United States Patent

Davis

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[54] METHOD FOR MAKING A TOBACCO SUBSTITUTE COMPOSITION

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Related U.S. Application Data

[63] Continuation-in-part of Ser. No. 473,916, July 21, 1965, which is a continuation-in-part of Ser. No. 32,211, May 27, 1960, abandoned, which is a continuation-in-part of Ser. No. 9,763, Feb. 9, 1960, abandoned.

[52] **U.S. Cl.** **131/2,** 131/17

[51] **Int. Cl.** **A24b 03/14**

[58] **Field of Search** 162/139, 157, 187; 131/2, 17, 131/15, 140

[56] **References Cited**

UNITED STATES PATENTS

3,145,717	8/1964	Osborne et al.	131/140
3,195,245	7/1965	Clarke	131/140
2,576,021	11/1951	Koree	131/140
2,738,791	3/1917	Schur et al.	162/139
2,987,434	6/1961	Murray et al.	162/157 X
3,003,895	10/1961	Grunwald	131/17
3,112,754	12/1963	Diaz	131/2
3,009,836	11/1961	Samfield et al.	131/17

OTHER PUBLICATIONS

Stedman, R. L. " The Chemical Composition of Tobacco and Tobacco Smoke" U.S. Dept. of Agriculture Publication 19118 p. 190 cited

Shmuk (text) " The Chemistry and Technology of Tobacco" pub. by Pishchepromizdat (Moscow) 1953 p. 497– 499 & p. 594– 595

" Smoking and Health" Report of the Advisory Committee to the Surgeon General of the Public Health Service, Public Health Service Publication No. 1103 (1964)

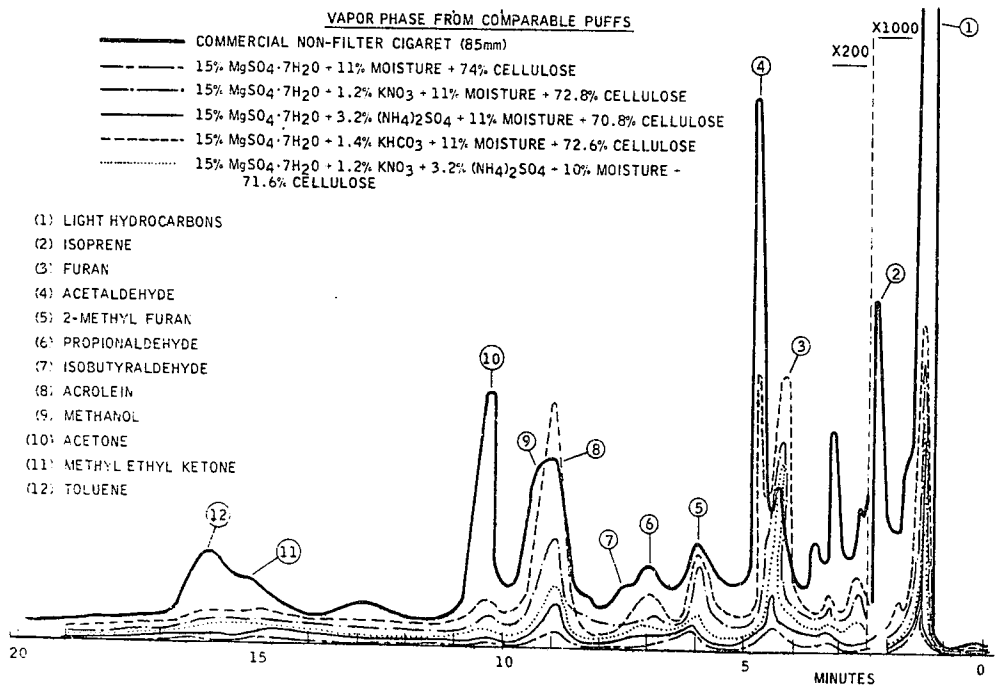
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[57] **ABSTRACT**

A method of making a tobacco substitute material is disclosed which involves the use of fibrous woodpulp which contains at least 90 percent of alpha cellulose. The selected wood pulp of high alpha cellulose count is lightly beaten to a Canadian Standard Freeness of between 400–700 ml. and is then combined with a nontoxic combustion modifier and formed into a sheet having a density of 12–35 lbs./ft.^{3}. The combustion modifiers useable are the potassium, sodium and magnesium sulphates, magnesium and potassium chlorides, sodium, potassium, magnesium and ammonium carbonates and bicarbonates, potassium nitrate, ferric oxide, ferric hydroxide, alumina, magnesium gluconate, citrate, citrate and acetate and gluconic acid. The metallic compounds utilized are preferably used in their hydrated form.

5 Claims, 5 Drawing Figures



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SHEET 1 OF 4

Fig. 1.

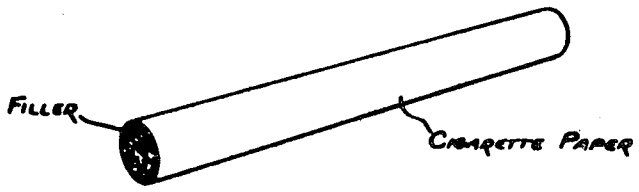
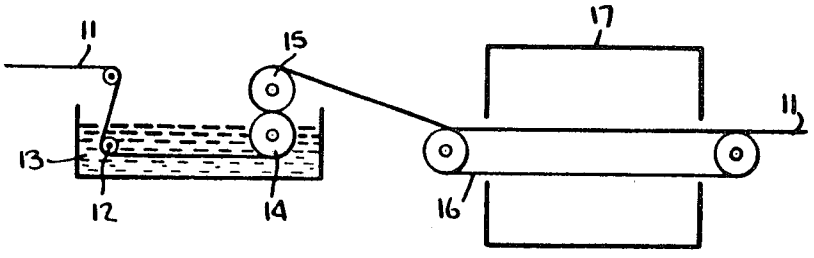


Fig. 2.



