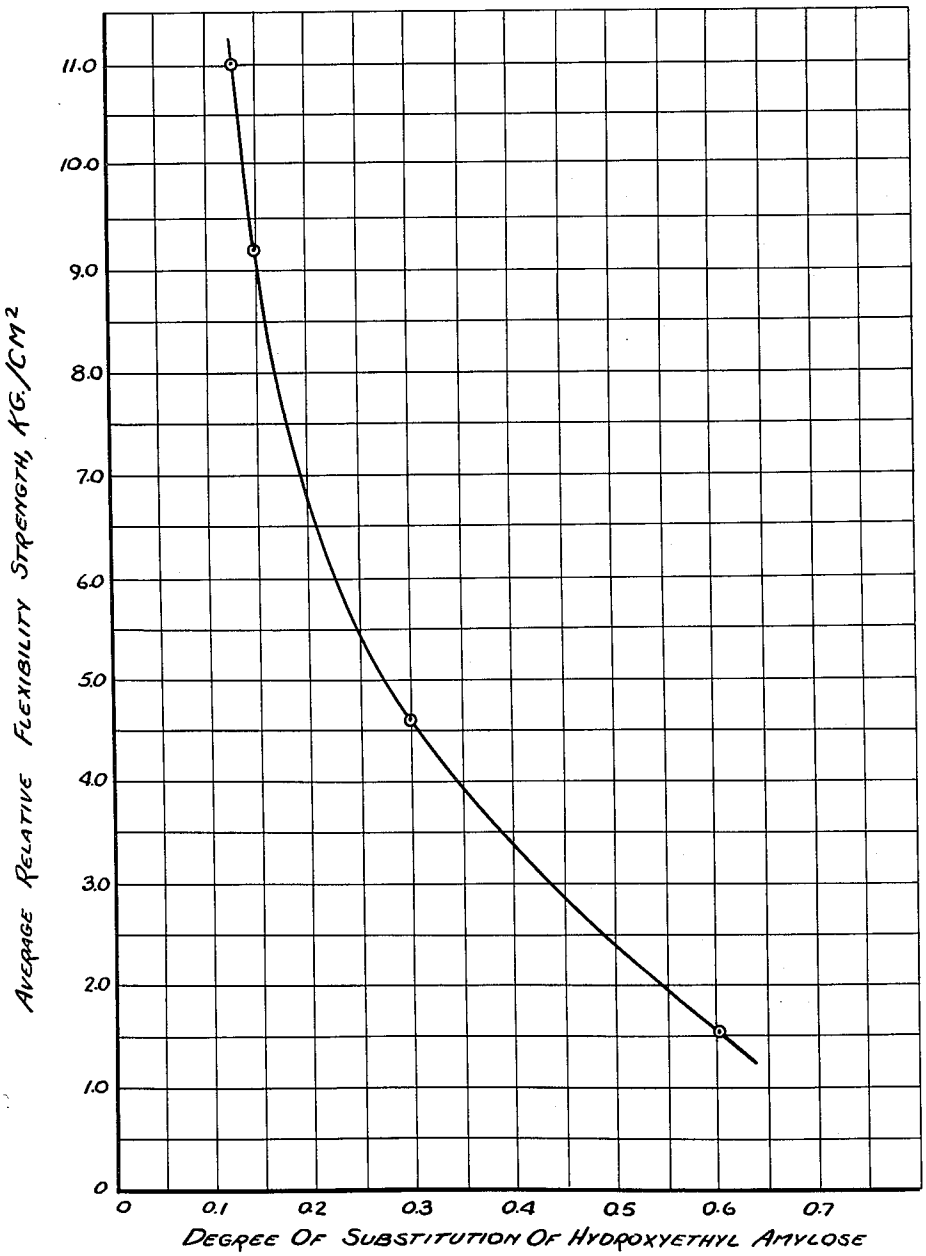
Nov. 21, 1961 M. M. SAMFIELD ET AL 3,009,835

NOVEL RECONSTITUTED TOBACCO COMPOSITIONS

Filed April 6, 1960

AVERAGE RELATIVE FLEXIBILITY STRENGTH VS.
DEGREE SUBSTITUTION OF HYDROXYETHYL AMYLOSE



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3,009,835
NOVEL RECONSTITUTED TOBACCO COMPOSITIONS

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21 Claims. (Cl. 131—17)

This invention relates to novel reconstituted tobacco compositions comprising primarily fragmented tobacco and a suitable binder for the tobacco fines. More particularly, this invention relates to novel reconstituted tobacco compositions which are suitable for blending with leaf tobacco in the manufacture of cigarettes and other smoking articles. Still more particularly, this invention relates to a novel process of preparing reconstituted tobacco compositions wherein the flexibility strength of the reconstituted tobacco sheets increases as the degree of substitution in the binder decreases within certain definite ranges and vice versa.

