tobacco plus glucose (2 gm.) plus water.....			25.2		
Washed tobacco plus fructose (2 gm.) plus water.....			24.7		
Washed tobacco plus sucrose, fructose, glucose (2 gm. each) plus water.....	7.2		26.8		
Washed tobacco plus tryptone (1 gm.) (casein digest) plus water.....	6.3		22.3	2.7	16.3
Washed tobacco plus tryptone plus sucrose (1 gm. each) plus water.....	6.1		22.9	2.5	17.3
Washed tobacco plus tryptone plus glucose (1 gm. each) plus water.....	6.1		22.2	3.3	17.5
Washed tobacco plus tryptone plus fructose (1 gm. each) plus water.....	6.1		22.0	2.5	17.1
Washed tobacco plus tryptone plus glucose, fructose, sucrose (1 gm. each) plus water.....	6.2		21.9	3.8	18.2
Washed tobacco plus water plus acidic polyphenols.....	5.2	4.5	22.6	3.5	16.3
Basic polyphenols plus washed tobacco plus water.....	9.3	6.3	12.4	5.8	15.0
Acidified basic polyphenols plus washed tobacco plus water.....	5.7	4.8	17.2	4.7	16.1
Washed tobacco plus P _b acetate sol. (10%) water.....			23.4		

NOTE.—Rd=Brightness factor; black=0; white=100; a+, b+=Coordinates on a chromaticity diagram; P_b Acetate=lead acetate.

The polyphenol fractions shown in table I were obtained as follows: 90 ml. of a tobacco-water extract, prepared as described above, was mixed with 20 ml. of a 10 percent lead acetate solution, and the formed precipitate was removed by centrifugation. This precipitate comprised the acidic polyphenol fraction.

Adding more lead acetate solution to the supernatant caused no more precipitation. An alkaline reagent, such as ammonium hydroxide (NH₄OH) was added to the supernatant until the pH was about 8.5—9. The formed precipitate, which was collected by centrifugation, constituted the basic polyphenol fraction.

As seen from the experimental results shown in table I, the basic polyphenols make a sheet darker than even the negative control (Rd readings 12.4 and 16.4, respectively). Furthermore, it is seen that while acidifying the basic polyphenols improves the color, it is still far below the positive control (Rd readings of 17.2 and 24.9, respectively). These results show that the sugars and amino acids do not affect the sheet color and similarly neither does the lead acetate solution used to precipitate the polyphenols.

Processes for the neutralization or removal of the polyphenol from tobacco with the addition of chemical substances are fully disclosed and described in our copending application, Ser. No. 639,066 filed even date herewith. The present disclosure confines itself to the removal of polyphenols without chemical substances.

In order to extract the basic polyphenols from the tobacco without adding any chemical additive to the reconstituted tobacco sheet, the tobacco water washings can be treated with adsorbent material, which adds nothing to the extract, but selectively removes the darkening agent or agents by adsorption. For example, tobacco water washings are passed through a column containing a selected adsorbent material, such as activated alumina. The adsorbent removes the darkening material, but leaves the other valuable tobacco constituents unchanged in the extract. The treated extract is then recombined with the original washed tobacco and the combination is processed into reconstituted tobacco sheet, which retains its commercially acceptable light color with no additives.

In accordance with the present invention, untreated tobacco, such as broken leaves, cuttings, stemmings, various fragments or waste, field scrap, is subjected to a water extraction operation so as to separate the water-soluble constituents.

The water-soluble constituents of the tobacco or extract are separated from the insoluble portion by any convenient process, such as draining, pressing, or centrifuging. The aqueous extract contains various water-soluble materials, including polyphenols. This aqueous extract can then be passed through a column containing an adsorbent material, such as activated alumina. Continuous operation in a column can be effected by placing the adsorbent material and passing the aqueous extract continuously through a column. The adsorbent material removes the color-forming bodies, but leaves the other valua-

ble tobacco constituents unchanged in the column effluent. The effluent is then recombined with the original washed tobacco and the combination is processed in a conventional form and a light colored reconstituted tobacco sheet is produced. The extracted coloring materials can be eluted from the adsorbent so that it can be reused as desired.

The following examples are set forth to demonstrate the method of this invention.

A glass column (25 cm. x 3 cm.) was filled with a slurry of 100 gm. of activated alumina (8—14 mesh) in water. The water was allowed to drain and the column was washed repeatedly with about 500—1000 ml. fresh water. A water extract of tobacco dust obtained by the method described previously was passed over the column at a flow rate such that effluent was collected at 0.5 ml. per minute. The column was then washed with water until the washings were colorless. The combined water washings and collected effluent were then concentrated to 100 ml. under vacuum in a flash evaporator using a temperature range of 36—38 C. The solids content in the concentrate, as well as in the original water tobacco extract were determined and the recovery of material from the column calculated from these values. The test results are presented in table 2.