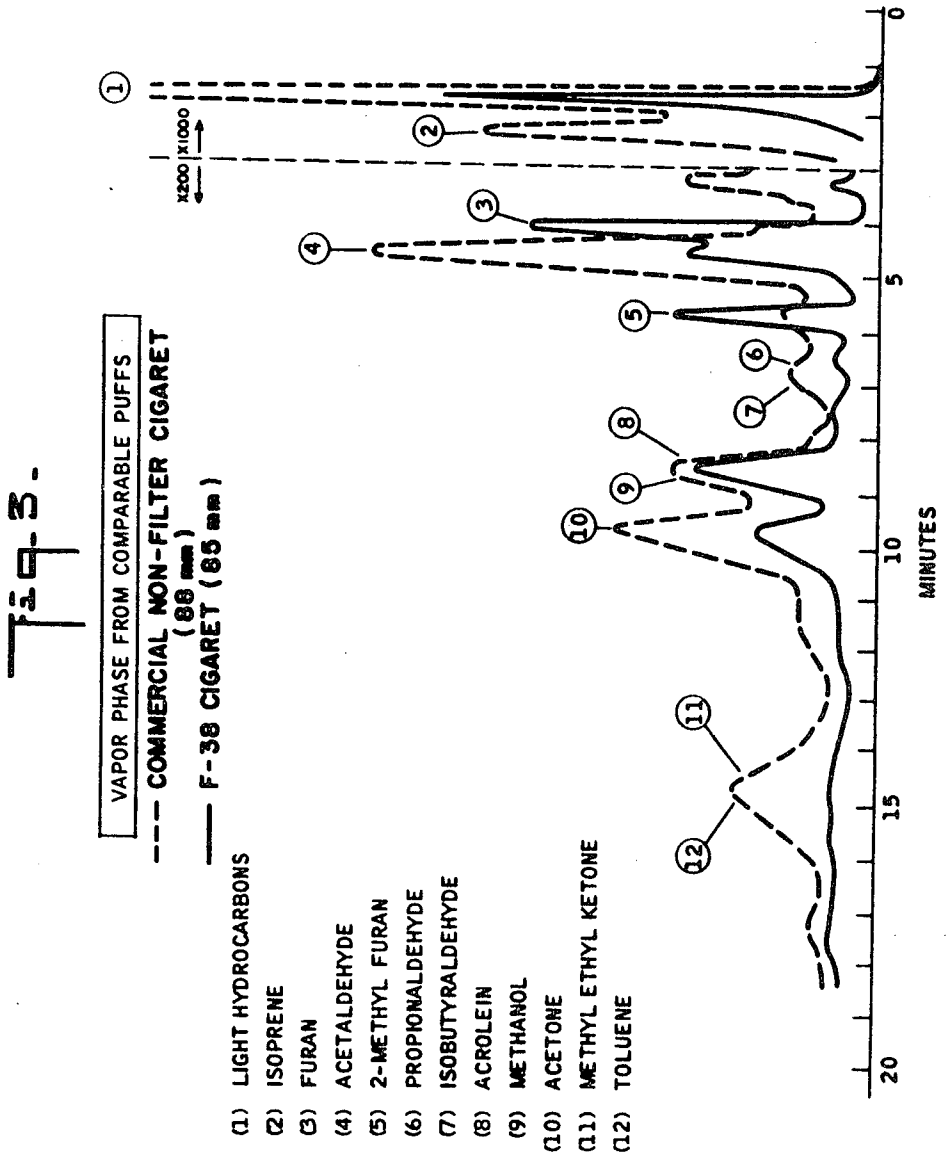
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Fig. 3.



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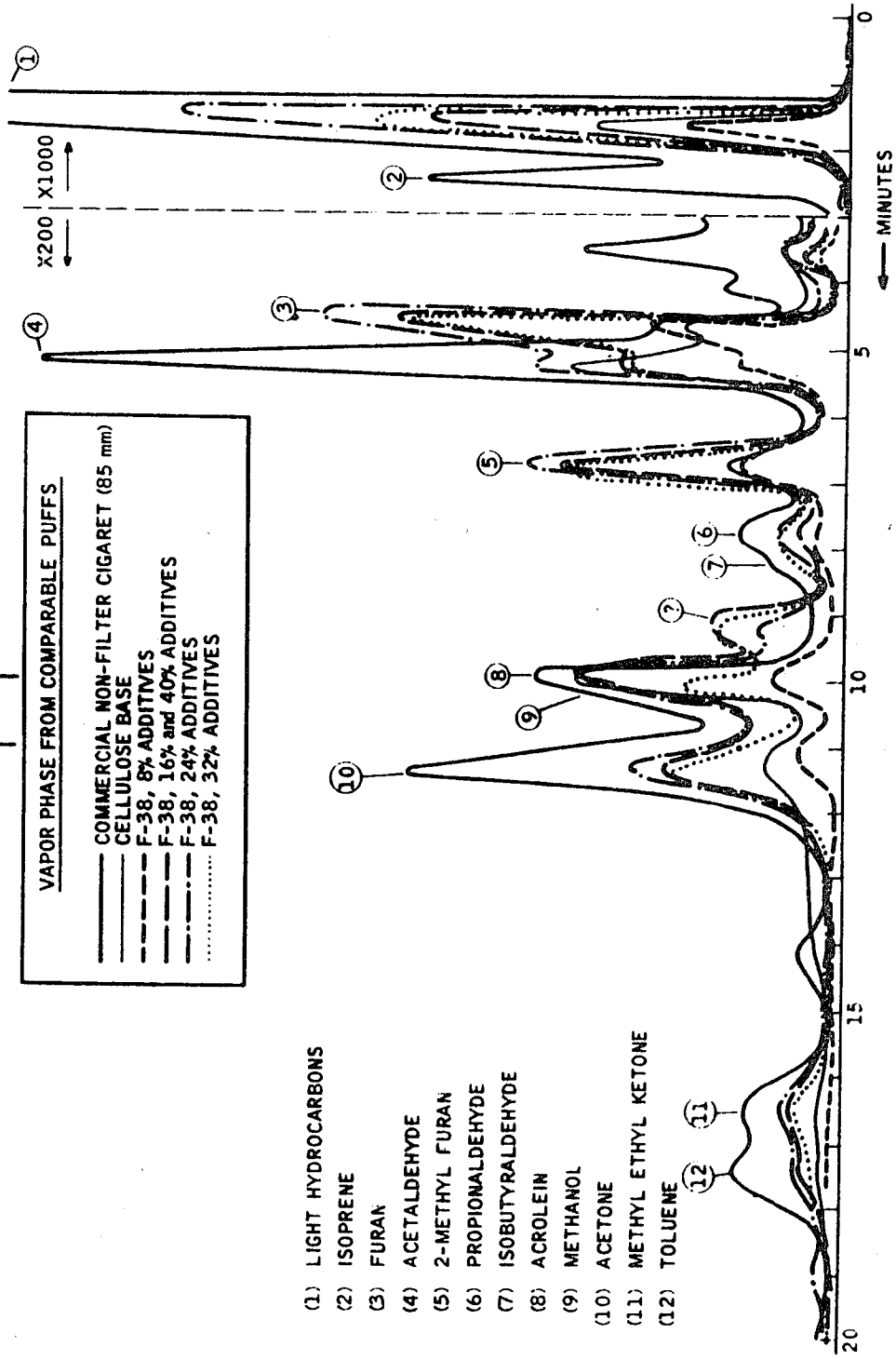
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Fig. 4.