This application is a continuation-in-part application of our co-pending application Serial No. 851,010, filed November 5, 1959.

The history of the art of reconstituting tobacco fragments or fines has been concerned largely with the problem of forming the fragments of tobacco which inevitably result from the manufacture of cigarettes and other smoking articles from natural tobacco leaves, into a composition suitable for smoking. Preferably, the fragmented tobacco is sought to be combined with natural tobacco leaves in the manufacture of cigarettes and other smoking articles, in such a manner that the process of manufacture of the smoking articles are not complicated by the presence of such reconstituted tobacco, and that the inclusion of such reconstituted tobacco does not adversely affect the qualities of the resulting cigarettes and other smoking articles from the point of view of their acceptability to the consumer. In the formulation of these compositions it has been found that it is desirable to employ a suitable binder to combine with the tobacco fragments in such manner as to preferably produce sheets of material, primarily consisting of tobacco fragments, which on the one hand possess physical characteristics such that the sheets can be put through the normal manufacturing processes which leaf tobacco goes through in the production of cigarettes and other smoking articles, and on the other hand have taste and smoking characteristics such that the reconstituted tobacco may be added to the leaf tobacco without adversely affecting the smoking quality of the resulting article. However, one of the primary problems in producing reconstituted tobacco compositions is the selection of a suitable binder which will impart satisfactory flexibility strength to the reconstituted tobacco

sheets and yet will not adversely affect the smoking qualities of the tobacco.

It is therefore an object of this invention to provide novel reconstituted tobacco compositions containing suitable binders.

It is a further object of this invention to provide novel reconstituted tobacco compositions which are suitable for blending with leaf tobacco in the manufacture of smoking articles.

It is still a further object of this invention to provide a process for controlling the flexibility strength of the reconstituted tobacco compositions.

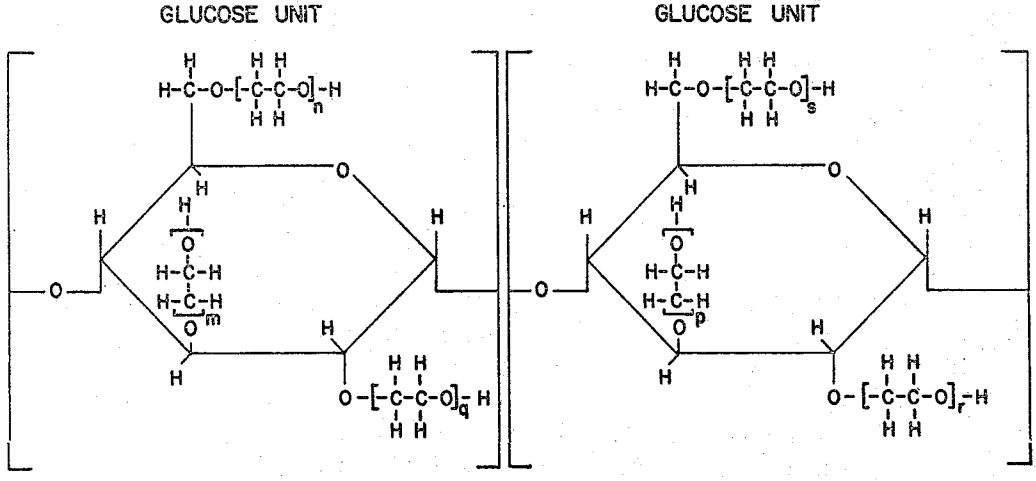
These and other objects will become apparent from the following detailed description.

Therefore, according to this invention there is provided a tobacco composition comprising fragmented tobacco and a binder consisting essentially of a small but effective amount of hydroxyethyl amylose having not more than 0.15 hydroxyethyl groups per glucose unit.

