

1

2,941,906

## TOBACCO COMPOSITION AND SMOKING UNIT CONTAINING MATERIAL FOR ELIMINATING DELETERIOUS MATTER

Walter L. Haden, Jr., Metuchen, N.J., assignor to Minerals & Chemicals Corporation of America, Menlo Park, N.J., a corporation of Maryland

No Drawing. Filed Dec. 26, 1957, Ser. No. 705,152

11 Claims. (Cl. 131—17)

This invention relates to smoking compositions and to smokable units including therein a novel particulate inorganic material intimately associated with tobacco, the additive being capable of substantially reducing the quantity of tars normally generated during the burning of the tobacco and improving the aroma of tobacco. For many reasons, clinical and aesthetic, reduction in the tar yielded during the smoking of tobacco is highly desirable. First, there exists strong clinical, statistical and pathological evidence that certain constituents of tobacco smoke condensate (or "tars" as such condensate is usually termed) are potentially carcinogenic when deposited on human tissue. These carcinogens have been reported to be mixtures of essentially neutral high-molecular weight polycyclic hydrocarbons, the carcinogenic potency of any mixture depending on the species present. Several general types of polycyclic aromatic hydrocarbons are usually present in tobacco smoke condensate and undoubtedly many individual molecular species are hence present. Deleterious polycyclic hydrocarbons are undoubtedly of pyrogenic origin since they cannot be isolated from raw tobacco prior to the pyrolysis thereof. These noxious substances are ordinarily carried as dispersed droplets or solid particles in the effluent smoke stream and thence into the smoker's respiratory system along with the aroma and taste producing constituents of the smoke. Secondly, aside from their alleged pathological effect, it appears that certain constituents of tars are responsible for the sharp irritating taste imparted to smoke from a cigarette or the like. Furthermore, tars produce disfiguring coloration of the teeth of many smokers and, if only for this reason, the lowering of total tars yielded during smoking is highly desirable.

Many efforts have been made in the past to reduce the yield of tars in the effluent smoke of tobacco. The use of filter-tips, for example, has met widespread acceptance both by the cigarette industry and the consumer. Another method involves the commingling with smoking tobacco of particulate porous solids having high sorptivity by virtue of their large specific surface, internal or external. The inclusion in smoking mixtures of a variety of sorptive clays, colloidal and non-colloidal, and various forms of sorptive aluminous gels has been suggested in the prior art. Reduction of tobacco oils, nitrogenous bases and tars in the smoke have been among purported benefits although reproducible quantitative evidence capable of being correlated with in vivo smoking conditions has not been set forth in support of purported benefits. The tobacco industry has apparently failed to find that the inclusion in tobacco compositions of such particulate solids as have been proposed heretofore to be of sufficient value to warrant their use in commercial smoking mixtures.

It is apparent that smoking mixtures which include an innocuous additive which is capable of substantially lowering the tar yield of said tobacco when smoked meets a long-felt need of the tobacco smoker.

Accordingly, it is a principal object of the instant

2

invention to provide a composition including tobacco which incorporates a novel aluminous additive which functions during the pyrolysis of the tobacco to reduce substantially the quantity of tars in the smoke effluent from said tobacco.

Another object of the invention is the provision of a composition including tobacco intimately associated with an innocuous aluminous additive capable at the elevated temperatures encountered in smoking tobacco of substantially lowering the yield of tars in the main stream smoke of tobacco.

It is another important object to provide smoking units including tobacco and a particulate aluminous additive dispersed through said tobacco and capable of curtailing the quantity of tar generated during pyrolysis of said tobacco.

Other objects will be apparent from a reading of a description of the invention which follows.