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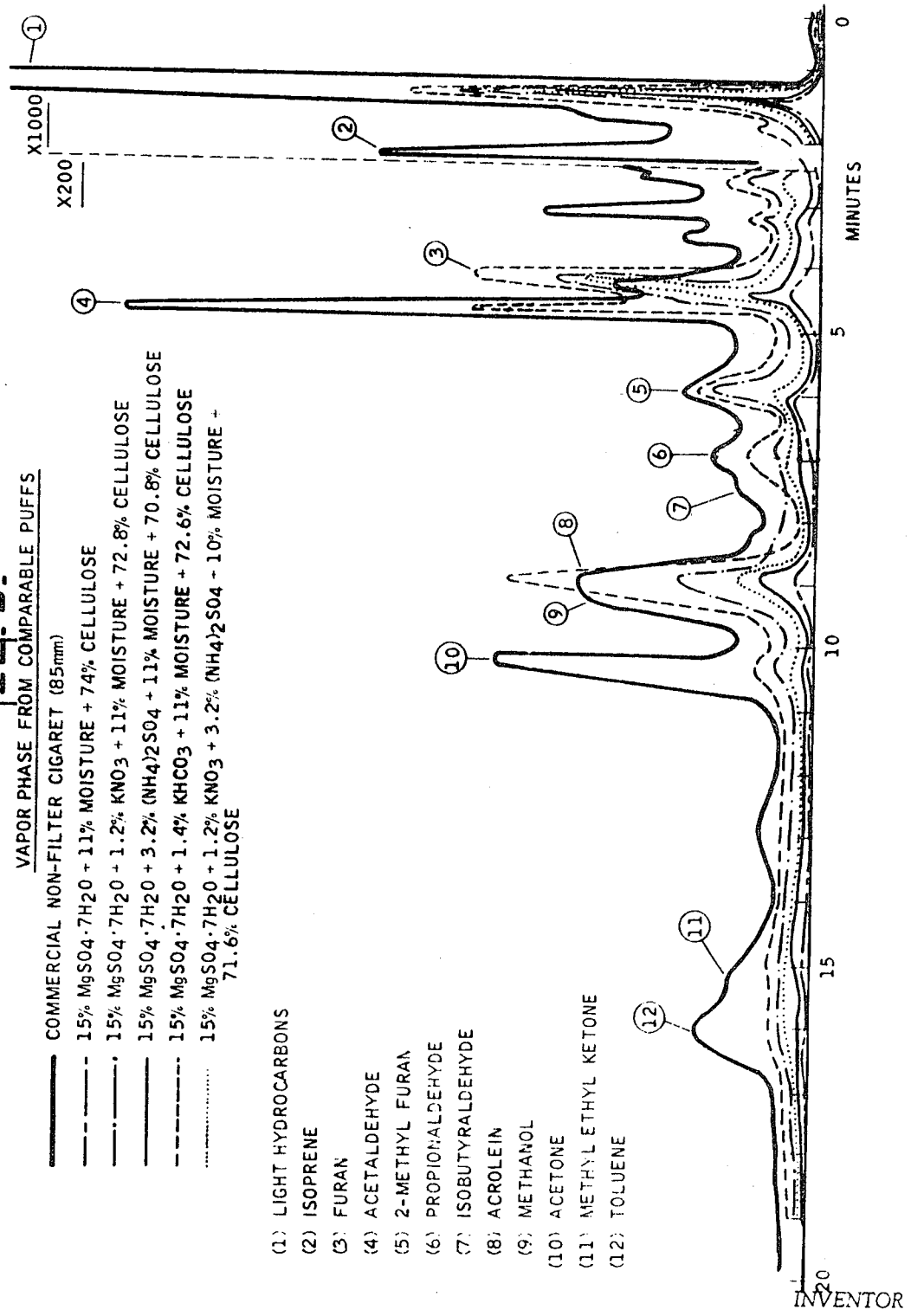
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Fig. 5.

VAPOR PHASE FROM COMPARABLE PUFFS



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METHOD FOR MAKING A TOBACCO SUBSTITUTE COMPOSITION

The present application is a continuation-in-part of Ser. No. 473,916, filed July 21, 1965, which is a continuation-in-part of application Ser. No. 32,211, filed May 27, 1960, now abandoned, which in turn is a continuation-in-part of application Ser. No. 9,763, filed Feb. 19, 1960, and now abandoned.

This invention relates broadly to smoking materials, and is more particularly concerned with cigarettes.

It is an object of this invention to provide a novel smoking material particularly suitable as a replacement, in whole or in part, for tobacco in cigarettes.

Another object is the provision of a cigarette or like smoking materials containing cellulose and additives which change the burning characteristics of the cellulose to produce a pleasing smoke.

An additional object of the present invention is to provide a smoking material for the manufacture of cigarettes which permits the control of, and more particularly the reduction of the smoke condensate from such cigarettes, as compared with that which is obtained from conventional all-tobacco cigarettes.

A further object of this invention is the provision of a smoking material which when smoked as a cigarette in a conventional smoking cycle produces a smoke condensate with a substantially lower polycyclic aromatic hydrocarbon content than that which is obtained from conventional tobacco cigarettes, and more particularly a smoke condensate whose content of 3,4-benzpyrene (a carcinogen) is lower than that which is commonly obtained from conventional all-tobacco cigarettes.

An additional object of the present invention is to provide an economical, quickly produced and readily available smoking material which can also be blended with tobacco to produce a more economical cigarette whose smoke contains a reduced amount of condensable material and a reduced amount of 3,4-benzpyrene, as compared with a conventional all-tobacco cigarette.

A still further object of the invention lies in the provision of a method of producing a smoking material at low density, featuring beating woodpulp to a Canadian Standard Freeness of between about 400 to 700 ml., whereby the resultant sheet has a relatively low density, resulting when blended with a hydrated metal compound and formed into a smoking material, a remarkably low tar content.

Other objects of this invention will be apparent from the following detailed description, particularly when taken in connection with the accompanying drawings.

In the drawings:

FIG. 1 is a view of my cigarette;

FIG. 2 is a schematic portrayal of an exemplary process for applying the additives to the paper, followed by a drying step; and

FIGS. 3, 4 and 5 are charts showing the results of vapor phase determinations on conventional nonfilter cigarettes and cigarettes produced in accordance with the present invention.

Cellulose, when used as a tobacco substitute in a cigarette, burns rapidly with a characteristic yellow flame and an odor like that of burning rags. When a hydrated metal compound exemplified by magnesium sulfate, magnesium acetate, magnesium citrate and the like is incorporated, e.g., in amount of 10 percent, into the cellulose the burning rate approaches that found in ordinary cured tobacco leaves, the odor of the side stream and the taste of the main stream are much more pleasant, the color of the flame front or coal is different and the ash is dense and self-supporting (and much lighter in color than the ash of the unmodified cellulose cigarette).

In accordance with one aspect of this invention, I have found that the amount of tar produced in the smoking of cigarettes containing the mixture of cellulose and magnesium sulfate and related compounds may be reduced by using as the cellulose a lightly beaten woodpulp of high alpha cellulose content. Beating of pulp is a conventional operation in the making of paper. It involves mechanically working the fibers

in the presence of water, usually on a machine called a beater, such as the Hollander, Horne, Niagara, Jones, Betram, Victory, Vortex and rod mill beaters described in Pulp and Paper manufacture, Vol. 2, Preparation of Stock for Paper Making, edited by J. N. stephenson, first edition, published in 1951 by McGraw-Hill Book Company, Inc. pages 200–212. I have obtained very good results by beating the pulp to such an extent that it will form a paper which withstands mechanical processing, particularly processing in the wet state (e.g., passage through an aqueous dipping bath without disintegrating), and stopping the beating before the pulp has lost most of its "freeness." Generally speaking, the beating should be stopped when the freeness of the pulp is in the range of 400–700 ml., preferably about 500–600 ml. (as measured in accordance with TAPPI Standard T227 *m*–50, which uses a Canadian Standard Freeness tester of the general type shown at pages 238 and 239 of Stephenson Pulp and Paper Manufacture Vol 2 (previously cited). The beaten pulp is then formed into paper, as by depositing the pulp on any suitable paper-forming screen e.g., a Fourdrinier screen or Valley Mold), pressing and drying. Suitably, the rate of deposition and amount of pressing are such that the resulting dried paper has an average thickness in the range of about 3 to 10 mils. Sheet thickness is inversely proportional to sheet bulk density, for fixed areal density. Tests have shown that relatively low squeeze pressures produce a lower density sheet and a lower total condensate when formed into a cigarette and smoked.

Best results are obtained when there are also present substances which modify the burning characteristics of cigarettes comprising cellulose and hydrated magnesium sulfate. It is especially desirable to add inorganic compounds, e.g., salts, which help sustain the burning of the cigarette, particularly during the resting period of the usual smoking cycle, when oxygen is not being forcibly drawn into the burning zone. Examples of such burning sustainers are potassium chloride, magnesium chloride, potassium sulfate, potassium nitrate, and ferric oxide and hydroxide.