This invention also provides for a process for controlling the flexibility strength of reconstituted tobacco sheets comprising fragmented tobacco and a binder consisting essentially of hydroxyethyl amylose, said process comprising controlling the degree of substitution of hydroxyethyl radicals in said binder such that the flexibility strength of the reconstituted tobacco sheets increases as the degree of substitution of hydroxyethyl radicals decreases and the flexibility strength of the reconstituted tobacco sheets decreases as the degree of substitution of the hydroxyethyl radicals increases.

Thus we have found that there is a direct correlation between the degree of substitution of hydroxyethyl units in the amylose molecule and the flexibility strength of the tobacco sheets. We are therefore able to control the flexibility strength of the tobacco sheets within desired limits by controlling the degree of substitution of hydroxyethyl radicals in the amylose molecule used as a binder in the reconstituted tobacco compositions of this invention.

The hydroxyethyl amylose employed in the compositions of this invention is a straight-chain derivative of the amylose fraction of potato starch and is sold in the United States in a granular form, by the Stein Hall Company. The hydroxyethyl amylose of this invention has far more desirable solubility characteristics than amylose which is only soluble in water under autoclave conditions, and is soluble in water by cooking to 95° C. Furthermore, hydroxyethyl amylose is soluble in glycerine to the extent of about 35% by weight at 180° F. The degree of polymerization of the hydroxyethyl amylose molecule is about 800 to about 1,000 glucose units with a resulting average molecular weight of about 150,000. Hydroxyethyl amylose is believed to have the general formula:



The lettered subscripts represent the number of hydroxyethyl radicals in each side chain. While the degree of hydroxyethylation will vary over wide ranges, we have found it desirable to maintain the number of hydroxyethyl radicals per amylose molecule within the range of about 0.11 to about 0.15 hydroxyethyl groups per glucose unit.

In accordance with our invention we have found that the flexibility strength of the sheets increases with decreasing degree of substitution in a consistent manner shown in the accompanying drawing. It will be seen from this drawing that the flexibility strength of the tobacco sheets increases substantially in a consistent manner as the number of hydroxyethyl units per amylose moleculedecreases with the greatest increase in flexibility strength occuring within the range of about 0.11 to about 0.15 of hydroxyethyl radicals per glucose unit.

The hydroxyethyl amylose is present in the compositions of this invention in a range of about 1 to 20%, and preferably about 3 to 12% based on the total weight of the composition.

It is also within the purview of this invention to employ a fibrous material, preferably alpha cellulose, in an amount of about 1 to 10%, preferably about 4 to 8% based on the total weight of the composition to furnish reinforcing fibers to increase the strength of the tobacco sheets. This invention also envisions the use of a suitable humectant such as glycerine and/or propylene glycol in an amount of about 5 to 12% based on the total weight of the composition. We have found it particularly satisfactory to use a glycerine-propylene glycol mixture in a 4:1 ratio.

In preparing the compositions of this invention, the hydroxyethyl amylose maybe added to the tobacco fines in dry powdered form or it may be added in liquid form. In preparing the composition of this invention, it is essential that a small amount of moisture always be present in order to facilitate the preparation of tobacco leaves. Thus, when it is added in dry powdered form, it is mixed intimately with the tobacco fines to which mixture is preferably added the fibrous material in fibrous form. A small amount of water with or without a humectant is then added. If the hydroxyethyl amylose is added in liquid form, it may be mixed with a humectant, such as glycerine or a mixture of humectants such as a glycerine-propylene glycol mixture, diluted with water and then mixed with the powdered tobacco fines which may be in admixture with the fibrous material.

A detailed process for the preparation of smoking compositions is set forth in the U.S. Patent No. 2,708,175 (Reissue Patent No. 24,424) and the process described in this patent is quite suitable for the preparation of the compositions of this invention.

Essentially, this process involves conveying the dry tobacco to a hammer-mill where it is ground until about 20% to 35% of the resulting fragments will pass through a 140 mesh screen and until about 0.5% to 10% will remain in a 30-mesh screen. The proportions of coarse and fine tobacco particles produced thereby are variable at random within these limits.