TABLE 2.—SOLIDS IN TOBACCO-WATER EXTRACTS AND IN COLUMN EFFLUENT FROM ALUMINA COLUMNS

Sample	Tobacco stems	Solids in H ₂ O extract, percent	Solids put on column (gm.)	Solids recovered in concentrated effluents (gm.)	Percent recovery
1.....	Bright.....	4.50	9.00	6.73	74.5
2.....	do.....	4.57	9.14	6.72	74.5
3.....	do.....	4.52	9.04	6.52	72.5
4.....	do.....	4.55	9.10		
5.....	Penn.....	3.17	6.34		
6.....	Same.....	3.15	6.30	5.63	89.2
7.....	do.....	3.15	6.30	5.64	89.3
8.....	do.....	3.14	6.28	5.67	90.5

The figures in table 2 show that the solids recovery is 73—75 percent for Bright stem extracts and 89—91 percent for Penn. stem extracts.

The material not removed from the column by water washing was eluted off by passing 2 normal formic acid through the column. Fractions were collected at a rate of 0.5 ml. per minute. The eluted samples were very dark in color. The 2 normal formic acid removed all the adsorbed material leaving the activated alumina free of color. The alumina was washed with water until free of acid, dried in the oven (130—140° C.) overnight and used again.

The formic acid was removed from the eluate (i.e., the basic-polyphenols) by evaporation under vacuum in a flash evaporator. The remaining dark brown material was made up to a 100 ml. volume with water and tested for carbohydrates by a thin layer chromatography method, such as described in

the Journal of Chromatography 18, 170—174 (1965). Samples of the effluent concentrate from the column previously referred to as well as the original tobacco-water extract were also examined for carbohydrates. The results showed that practically all of the carbohydrates found in the original tobacco-water extract were recovered in the concentrated effluent while only significant amounts were found in the formic acid eluate.

It should be noted that the material, which could not be removed from the column by water could be eluted off by other suitable solvents, such as 50 percent ethanol containing 2 normal HCl or by 2 normal HCl solution. Advantageously, formic acid is preferred, since it is easily removed by evaporation under vacuum. This makes it possible to recover the solids from the formic acid eluate and their subsequent utilization in the color experiments. In an operation where these solids are of no consequence, their removal from the alumina would be carried out more economically with a 2 normal HCl solution.

The effluent concentrates were made up to volume and recombined with the washed tobacco. The slurries were treated in a manner as described above. The obtained sheets were examined in a Gardner colorimeter, and the results are presented in table 3.

TABLE 3.—GARDNER READINGS ON SHEETS PREPARED WITH VARIOUS EFFLUENTS AND ELUATES FROM ALUMINA COLUMNS

Tobacco	Fraction	pH of tob. slurry	Rd	a+	b+
Bright.....	Effluent concentrate.	5.8	21.5	3.7	17.5
Do.....	do.....	5.6	24.6		
Do.....	do.....	4.7	24.6	5.4	34.4
Do.....	Formic acid eluate..	3.2	17.4	4.1	22.7
Bright (positive control).....		5.3	24.8	3.9	35.0
Bright (negative control).....		4.8	16.8	4.8	22.3
Penn.....	Effluent concentrate.	6.3	19.6	5.0	24.6
Penn.....	Formic acid eluate..	3.8	17.1	6.1	23.3
Penn. (positive control).....		7.0	19.3	5.2	24.6
Penn. (negative control).....		6.2	17.4	5.3	22.8
Bulgarian.....	Effluent concentrate.	5.1	15.8		
Bulgarian (positive control).....		5.7	15.3		
Bulgarian (negative control).....		5.2	11.3		

Note.—Positive control consists of washed tobacco (7 gm. dry weight) plus new water. Negative control consists of washed tobacco plus the obtained washing.

The figures in table 3 show that the effluent concentrate from the adsorbent column is similar in its color properties to the positive control, while the formic acid eluate, after removing the formic acid and adjusting to volume with water, is similar to the negative control. This is particularly apparent in the Rd values.

Adsorbents, other than activated alumina, were examined. Regenerated alumina worked as well as unused alumina. Polyamide and silica gel also gave satisfactory results.

Other adsorbent materials could be used, such as magnesol, silicic acid or adsorption alumina, and the like. However,

these materials are fine meshed and pack easily in the columns, so as to reduce flow through the columns. A pressure source would be necessary to obtain satisfactory flow through columns using these adsorbent materials. It is understood that the preferred adsorbents need not be used in a column. They can also be employed effectively in slurry-type removal polyphenols.

By the practice of the present invention, there is produced a reconstituted tobacco sheet using heat, which contains all the desirable portions of the original tobacco, and which is lighter in color than sheets made of untreated tobacco.

Various changes and modifications may be made in practicing this invention without departing from the spirit and scope thereof, and therefore, the invention is not to be limited, except as defined in the appended claims.

We claim:

1. The method of removing basic polyphenols from tobacco which comprises treating tobacco with water to create an aqueous solution in which the water soluble components of tobacco including basic polyphenols are extracted from the fibrous components thereof, separating the said aqueous solution containing the basic-polyphenols from the extracted tobacco, removing a material portion of said basic-

polyphenols from the extract solution by passing said solution into contact with an adsorbent material selected from the group consisting of activated alumina and polyamids, separating the solution from the adsorbent material, and finally combining the said extract solution from which the basic-polyphenols have been removed with the fibrous extracted tobacco so as to form an aqueous tobacco mixture which includes extracted water soluble components of the original tobacco minus a material amount of the basic-polyphenols originally therein.