Certain burning sustainers have a particularly desirable additional effect, in that they increase the rate of burning; examples of such materials are the carbonates and bicarbonates, e.g., potassium, sodium, magnesium and ammonium carbonate or bicarbonate. It is believed that the potassium, sodium and ammonium carbonates react with part of the magnesium sulfate to form magnesium carbonate during the application of these compounds. The presence of the carbonate during burning is believed to cause an expansion of the burning zone and is believed to permit more air to penetrate to all parts of the burning particles, thus promoting more complete combustion. The presence of the carbonate also causes an expansion of the ash so that the ash is more porous. In addition, the use of carbonates materially decreases the acid content of the smoke. Potassium carbonate is particularly suitable since it not only is a carbonate, but also supplies potassium which is very effective to maintain the burn. Other burning sustainers which increase the rate of burning, when used in suitable concentrations, are sodium sulfate, potassium nitrate and similar salts.

The flavoring of the cigarette may be improved by the incorporation of flavoring agents. These may be of the type used commercially for the flavoring of tobacco such as menthol, tonka bean, powdered deer tongue, licorice or the proprietary mixtures known as "Alva: G–1, G–6, G–16, G–189, G–32 and G–327"supplied by Van Ameringen-Haebler, Inc., Division of International Flavors and Fragrances, Inc. Vanillin has an excellent effect on the flavor and odor of the cigarette, making these considerably milder. The usual household vanilla extract may be incorporated in the cellulose for this purpose.

It is also desirable to have a humectant present in order to prevent the tobacco substitute from drying out unduly. Any of the usual humectants used in cigarette manufacture may be employed; for example, sorbitol, which is preferred, or other humectant polyhydric alcohol, e.g., glycerol, may be used. The moisture content of the tobacco substitute may be within

the range of about 5 to 25 percent, preferably about 10 to 15 percent, exclusive of the moisture present as water of hydration.

I may also incorporate nicotine or other alkaloid in the synthetic smoking material. The presence of the alkaloid gives an increased feeling of satisfaction to the users of the synthetic smoking material. The nicotine may be supplied, for example, as such or in the form of its sulfate, or citrate or citrate-sulfate, malonate or malate. Preferably, the amount of nicotine used is insufficient, in itself, to impart any pronounced color to the synthetic smoking material; its effect in this respect, even if impure colored nicotine is used, is merely a slight tinting of the product.

I may also incorporate an ammonium compound. Thus, compounds such as ammonium sulfate, ammonium carbonate, ammonium persulfate and ammonium perchlorate act to release ammonia and thus raise the pH of the side stream. The presence of the ammonia thus formed is also believed to have an effect in inhibiting the production of undesirable 3,4-benzpyrene during smoldering. The use of ammonium compounds also results in the formation of whiter ash.

To enhance the appearance of the tobacco substitute, a coloring agent may be incorporated. Although the color is preferably a tobaccolike brown or yellow-brown, colors such as purple and pink may be used. Among the colors are those certified by the Food and Drug Administration such as F.D. and C. (Food, Drug & Cosmetic Act) Yellow No. 5; F.D. and C. Chocolate Brown, New Shade B; C.I. 17590; **Brown PG**; 20170 **Brown Y**; 30045 Yellow-Brown K.

This invention makes it possible to produce a cigarette which yields a smoke containing a very small amount of condensate (tar), and in which the burning temperature during smoking is in the critical range of about 750° to 900° C. for only a very small fraction of the total smoking time. It is known that burning temperatures in the aforesaid critical range promote the formation of certain polycyclic aromatic compounds, such as 3,4-benzpyrene, which are carcinogenic. In conventional cigarettes the burning temperature is in the critical range for much longer times. This is readily demonstrated by standard burning temperature profiles. Burning temperature profiles of cigarettes are obtained, in a standard manner, by continuously recording the temperature at a fixed point of the cigarette while the cigarette is being smokes in a standard manner, as described by G. P. Touey and R. C Mumpower, Tobacco 144, 18 (1957). More particularly, a platinum/87 percent Pt. 13 percent Rh fine wire thermocouple is placed at a point 25 mm. back from that end of the cigarette which is to be lit. The junction of the thermocouple is at the longitudinal axis of the cigarette while the two fine wires leading to the junction are at right angles to that axis and pass, through the cigarette paper, on opposite sides of the junction, the paper being suitably sealed about the wires to prevent entrance of air and movement of the thermocouple. The cigarette is smoked by means of a machine which draws air through the lit cigarette and then expels the indrawn gas and smoke into the atmosphere. More particularly, in the work described in the atmosphere. More particularly, in the work described in this application, the machine has a piston operating in a cylinder connected to the cigarette through a trap cooled by a dry ice-acetone mixture. The volume displaced by movement of the piston in the cylinder is 35 ml., and the cylinder is maintained at room temperature (25° C.). The smoking machine is operated on a 30 second cycle, in which air is drawn through the lit cigarette and into the cylinder, by movement of the piston, for 2 seconds; the gas in the cylinder is then expelled to the room, by movement of the piston for 2 seconds; and the apparatus then remains stationary for 26 seconds, so that the cigarette is in the resting stage for 28 seconds. With this arrangement a standard tobacco cigarette, 70 mm. long and 22–26 mm. in circumference and having no filter tip, will smoke down to a 25 mm. butt in about 12 to 14 cycles. Alternatively, I may use a 60 second cycle, having a resting stage of 58 seconds, with similar two second intake strokes and two second exhaust strokes.

The instrument used for measuring temperature must have a high speed of response. For this purpose I have used a Sargent Multi-range Recorder (Cat. No. S72151, E. H. Sargent & Co.) having a temperature response characteristic of 1¼ seconds for the full scale of recording, the setup being such that the full scale of recording covers 1,150° C.

In a conventional tobacco cigarette, during each puff of the smoking cycle (that is, during the 2-second intake of the gas) the temperature at the thermocouple junction rises as the cigarette burns back towards the junction. The temperature at the thermocouple drops slightly after each puff, at the beginning of each rest period, but when the temperature during the puff rises into the critical temperature range the resting temperature stays within the critical range. There is a long period of time when the temperature in a substantial portion of the cigarette, extending back from the lit end, is in the critical range. The temperature profiles of cigarettes of typical tobacco substitutes made in accordance with this invention are in sharp contrast to those of typical tobacco cigarettes. The temperature drops off sharply after each puff and even after the puff temperature has come into the critical temperature range, the resting temperature is well outside that range. In addition, only a very small part of the cigarette is at a temperature in the critical range during the smoking cycle. For example, in the cycle just before the cigarette burns back to the thermocouple, the thermocouple temperature is in the critical range for less than 50 percent, preferably less than 10 percent, and more preferably less than 5 percent, of the time. The initial slope of the curve descending from each peak temperature within the critical range is much steeper than that of the tobacco cigarette, being considerably steeper than −3,000° C./min., usually sharper than about −6,000° C./min. and preferably sharper than about −9,000° C./min.

The slope of the curve ascending to each peak temperature within the critical range is also much higher than that of the tobacco cigarette, being well above 3,000° C./min., usually sharper than about 5,000° C./min. and preferably sharper than 6,000° C./min. The difference between the peak temperature and the next succeeding minimum of the resting temperature during the two time cycles starting one puff before the highest temperature puff is usually at least about 200° C., preferably at least about 250° C.