When the hydroxyethyl amylose is added in dry form, it is mixed together with the ground tobacco and preferably the fibrous material. The liquid ingredients, i.e., water, with or without humectants such as glycerine and propylene glycol, are then added to the pre-mixed dry ingredients, i.e., the dry-ground tobacco, floc and hydroxyethyl amylose, with constant stirring. The resulting damp mix is then processed by any suitable apparatus adapted to apply a shearing action to the mixture while it is in moist condition. The shearing action is valuable because it tends to distribute the hydroxyethyl amylose throughout the mixture. Suitable apparatus includes, for example, six pairs of rollers, each pair having a space between them of about 0.004", with one of each pair of rollers revolving at such a speed that its rolling surface when acting upon the material being rolled between the pair, travels faster than the other rolling surface when so acting, thus producing a shearing or wiping action on the material of the mixture. The roller material will tend to stick to the faster moving surface, and should be removed therefrom by a doctor blade. The pairs of rollers may be conveniently placed so that the sheared material will drop by gravity from one pair of rollers to the next, and seriatim; and finally, the sheared material may be deposited on a moving belt or screen in the form of coherent and continuous damp sheets.

The sheets are then dried to a degree such that the moisture content approximately equals that of natural leaf tobacco and are then ready to be blended with natural leaf tobacco in the manufacture of cigarettes and other smoking articles. During the drying process the sheets lie in an undisturbed position upon the belt or screen upon which they have been deposited on leaving the last pair of shearing rollers. An overhead infrared electric resistance heater is suitable for the drying step. The tensile strength of the sheet produced is approximately the same as that of leaf tobacco, and similarly the thickness of the sheet of finished composition is about that of natural leaf tobacco while its ultimate moisture content, after drying, is about that of natural leaf tobacco when ready for blending, i.e., about 9% to 13% by weight of the total composition.

Alternatively, the hydroxyethyl amylose may be added to the dry mixture of dry-ground tobacco fragments which may contain the fibrous material, in the form of a solution. In this case, the dry-ground tobacco fragments and fibrous material are mixed together as a dry mix, while water with or without humectants and hydroxyethyl amylose are mixed together in the form of a solution. Thereafter, this solution is cooked for about 15 minutes at about 92° C. This hot solution or the solution after cooling may be added to the dry mix while the resultant mixture is constantly stirred. The resulting damp mix is then processed in a manner similar to that described above. In any case, a part of the liquid, whether water, a humectant or humectants, or both, may be supplied to the damp mix at the rollers, for instance, by moistening the latter with a measured part of the total liquid which is to be included in the mixture of dry and liquid mixes.

The following examples, without limiting in any way the scope of the invention which we claim, are specific embodiments of the invention showing illustrative proportions of the component materials which may be used.

In carrying out the following examples, tobacco compositions were prepared employing as the binder, hydroxyethyl amylose with different degrees of hydroxyethyl substitution. The resulting tobacco composition was then tested to determine the relative flexibility strength of the composition.

In carrying out the relative flexibility strength test, strips 16 mm. wide are cut from the equilibrated, finished tobacco sheets. Each strip is measured for thickness with a paper thickness gauge. The strips are then folded by hand without creasing. A 1,450 gram weight is then placed on the folded strip to crease it, the crease being made near the center of the strip.

The creased strips are then carefully opened up and carefully are lined between the jaws of a Louis Schopper tensile tester. The force, in kilograms, required to break the strip is then measured. Since the thickness and width of the strips are known, the relative flexibility strength of the specimens in kilograms per square centimeter cross section area of the strip may be calculated.

The term flexibility strength is used here to indicate the strength of the strips after creasing to distinguish the test from standard tensile strength measurements made on uncreased materials. We have found that the strength after creasing is a better measure of the ability of the

material to withstand the numerous handling operations in the cigarette factory.

Example No. 1

A dry mixture was prepared by blending the following ingredients:

454 grams dry ground tobacco
40 grams alpha cellulose
51 grams hydroxyethyl amylose with a degree of substitution (D.S.) of 0.60

A liquid mixture was prepared by blending the following ingredients:

375 ml. water
28 ml. glycerine
8 ml. propylene glycol

The liquid mixture was then added to the dry mixture while stirring constantly. The resulting damp mixture was then processed in a reconstituted tobacco plant similar to that described in U.S. Patent No. 2,708,175 (Reissue Patent No. 24,424) and U.S. Patent No. 2,845,933. The damp mixture was introduced directly into the rollers, bypassing the wet and dry mixers. The dried, finished sheet had the following composition (based on 12.5% moisture in the finished sheet):