I have discovered that substantial benefits are realized when a unique form of alumina is included in tobacco mixtures. Briefly, compositions of my invention comprise tobacco intimately associated with alumina obtained by thermal desulfation of a material comprising aluminum sulfate. Tobacco compositions made in accordance with the instant invention are characterized by a smooth, bite-free taste when smoked. The smoke effluent from said compositions is distinguished by a tar content substantially reduced from that of a smoking mixture including the same tobacco specie or species but devoid of the novel additive. The additive is one of a chemical composition that is recognized by competent contemporary authorities as being essentially innocuous. The aluminous additive within the scope of my invention improves the smoking characteristics to a hitherto unexpected degree. Although the invention will be described with particular reference to cigarettes, it will be understood that comparable benefits are realized when compositions of my invention are cigar and pipe smoking mixtures.

Prior art workers have suggested the dispersion in tobacco mixtures of colloidal alumina, colloidal aluminum hydroxide and so-called "light alumina hydrate" for such purposes as sorbing deleterious pyrogenic or otherwise produced materials. Activated bauxite has been suggested as a tobacco additive to control the release of such tobacco treating agents as moisture or menthol during smoking of tobacco. None of these materials is within the compass of my invention nor is their performance in smoking mixtures comparable. Light alumina hydrate, for example, is a crystalline sulfated aluminum hydrate of the approximate formula  $5Al_2O_3 \cdot 2SO_3 \cdot xH_2O$  whereas my additive is essentially sulfur-free alumina of non-colloidal substantially amorphous character, said alumina being profoundly different from aluminas otherwise prepared. I have dispersed several aluminous materials in tobacco mixtures but have found that they do not produce the outstanding benefits that are realized when a thermally desulfated aluminum sulfate is used. I have found, for example, that neither alpha-alumina trihydrate nor alumina obtained therefrom by thermal activation will reduce the tar generated from tobacco mixtures.

More specifically, in accordance with my invention I intimately associate from about 1 to 20%, and more preferably from about 2 to 15%, based on the weight of the tobacco, of the finely-divided alumina produced by the controlled thermal desulfation of an aluminum sulfate or a hydrate thereof. The aluminum sulfate material which is the precursor of the particular alumina useful in compositions of my invention may be of natural or synthetic origin. The source may be pure aluminum sulfate or may be aluminum sulfate associated with a minor amount of an impurity mechanically or chemically associated with said aluminum sulfate provided that impurity

3

may be removed from the aluminum sulfate substantially completely prior to its desulfation. For example, ammonium alums may be used as the starting material. Useful naturally-occurring sources of aluminum sulfate or hydrates include the minerals alunogenite and aluminite, the former consisting essentially of the octadecahydrate of aluminum sulfate and the latter corresponding approximately to the formula  $\text{Al}_2(\text{OH})_4\text{SO}_4 \cdot 7\text{H}_2\text{O}$ . Aluminum sulfate or hydrates thereof may be of synthetic origin, such as, for example, from "aluminum cake" which may be made by digesting ground bauxite with sulfuric acid, removing silica and other insoluble materials, evaporating the clarified filtrate to the described density and cooling the resultant hot concentrated solution. A more pure sulfate of alumina hydrate which is a product of the Bayer process may be used in place of bauxite.

The sulfate of alumina is calcined for a sufficient time and at a suitable elevated temperature to eliminate substantially all of the sulfate content and leave an essentially pure alumina of the requisite porosity. The desulfation will be carried out between 1235° F., the decomposition temperature for aluminum sulfate, and below the temperature at which alpha-alumina is formed, the latter temperature varying considerably with operating conditions. Desulfation may be carried out in a reducing atmosphere, such as in the presence of carbon monoxide, hydrogen or methane or mixtures thereof, or in the presence of carbon or coke. The advantage of conducting the desulfation in a reducing atmosphere is that desulfation of the aluminum sulfate may be carried out at a lower temperature and produce a material of somewhat greater activity. Ordinarily a very satisfactory material will be obtained by calcination at temperatures of the order of 1300–1500° F. when a reducing atmosphere is employed whereas calcination at temperatures of from about 1500–1800° F. is particularly suitable in a non-reducing atmosphere. Ordinarily calcination times within the range of from about 1 to 24 hours will suffice although variations in the heating time may sometimes be indicated. The criterion of calcination completion will be a sulphate content in the product of less than about 0.5% and preferably less than 0.1%.