Acceptable results are attained by the use of hydrated magnesium sulfate as the hydrated metal compound to favorably modify the burning rate of the cellulose tobacco substitute; this compound markedly improves the flavor and odor of burning cellulose. Other hydrated metal compounds are sodium sulfate, hydrated alumina, magnesium citrate, magnesium acetate and calcium tartrate. Mixtures of compounds may be used; thus magnesia and sodium sulfate may be applied so as to form magnesium sulfate in situ. Similarly, magnesium chloride and potassium sulfate may be applied for the same purpose. Also, gluconic acid or magnesium gluconate are effective substitutes. It will be understood that, like hydrated magnesium sulfate, the other hydrates may be used with burning sustainers, flavoring agents, humectants, nicotine, ammonium compounds, or other modifying agents, individually or in any combination.

The proportion of the burning rate controller of hydrated metal compounds mentioned is desirably in the range of about 10 to 15 percent, based on the total air-dry weight of the tobacco substitute. The proportion of the salt which is a burning sustainer is desirably in the range of about ½ to 5 percent, preferably about 1 to 4 percent, based on the total air-dry weight of tobacco substitute. Advantageously, the proportion of burning sustainer is such as to insure that the cigarette remains lit during the resting portion of the cycle, but should not be so high that the cigarette will flare or flame during the puffing portion of the cycle, and preferably not so high as to seriously impair the taste or odor. In general, the precise amount of burning sustainer for best results will depend on the physical form of the cellulose and the amount of hydrated $MgSO_4$ (or other compound which lowers the burning rate). When potassium carbonate or bicarbonate is used as a burning

sustainer the proportion for best results generally falls in the range of about ½ to 2 percent, preferably about 1 to 1.8 percent, based on the total air-dry weight of the tobacco substitute. For other carbonates, proportions providing the same content of CO_3 are desirable.

The amount of flavoring agent is desirably in the range of about 0.001 to 10 percent with the best results usually obtained with the proportions within the range of about 1/10 to 1/12 percent, based on the total air-dry weight of the product. The proportion of humectants is desirably in the range of about 0.001 to 10 percent, preferably about 1½ to 2½ percent, based on the total air-dry weight of the tobacco substitute. The proportion of nicotine, when used, is desirably in the range of up to about 2 percent, preferably about ½ to 1½ percent, based on the weight of the cellulose. The amount of the ammonium compound, when used, is desirably in the range of about ½ to 2 percent, preferably about ½ to 1¼, calculated as NH_3, and based on the total air-dry weight of the tobacco substitute. A suitable proportion of coloring agent, when used is desirably in the range of about 0.1 to 3 percent, preferably when 1/5 to 1 percent, based on the weight of the cellulose.

The cellulose used should have an alpha cellulose content of at least 90 percent, preferably at least 92 percent, e.g., 95 percent, such as acetate grade or viscose grade wood pulp. It is desirable that the alpha cellulose constitute a major portion, preferably at least about 60 percent, of the total air-dry weight of the tobacco substitute. Some suitable sources of wood pulp are hemlock, spruce and other coniferous firs.

The cellulose should be combined intimately with the additive or additives to be employed. This may be done conveniently by saturating a sheet of paper made from the lightly beaten cellulose with a solution, dispersion or emulsion of the additives, which may be applied separately or all together, followed by drying and shredding the sheet or cutting it into strips. If desired, a sheet may be cut into strips or shreds and sprayed with a solution or dispersion of the additive or additives and then aged or tumbled to assist in the uniform distribution of the additive material on the cellulose, and thereafter dried. All or any part of the additive material may be incorporated into the cellulose in the course of a papermaking operation. One convenient way of doing this is by incorporating the water-soluble additive material into a beaten pulp stock, then forming the stock into paper on a conventional papermaking machine, thereafter treating the paper, preferably while still wet, with a solution or emulsion of nonwater soluble materials and of the more expensive water-soluble constituents (e.g., nicotine), followed by drying and shredding. In this case the white water, drained from the papermaking machine, contains the water soluble additives and may be reused. More economically the other constituents are applied to the paper after it has left the Fourdrinier screen of the papermaking machine, as by spraying an aqueous solution thereof onto the wet sheet on the papermaking screen or, more advantageously, by passing the formed paper continuously through said solution.

When incorporating the additives into performed paper it is convenient to pass the paper through an aqueous bath in an apparatus such as shown in FIG. **2**, in which the paper **11** is passed under an idler roll **12** in the bath **13** and then moves out of the bath, but still carrying a film of the bath liquid, through a pair of precisely spaced squeeze rolls **14** and **15**, which press out any excess liquid. The wet paper, supported on a belt **16**, is then dried by hot air in a drier **17**. The paper **11** may, if desired, have had added thereto 1 to 2 percent of a conventional wet strength addition.

As previously discussed, potassium carbonate and other carbonates tend to react with part of the magnesium sulfate to form magnesium carbonate. When both the carbonate and the magnesium sulfate are present in the same aqueous bath the resulting insoluble magnesium carbonate must be kept in suspension if it is desired that it be incorporated in the paper uniformly with the water soluble constituents. It is sometimes more advantageous to apply the soluble carbonate separately. This may be done by applying an aqueous solution of the carbonate to the paper before or after the solution of the sulfate is applied. The amount of carbonate is small relative to the amount of magnesium sulfate and a small amount of the carbonate solution may be applied, as by a coating roller, just before the paper is saturated with the magnesium sulfate solution, without any intermediate drying step.

The amount of solution or dispersion, and of the constituents of such solution and dispersion, taken by the paper will depend on such factors as the moisture content and porosity of the paper, the length of time the paper is immersed in the solution and the extent to which the paper is pressed or squeezed after it leaves the saturating bath. Accordingly, it is best to determine the concentrations of the solutions by preliminary trials for each type of machine and process. Suitable concentrations of magnesium sulfate in water are, for example, in the range of about 2 to 40 percent, with the other constituents in proportion, in accordance with the ratio of such constituents to magnesium sulfate desired in the product. When the potassium carbonate is applied separately its concentration in water may be, for example, in the range of about ½ to 6 percent. When the carbonate is applied after the sulfate (or other water-soluble hydrate) the tendency for the latter to be extracted from the cellulose by the carbonate solution may be reduced by using a mixture of water and a water-miscible organic solvent, such as ethanol, to dissolve the carbonate; in this case the carbonate tends to deposit preferentially on the cellulose, in a proportion greater than when a water solution of the same concentration of the same carbonate is applied. Loss of sulfate by extraction into the carbonate solution may also be reduced by spraying the carbonate solution onto the sulfate-cellulose blend, rather than dipping the blend in the aqueous carbonate solution.

A particularly good cigarette is obtained when the tobacco substitute is in the form of thin, narrow strips of nonuniform, e.g., random, length and width the cigarette is easier to light, the filler has better packing quality and the cigarette has a better appearance. Strips may be for example from 0.2 to 2 mm. in width and from 1 to 25 mm. in length. The paper need not be uniform in thickness. I have found it desirable to use paper of variable thickness and low density; in fact portions of the paper may be less than 3 mils in thickness. Such nonconformity may be attained by depositing the pulp slurry onto the paper-forming screen nonuniformly, as by tilting the screen slightly transversely across the width of the sheet, or by only lightly stirring the mass of slurry so that it is relatively heterogeneous (of varying concentration in different parts of the slurry) during the operation of depositing the slurry onto the screen through a slit of nonuniform width.