Tobacco	74.4%	(8.5% moisture)
Alpha cellulose	6.5%	(8.0% moisture)
Hydroxyethyl amylose (D.S.=0.60)	8.3%	(12.0% moisture)
Glycerine	5.5%	(5.0% moisture)
Propylene glycol	1.4%	
Excess water	3.9%	
	100.0%	

Sheet thickness and relative flexibility determinations were made on the finished sheet, in the manner described above after the sheets were equilibrated for five days in a room maintained at 75° F. and 60% relative humidity. At an average sheet thickness of 0.15 mm., the above sheets had a relative flexibility strength of 1.5 kilograms per square centimeter (kgms./cm.^{2}).

Example No. 2

A dry mixture was prepared by blending the following ingredients:

454 grams dry ground tobacco
40 grams alpha cellulose
51 grams hydroxyethyl amylose with a degree of substitution (D.S.) of 0.30

A liquid mixture was prepared by blending the following ingredients:

375 ml. water
28 ml. glycerine
8 ml. propylene glycol

The liquid mixture was then added to the pre-mixed dry ingredients while stirring constantly. The resulting damp mixture was then processed as described in Example 1.

The finished sheet had the following composition (based on 12.5% moisture in the sheet):

Tobacco	74.4%	(8.5% moisture)
Alpha cellulose	6.5%	(8.0% moisture)
Hydroxyethyl amylose (D.S.=0.60)	8.3%	(12.0% moisture)
Glycerine	5.5%	(5.0% moisture)
Propylene glycol	1.4%	
Excess water	3.9%	
	100.0%	

Sheet thickness and relative flexibility determinations were made on the finished sheet after equilibrating in the manner described in Example 1.

At an average sheet thickness of 0.14 mm., the average relative flexibility strength of the sheets was determined to be 4.6 kgms./cm.^{2}.

Example No. 3

A dry mixture was prepared by blending the following ingredients:

454 grams dry ground tobacco
40 grams alpha cellulose
51 grams hydroxyethyl amylose with a degree of substitution (D.S.) of 0.15

A liquid mixture was prepared by blending the following ingredients:

375 ml. water
28 ml. glycerine
8 ml. propylene glycol

The liquid mixture was then added to the pre-mixed dry ingredients while stirring constantly. The resulting damp mixture was then processed as described in Example 1.

The finished sheet had the following composition (based on 12.5% moisture in the finished sheet):

Tobacco	74.4%	(8.5% moisture)
Alpha cellulose	6.5%	(8.0% moisture)
Hydroxyethyl amylose (D.S.=0.15)	8.3%	(12.0% moisture)
Glycerine	5.5%	(5.0% moisture)
Propylene glycol	1.4%	
Excess water	3.9%	
	100.0%	

Sheet thickness and relative flexibility determinations were made on the finished sheet after equilibration in the manner described in Example 1. At an average sheet thickness of 0.14 mm., the average relative flexibility strength was found to be 9.2 kgms./cm.^{2}.

Example No. 4

A dry mixture was prepared by blending the following ingredients:

454 grams dry ground tobacco
40 grams alpha cellulose
51 grams hydroxyethyl amylose with a degree of substitution (D.S.) of 0.13

A liquid mixture was prepared by blending the following ingredients:

375 ml. water
28 ml. glycerine
8 ml. propylene glycol

The liquid mixture was then added to the pre-mixed dry ingredients while stirring constantly. The resulting damp mix was then processed into sheets as described in Example 1.

The finished sheet had the following composition (based on 12.5% moisture in the finished sheet):

Tobacco	74.4%	(8.5% moisture)
Alpha cellulose	6.5%	(8.0% moisture)
Hydroxyethyl amylose (D.S.=0.13)	8.3%	(12.0% moisture)
Glycerine	5.5%	(5.0% moisture)
Propylene glycol	1.4%	
Excess water	3.9%	
	100.0%	

Sheet thickness and relative flexibility determinations were made of the finished sheet after equilibration in the manner described in Example 1.

At an average sheet thickness of 0.14 mm., the average relative flexibility strength was found to be 11.0 kgms./cm.^{2}.

Having thus provided a written description of the invention along with specific examples thereof, it should be understood that no undue limitations or restrictions are

to be imposed by reason thereof, but that the present invention is defined by the appended claims.