The cut or shredded tobacco substitute may be processed exactly like tobacco on the usual cigarette-making machines, such as those shown in U.S. Pat. Nos. 1,787,551; 2,190,032; 2,208,504; 2,247,413; 2,236,579; or 2,671,452.

The cut or shredded tobacco substitute may also be blended with tobacco itself, with tobacco extract or with any of the usual reconstituted tobaccos. The proportion of cured tobacco thus blended with tobacco substitute may be in the range of about 5 to 75 percent, for example. When the proportion of tobacco is considerable, e.g., over 50 percent, it may be desirable to decrease the proportion of, or omit entirely, the nicotine, and flavoring agent in the tobacco substitute.

As previously stated, the use of low density paper base from the lightly beaten pulp makes possible a considerable reduction in the amount of smoke condensate (tar) produced in the smoking of the cellulose-based cigarette. Thus, in one experiment comparing cigarettes made from equal weights of pulps having freeness values of about 560 ml. and about 250 ml., respectively, the smoke of the former contained about one-third the tar of the latter. I do not have any clear explanation for this unpredictable effect. It may be that the greater porosity leads to more complete combustion or to better filtration of the smoke passing through the cigarette. Other complex physi-

cal and physico-chemical interations probably occur. This invention makes not only for a reduction in the amount of smoke condensate but also provides a condensate very low in polycyclic aromatics (as indicated by its very low fluorescence).

If desired, the entire tobacco substitute may be diluted by mixing the same with a noncombustible filler. Examples of suitable fillers are clay (e.g., bentonite or attapulgus clay) and alumina, e.g., in proportion of about 30 percent.

The following examples are given to illustrate this invention further.

EXAMPLE I

A 4 percent aqueous slurry of viscose grade sulphite wood pulp containing about 92 percent alpha cellulose was beaten in a hollander beater until a freeness value, determined according to TAPPI Standard T 227 *m*–50, of 574 ml. was obtained. Beating was then discontinued. The beaten pulp stock was diluted to 1 percent consistency. A measured volume of the 1 percent slurry was suspended in a measured volume of concentrated aqueous solution of $MgSO_4 \cdot 7H_2O$, $(NH_4)_2SO_4$ and sorbitol. The additives-treated pulp suspension was formed into a paper sheet in conventional manner, at low squeeze pressures to produce a low-density sheet, and dried. The dried sheet was dipped into a 60/40 (by volume) solution of ethanol/water at 25° C. containing 5.0 grams potassium carbonate and 4.0 grams nicotine per liter. The residence time of the sheet in the dipping solution was about 2 seconds. The impregnated paper, which did not disintegrate during dipping, was dried and then cut and shredded into filler strips whose lengths varied randomly from 1 millimeter to 25 millimeters and whose widths varied randomly from 0.3 millimeter to 1.3 millimeter. The filler was conditioned overnight at 65 percent R.H. and 75° F. and made into cigarettes, using standard cigarette paper for the wrapper. The average weight of these cigarettes was 0.96 grams of which 50 milligrams was the weight of the cigarette paper wrapper and the cigarette length was 70 millimeters.

In this example the proportions used were so chosen that the air dried tobacco substitute showed the following analysis:

$MgSO_4 \cdot 7H_2O$	13.3%
$Mg(NO_3)_2 6H_2O$	1.8%
Sorbitol	1.2%
$(NH_4)_2SO_4$	3.3%
K_2CO_3	1.2%
Nicotine	1 %

EXAMPLE II

Example I was repeated except that the aqueous solution containing magnesium sulfate, magnesium nitrate, sorbitol and ammonium sulfate was used to impregnate paper made from the beaten pulp stock, instead of adding the solution to the stock.

EXAMPLE III

Cigarettes were prepared in generally the manner of example I, except that the saturating solutions contained solely nicotine, sorbitol and two levels (5 and 25 percent) of either magnesium sulfate, magnesium acetate or magnesium citrate. The ratio of salts to sorbitol to nicotine was 50:4.3:3.9 in all samples.

The following temperature profiles were obtained:

	Peak ° C.	Rest ° C.
$MgSO_4 \cdot H_2O$		
at 5% level	850–1,025	675–700
at 25% level	765–1,065	550–685
$Mg(OAc)_2 \cdot 4H_2O$		
at 5% level	850–1,150	640–640
at 25% level	910–1,150	640–725
$Mg_3(Citrate)_2 \cdot ?H_2O$		
at 5% level	925–1,150	550–685
at 25% level	870–1,150	—

The cigarettes as above were smoked, and other cigarettes were prepared in which the treated and shredded cellulosic base was mixed with 5 and 95 percent tobacco. The magnesium acetate samples burned very well with no difficulty in sustaining burning. The ash appearance was good, although somewhat fluffy and white. The taste was much better than the magnesium sulfate samples, however, the smoke was somewhat difficult to inhale. The citrate samples burned well at both levels and at both tobacco levels. The smoke was milder and could be inhaled. No sulfide odors were noticed, and the aftertaste was less. The ash at the lower level closely resembled that of a commercial cigarette.

EXAMPLE IV

Cigarettes were again prepared in the manner of example I, except that in one series of tests about 15 percent of gluconic acid was substituted for the magnesium salts, and in another series about 15 percent magnesium gluconate was used. In the first case the peak temperature was in the range of 960° to 1,150° C., the rest temperature ranged from 640° to 810° C., while with magnesium gluconate these temperatures were 845° to 990° and 595° to 675°, respectively.

In both cases the shredded and treated cellulosic base was blended with 50 percent tobacco from a conventional cigarette and smoked. The smoke was extremely mild, pleasant and could be inhaled.

The following examples will illustrate the invention further.

A high alpha cellulose content (acetate grade) woodpulp slurry, made from 1.5 pounds of woodpulp and 48 pounds of water, was charged into a beater purchased from Valley Iron Works. The slurry was beaten to a freeness of approximately 645 ml. (as measured in accordance with TAPPI Standard T 227M–50), using a Canadian Standard Freeness tester.

Approximately 100 cc. of the pulp slurry was removed from the beater, diluted with water to 1,000 cc. to give a pulp concentration of about 0.3 percent by weight, and was then added to a Valley Sheet mold which had previously been substantially filled with water. The excess water was drained off, the sheet produced was removed from the screen, placed between sheets of blotter paper, rolled lightly and carefully dried upon a steam plate drier with a canvas cover on the sheet. The low density sheet obtained was then saturated with the following formulation, providing when air dry a sheet containing approximately a 15 percent magnesium sulfate heptahydrate:

	Wt. % in product
Cellulose	66.5
Magnesium sulfate	8.0
Potassium nitrate	1.17
Ammonium sulfate	3.23
Potassium bicarbonate	1.40
Sorbitol	1.24
Nicotine	0.95
Food colors	0.40
Flavors	0.10
Water	17

When dry, the saturated sheet was made into cigarettes in the manner described in the subject patent application, and the cigarettes were machine smoked. Ten cigarettes were smoked and the volatiles are estimated to comprise about 50 percent water and the remainder organic tars. After the trap was allowed to reach room temperature it was weighed, and the weight difference represents total condensate.

It was found that when the pulp was beaten to a freeness of approximately 645 ml. as described above and using low squeeze pressure to get a low-density paper, the total condensate obtained was 12.3 mg. per cigarette with each cigarette in these tests weighing approximately 0.8 grams.

The procedure outlined above was again followed, except that the pulp slurry was beaten for a total time of approximately 40 minutes to a freeness of value of 290 ml. The total condensate obtained was 49.2 mg. per cigarette as measured in the manner earlier described.