We claim:

1. A composition of matter suitable for smoking comprising fragmented tobacco and a binder consisting essentially of a small but effective amount of hydroxyethyl amylose having not more than 0.15 hydroxyethyl radicals per glucose unit.

2. A composition of matter suitable for smoking comprising fragmented tobacco and a binder consisting essentially of hydroxyethyl amylose having between about 0.11 to 0.15 hydroxyethyl groups per glucose unit.

3. A composition of matter suitable for smoking consisting by weight of a minor proportion, about 1 to 20%, of hydroxyethyl amylose, having not more than 0.15 hydroxyethyl radicals per glucose unit, approximately 9 to 13% of moisture and the balance essentially all dry-ground tobacco with the individual finely divided fragments thereof cohered together so as to have when formed in sheets of about the thickness of natural leaf tobacco, a tensile strength approximately equal to the tensile strength of such leaf tobacco.

4. A composition of matter suitable for smoking consisting by weight of a minor proportion, about 1 to 20%, of hydroxyethyl amylose having between about 0.11 to 0.15 hydroxyethyl radicals per glucose unit, about 1 to 10% of a fibrous material, approximately 9 to 13% moisture and the balance essentially all dry-ground tobacco with the individual finely divided fragments thereof cohered together so as to have, when formed in sheets of about the thickness of natural leaf tobacco, a tensile strength approximately equal to the tensile strength of such leaf tobacco.

5. A composition according to claim 4 wherein said fibrous material is alpha cellulose.

6. A composition according to claim 4 wherein said hydroxyethyl amylose is present in an amount of about 3 to 12% by weight.

7. A composition according to claim 4 wherein said fibrous material is present in an amount of about 4 to 8% by weight.

8. A composition of matter suitable for smoking consisting by weight of a minor proportion, about 1 to 20%, of hydroxyethyl amylose having about 0.11 to about 0.15 hydroxyethyl radicals per glucose unit, and moisture comprising water in an amount between 9% to 13% by weight of the total composition, and the balance essentially all dry ground tobacco with the individual finely divided fragments thereof cohered together so as to have, when formed in sheets of about the thickness of natural leaf tobacco, a tensile strength approximately equal to the tensile strength of such leaf tobacco.

9. A composition of matter suitable for smoking consisting by weight of a minor proportion, about 1 to 20%, of hydroxyethyl amylose having about 0.11 to about 0.15 hydroxyethyl radicals per glucose unit, about 1 to 10% of a fibrous material, at least one humectant in an amount not exceeding 12%, and the balance essentially all dry ground tobacco, with the individual finely divided fragments thereof cohered together so as to have, when formed in sheets of about the thickness of natural leaf tobacco, a tensile strength approximately equal to the tensile strength of such leaf tobacco.

10. A composition according to claim 9 wherein said fibrous material is alpha cellulose.

11. A composition according to claim 9 wherein said humectants are selected from the group consisting of glycerine propylene glycol, and mixtures thereof.

12. A composition according to claim 9 wherein said hydroxyethyl amylose is present in an amount of about 3 to about 12% by the weight.

13. A composition according to claim 9 wherein said fibrous material is present in an amount of about 4 to 8% by weight.

14. A composition of matter suitable for smoking consisting by weight of a minor proportion, about 1 to 20%, of hydroxyethyl amylose having about 0.11 to about 0.15 hydroxyethyl units per glucose unit, about 9 to 13% of water, a humectant in an amount not exceeding about 12%, and the balance essentially all dry ground tobacco with the individually finely divided fragments thereof cohered together so as to have, when formed in sheets of about the thickness of natural leaf tobacco, a tensile strength approximately equal to the tensile strength of such leaf tobacco.

15. A composition according to claim 14 wherein said hydroxyethyl amylose is present in an amount of about 3 to 12% by weight.

16. A composition according to claim 14 wherein said fibrous material is present in an amount of about 4 to 8% by weight.

17. A composition according to claim 14 wherein said humectants are selected from the group consisting of glycerine, propylene glycol, and mixtures thereof.

18. A process for controlling the flexibility strength of reconstituted tobacco sheets comprising fragmented tobacco and a binder consisting essentially of hydroxyethyl amylose, said process comprising blending fragmented tobacco with a small but effective amount of hydroxyethyl amylose having not more than 0.15 hydroxyethyl radicals per glucose unit, forming sheets of reconstituted tobacco from the blend of fragmented tobacco and hydroxyethyl amylose and then drying said sheets to approximately the moisture content of natural leaf tobacco, the flexibility strength of the reconstituted tobacco sheet being controlled such that the flexibility strength of the reconstituted tobacco sheet increases as the degree of substitution of hydroxyethyl radicals in said binder decreases and the flexibility strength of the reconstituted tobacco sheet decreases as the degree of substitution of the hydroxyethyl radicals in said binder increases.

19. A process for controlling the flexibility strength of reconstituted tobacco sheets comprising fragmented tobacco and a binder consisting essentially of hydroxyethyl amylose, said process comprising blending fragmented tobacco with a small but effective amount of hydroxyethyl amylose having from about 0.11 to about 0.15 hydroxyethyl radicals per glucose unit, forming sheets of reconstituted tobacco from the blend of fragmented tobacco and hydroxyethyl amylose and then drying said sheets to approximately the moisture content of natural leaf tobacco, the flexibility strength of the reconstituted tobacco sheet being controlled such that the flexibility strength of the reconstituted tobacco sheet increases as the degree of substitution of hydroxyethyl radicals in said binder decreases and the flexibility strength of the reconstituted tobacco sheet decreases as the degree of substitution of the hydroxyethyl radicals in said binder increases.

20. A process for controlling the flexibility strength of a composition suitable for smoking consisting by weight of a minor proportion, about 1 to 20% of hydroxyethyl amylose having not more than 0.15 hydroxyethyl radicals per glucose unit, approximately 9 to 13% of water, a humectant in amount not exceeding about 12%, and the balance essentially all dry ground tobacco with the individual finely divided fragments thereof cohered together so as to have, when formed in sheets of about the thickness of natural leaf tobacco, a tensile strength approximately equal to the tensile strength of such leaf tobacco, said process comprising blending together said hydroxyethyl amylose, water, humectant and finely divided tobacco fragments, forming sheets of reconstituted tobacco from these ingredients and then drying said sheets to approximately the moisture content of natural leaf tobacco, the flexibility strength of the reconstituted tobacco sheet being controlled such that the flexibility strength of the smoking composition increases as the degree of substitution of hydroxyethyl radicals in said hydroxyethyl amylose decreases and the flexibility strength of the smoking composition decreases as the degree of substitution of the

hydroxyethyl radicals in said hydroxyethyl amylose increases.

21. A process for controlling the flexibility strength of reconstituted tobacco sheets consisting by weight of a minor proportion, about 1 to 20% of hydroxyethyl amylose, having not more than 0.15 hydroxyethyl radicals per glucose unit, approximately 9 to 13% of water, a humectant in an amount not exceeding about 12%, and the balance essentially all dry ground tobacco with the individual finely divided fragments thereof cohered together so as to have, when formed in sheets of about the thickness of natural leaf tobacco, a tensile strength approximately equal to the tensile strength of such leaf tobacco, said process comprising blending together said hydroxyethyl amylose, water, humectant and finely divided tobacco fragments, forming sheets of reconstituted tobacco from the aforesaid ingredients and then drying said sheets to approximately the moisture content of natural leaf tobacco, the flexibility strength of the reconstituted tobacco sheet being controlled such that by controlling the degree of substitution of hydroxyethyl radicals in said hydroxyethyl amylose in accordance with the curve shown in FIG. 1 of the accompanying drawing, the flexibility strength of the reconstituted tobacco sheet increases as the degree of substitution of hydroxyethyl radicals decreases and the flexibility strength of the reconstituted tobacco sheet decreases as the degree of substitution of the hydroxyethyl radical increases.

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UNITED STATES PATENT OFFICE

CERTIFICATION OF CORRECTION

Patent No. 3,009,835 November 21, 1961

Max M. Samfield et al.

It is hereby certified that error appears in the above numbered patent requiring correction and that the said Letters Patent should read as corrected below.

Column 5, line 66, for "(D.S.=0.60)" read -- (D.S.=0.30) --.

Signed and sealed this 10th day of April 1962.

(SEAL)
Attest:

ERNEST W. SWIDER
Attesting Officer

DAVID L. LADD
Commissioner of Patents