The beating was allowed to continue for a total time of approximately 100 minutes in a third trial, providing a freeness value of 62 ml. Following the same procedure as in the first trial, the total condensate was about 80.5 mg. per cigarette.

The foregoing trials clearly demonstrate that by employing a low-density base paper from a lightly beaten pulp, that is, one having a high-freeness value, the total condensate and consequently the tars therein are held to a very low value. On the other hand, beating to a low freeness value markedly increases the amount of total tars in the smokeable material produced.

Both high- and low-density paper can be made from a high freeness pulp, using low squeeze pressure to get a low-density paper. The effect on tar values of employing a low density sheet (12–35 lbs./ft.^{3}), preferably 15–25 lbs./ft.^{3}, made from pulp with a high freeness value, was confirmed by a series of trials which demonstrated the relationship sheet density to tar values.

In this series of trials a pulp slurry of the same formulation as above described as beaten for approximately 10 minutes to a freeness value of about 600 ml. After being processed through the same sheet mold, dried and saturated with the formulation above set forth, the sheets obtained, which measured 8 inches square and contained 3 grams of cellulose, were placed in a hydraulic press and subjected in a group of three experiments to differing ram or squeeze pressures.

In a first test relatively light pressure of 5 p.s.i.g. was employed to obtain a sheet having a thickness of about 8 mils. The sheet was then made into cigarettes, machine smoked as previously described, and the total condensate measured. It was found that a sheet subjected to the pressure noted and having the thickness mentioned provided a total condensate of 25±5 mg. per cigarette when measured as above. The sheet had a density of 20 pounds per cubic foot, or 0.32 grams per cubic centimeter.

A second test was performed in which the sheet was subjected in the hydraulic press to a ram pressure of about 65 p.s.i.g. to produce a sheet thickness of 5.1 mils. Cigarettes made from such a sheet when machine smoked as previously gave a total condensate value of 50±5 mg. per cigarette. The sheet had a density of 32 pounds per cubic foot or 0.51 grams per cubic centimeter.

In a third test a sheet in the hydraulic press was subject to a pressure of about 300 p.s.i.g., to provide a sheet having a thickness of 4.5 mils. The total condensate obtained from cigarettes produced and smoked was 70± mg. per cigarette. The sheet density was 35 pounds per cubic foot or 0.56 grams per cubic centimeter.

In another set of trials using pulp specified to contain alpha cellulose in excess of 94 percent, hand sheets were made by procedures outlined above, and using the aforesaid sheet mold, the pulp was beaten as previously described to a Canadian Freeness value of 585 ml. ± ml.±35 ml. and dried under varying light pressures to yield the following range of sheet densities and thicknesses:

Trial	Thickness in inches	Density lb./ft.^{3}	Tar mg./cigarettes
A	0.0080	20	26.7
B	0.0051	32	47.2
C	0.0045	35	67.0

The sheets were saturated as disclosed above, shredded, and made into cigarettes weighing about 1 gram each. The values of smoke condensate found are given in the above table in the column labeled "Tar".

The same pulp was shipped to a commercial paper maker and beaten in commercial equipment to the aforementioned freeness specification, converted on a commercial Fourdrinier paper making machine and dried under minimum drying pressures, (about 5 p.s.i.g.). The sheets so produced had a thickness of 0.008 inches and a density of 21 lbs./ft.^{3}. After saturating as before, cigarettes were made from this commercial sheet and the condensate yield was found to be about 25 mg./cigarette.

The above described trials clearly demonstrate the importance of employing a low density sheet obtained by beating the pulp to a high freeness value in order to provide a low-tar smoking material.

A series of tests have also been conducted which demonstrate that when the cigarettes of this invention are smoked there is present in the smoke less total condensate and total phenols, as compared with conventional nonfilter cigarettes, and further, which display in vapor phase analyses relatively lesser amount of the major molecules components reported to be harmful substances in the smoke of conventional nonfilter cigarettes.

A first group of cigarettes (designated in table A below as group I) was prepared for the above determinations essentially as described in examples I and II of the (instant) patent application, using substantially the formulation appearing therein.

A second group of cigarettes (designated as group II in table A) was prepared in a like manner, except that the total amount of additives was varied from about 8 to 16 to 24 to 32 to 40 percent by weight of air dried product. A third group of cigarettes (designated as group III in table B below) was prepared employing simply magnesium sulfate and high-alpha cellulose, and in a successive series of formulations, by adding to the sulfate and cellulose base, certain additional salts. A fourth group of cigarettes (referred to as group IV in table B) was provided by conventional all-tobacco nonfilter cigarettes.

	Phenol	Cresol, o.p.m.	Total phenolics	Condesate (wet) 22±3 puffs
Group I	33	9, 5, 7	54	22.0
Group II:				
(a) 81% cellulose plus 11% moisture plus 8% additives (incl. 5% $MgSO_4.7H_2O$)	22	6, 5, 5	38	27.1
(b) 74% cellulose plus 10% moisture plus 16% additives (incl. 10% $MgSO_4.7H_2O$)	22	7, 4, 6	39	18.7
(c) 66% cellulose plus 10% moisture plus 24% additives (incl. 15% $MgSO_4.7H_2O$)	33	9, 5, 7	54	22.0
(d) 69% cellulose plus 9% moisture plus 2% additives (incl. 20% $MgSO_4.7H_2O$)	22	5, 8, 7	42	23.2
(e) 53% cellulose plus 7% moisture plus 40% additives (incl. 25% $MgSO_4.7H_2O$)	15	3, 2, 2	22	26.1
				Condensate (wet) 23±3 puffs
Group III:				
(a) 15% $MgSO_4.7H_2O$ plus 11% moisture plus 74% cellulose	35	12, 8, 8	63	54
(b) 15% $MgSO_4.7H_2O$ plus 1.2% KNO_3 plus 11% moisture plus 72.8% cellulose	25	10, 5, 8	48	66
(c) 15% $MgSO_4.7H_2O$ plus 3.2% $(NH_4)_2SO_4$ plus 11% moisture plus 70.8% cellulose	50	14, 9, 10	83	38
(d) 15% $MgSO_4.7H_2O$ plus 1.4% $KHCO_3$ plus 11% moisture plus 72.6% cellulose	28	10, 5, 7	50	62
(e) 15% $MgSO_4.7H_2O$ plus 1.2% KNO_3 plus 3.2% $(NH_4)_2SO_4$ plus 10% moisture plus 71.6% cellulose	38	11, 7, 8	64	47
				Condensate (wet) 13±1 puffs
Group IV	117	25, 45, 20	207	53

1. The phenol determinations were conducted using generally the techniques described by D. Hoffman & E. L. Wynder, Beitrage zur Tabakforschung, No. 3, pp. 101–106 (1961), and a smoking machine substantially as disclosed by A. O'Keefe and R. Lieser, Tobacco Science, 2, 73 (1958) was employed therein. A 2-second puff of 35 cc. volume every 30 seconds was employed. The mainstream cigarette smoke was condensed on a glass fiber filter pad at room temperature, located between the smoking machine and cigarette holder. All cigarettes were 85 mm. in length and were smoked to a 25 millimeter butt, and also were conditioned and smoked at 60 percent relative humidity at 72° F.

The cigarettes were screened by weight prior to smoking, and cigarettes weighing within ±0.08 grams of the average cigarette weight were selected. Each determination of phenol in the smoke consisted of smoking six cigarettes on a single smoking machine position, and collecting the total condensable material in a single trap. The total condensate weight was obtained from the difference in pad weight before and after smoking. Aqueous sodium hydroxide solution (5percent NaOH) was then added to the pad to remove the condensate.

A Perkin-Elmer Model 225 Gas Chromatograph was used in the analysis of the phenolic fraction of the smoke condensate. This unit is equipped with a flame ionization detector. Chromatograph separation of the phenolic fraction of the condensate was achieved with a 100 foot Golay (capillary) column having a 0.02 inch internal diameter coated with a 85/15 mixture of Cellulube 550/Empol Dimer Acid.

The tabulation appearing as table "A" clearly demonstrates by a comparison of the results obtained from the cigarettes of groups I and II that the instantly disclosed smoking materials have a markedly lower total phenol and cresol content, as well as a lower total condensate, on a per puff basis, when measured under experimentally equivalent conditions with a conventional all-tobacco nonfilter cigarette of Group IV.

As well, when reference is made to the results set forth for the cigarettes noted as group III in table "A", the instant applicant's novel combination of magnesium sulfate and cellulose, by themselves, is much lower in phenolics, cresols and total wet condensate on a "per puff" basis, again compared with a conventional all-tobacco nonfilter cigarette. This same tabulation also demonstrates without question that when certain listed slat constituents are added to the sulfate and cellulose base, quite unexpectedly the phenolic compounds and cresols are much below those for a conventional all-tobacco nonfilter cigarette, and the total wet condensates are also considerably less when compared on an equivalent puff basis.

Additionally, it is manifest from a review of the results noted for the cigarettes identified as group II that even through the magnesium sulfate content disclosed in the present application is controllably varied over the range set forth in the application, and proportionate modifications are made in the other salt additives, the value for total phenols and cresols, as well as the total wet condensate, consistently remains at values drastically below those for a conventional all-tobacco nonfilter cigarette.

In the tables to which reference was made, the value for total phenol and cresols is measured in micrograms per cigarette, while condensate is measured in milligrams per cigarette. The total wet condensate is collected and weighed at room temperature and includes water of combustion.

A second series of determinations, namely vapor phase analyses from puffs comparable to the fifth puff from a conventional cigarette, were conducted by locating a precision gas sampling valve in the live between the glass fiber filter pad and the chromatograph.

The results of these extensive studies are summarized on the attached charts designated as FIGS. 3, 4 and 5. These are precise reproductions of typical chromatographs obtained from the vapor phase of smoke, FIG. 3 showing the results obtained from the cigarettes of group I (F–38) and group IV, and FIG. 3 portraying the vapor phase results from the cigarettes of group II. The vapor phase analysis from the cigarettes of group III will be discussed later. In the attached charts, the major molecular components are listed thereon, and the relative amounts present in the substantially comparable puffs are proportional to the peak areas displayed.

It is observed from FIG. 3 that the light hydrocarbons such as methane, ethane and the like are much lower than a commercial all-tobacco nonfilter cigarette, which is indicative of a more complete combustion epoch. Notably, isoprene is barely detectable This highly reactive compound in both chemical and physiological systems, is a known precursor of known toxins, as well as carcinogenic polynuclear substances. Similarly, a much lesser amount of acetaldehyde was found to be present. As is known, acetaldehyde has been reported to have a synergistic effect with acrolein, which is a harmful component of cigarette smoke in inhibiting normal ciliary activity. A publication on this point is *Inhibitory Effects of Tobacco Smoke on the Ciliary Activity of the Respiratory Epithelium and the Nature of the Compounds Responsible* R. Guillerm, R. Badre and B. Vignon.

Additionally, by reference to FIG. 3, it can be seen that even the instantly disclosed cellulose base by itself, while of course not completely suitable in unmodified form as a smoking material, displays generally lesser amounts of the listed molecular components. Even more important, however, is the fact that when the total amount of disclosed additives was widely varied, major improvements are seen when compared with a commercial all-tobacco nonfilter cigarette.

Vapor phase determinations have also been made on the cigarettes of group III, and these appear as FIG. 4. It has been found with the exception of the cigarettes made from cellulose and 15% $MgSO_4 \cdot 7H_2O$ plus 3.2% $(NH_4)_2SO_4$, which exhibited a slightly lower reduction in the charted molecular components, the cigarettes in this group showed over a 75 percent reduction in light hydrocarbons, greater than a 90 percent reduction in isoprene, more than an 80 percent lowering of acetaldehyde, a reduction exceeding 70 percent in methanol and acrolein, a lessening of methyl ethyl ketone by over 90 percent, and a reduction of more than 90 percent in toluene.

The total condensate from 40 cigarettes is subjected to a solvent partitioning scheme to enrich the polynuclear aromatic hydrocarbons (PAH). Further fractionation is achieved on a silica gel adsorption column eluted with methanol. Final fractionation is achieved on acetylated paper chromatograms. The 3,4-benzpyrene spot is excised and extracted with methanol and quantitatively measured with a spectrophotofluorometer of appropriate sensitivity.

The 3,4-benzpyrene content from machine made cigarettes produced in accordance with example I was found to be 0.6 micrograms per 100 cigarettes. Quite by contrast, the 3,4-benzpyrene content from conventional all-tobacco nonfilter cigarettes was found to be 4 to 6 micrograms per 100 cigarettes.

It is to be pointed out that the cellulose base employed in each of the above-described trials was beaten to within a Canadian Standard Freeness range of 400 to 700 ml., and specifically, to about 550 ml.

In the description and claims all proportions are by weight unless otherwise indicated. In the above specification, the proportions are expressed in terms of the compounds originally supplied, disregarding any chemical reaction between the compounds. Thus, although potassium carbonate interacts with part of the magnesium carbonate, the analyses, (based on the amounts of K.,SO_4,Mg and CO_3 present in the product) are expressed as percentages of potassium carbonate and magnesium sulfate.

In the above examples, the cigarette paper used was a standard nonporous cigarette paper (e.g., "Yorkshire" paper sold by Sears Roebuck & Co.). If desired microperforated paper, well known to the cigarette art, may be used instead.

It is to be understood that the foregoing detailed description is given merely by way of illustration and that many variations may be made therein without departing from the spirit of my invention.

The embodiments of the invention in which an exclusive property or privilege is claimed are defined as follows:

1. A method for producing a tobacco substitute comprising lightly beating fibrous woodpulp containing at least 90 percent alpha cellulose to a Canadian Standard Freeness of between 400 and 700 ml., intimately combining said woodpulp with from 2 to 25 percent, based on the weight of the woodpulp, of an orally nontoxic combustion modifier selected from the group consisting of the sulfates of magnesium, sodium and potassium; the chlorides of potassium and magnesium; the carbonates and bicarbonates of sodium, potassium, magnesium and ammonia; potassium nitrate; ferric oxide; ferric hydroxide; alumina; the gluconates, citrates and acetates of magnesium; and gluconic acid, and forming a sheet therefrom, said sheet having a density of 12 to 35 lbs./ft.^{3}.

2. The method of claim 1 wherein the said combustion modifier is a hydrated metal compound.

3. The method of claim 1 wherein said woodpulp contains at least 92 percent alpha cellulose.

4. The method of claim 1 further comprising shredding said paper sheet into narrow strips of nonuniform length and width.

5. The method of claim 1 wherein an amount of alpha cellulose is added such that said alpha cellulose constitutes at least 60 percent of the total weight of the tobacco substitute.

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