The additive may be applied to whole leaf, reconstituted tobacco or mixtures thereof. Also, the additive may be an integral component of reconstituted tobacco, as will be accomplished by including the additive in the pulverulent tobacco mass from which sheets of reconstituted tobacco are formed. The additive may be applied by dusting the dry material on the tobacco, by spraying or by any other well-known means to apply a finely-divided material to a substrate.

The aluminous additive is preferably incorporated in cigarette tobacco in comminuted form, usually about 300-mesh or finer and preferably of a particle size distribution such that a preponderating portion by weight is less than about 10 microns. In general, the more finely divided the aluminous additive the greater the adhesion to the tobacco. Of course the particular particle size used in a composition will depend on such factors as moisture content, presence of tacky humectants, mode of application to the tobacco, size of tobacco particles, presence and nature of filter-device, quantity of aluminous additive and locus of additive placement.

The aluminous additive may be distributed substantially uniformly throughout the body of the cigarette or, as in an embodiment of the invention, the material may be placed selectively within the cigarette to effect maximum benefits. Accordingly, the material may be advantageously gradated throughout the cigarette, with maximum content proximate the inhaling end where, during the smoking of the cigarette, maximum tar deposition is encountered.

A filter, either of the well-known type which is integral with the smoking unit or of the holder type including a filter, may be employed in a smoking unit when tobacco is treated by the process herein taught. The

4

filter may be fibrous and/or include adsorbents such as silica, clays or the like. The purpose of the filter is to prevent inspiration of finely-divided aluminous particles which may occur if the smoking unit is subjected to dry warm weather for prolonged periods. The filter interposes a bed of porous material, or the like, between the tobacco-alumina mixture and the smoker's mouth and imprisons any alumina particle which might otherwise be drawn into the smoker's mouth. However, by proper selection of moisture content and, in some cases, inclusion of about 2–4% by weight of an organic humectant, the filter may be omitted.

The following embodiment of the invention is given only for the sake of more fully illustrating the invention and is not to be construed as limiting the scope thereof.

#### EXAMPLE I

A tobacco composition of my invention was prepared by admixing shredded humected tobacco with thermally desulfated aluminum sulfate in the ratio of 10 parts of tobacco to 1 part of aluminous additive. The mixture was packed into cigarette paper, each cigarette weighing approximately 1.0 grams. The thermally desulfated material was obtained by calcining aluminum sulfate, dried powder, Bakers analyzed reagent grade at 1500° F. for 6 hours, 1600° F. for 6 hours, followed by calcination at 1800° F. for 3 hours.

#### EXAMPLE II

The benefits realized by dispersing an aluminous additive within the scope of my invention in cigarettes was explored with respect to effect on tar content of the smoke effluent from a unit weight of tobacco pyrolyzed in the presence of the additive. A control cigarette was prepared using the same quantity and stock of tobacco as a cigarette made in Example I but devoid of additive; the tar content of the smoke of control cigarettes, mechanically smoked, was compared to that of cigarettes including the same weight of tobacco but including the novel aluminous additive.

In the example all cigarettes were mechanically smoked by the method described in detail in Industrial and Engineering Chemistry, vol. 28 No. 7 in an article entitled "Nature of Cigarette Smoke-Technic of Experimental Smoking," J. A. Bradford et al., pp. 836–839, inclusive (1936). Analysis of total tar content was made by collecting the tobacco smoke in a glass wool filter tube at room temperature and evacuating the tube at room temperature to eliminate moisture. Quantitative determination of tar yield was measured by calculation of the weight pick-up of the tube after removal of moisture therefrom. The mechanical smoking method employed approximates the conditions of human smoking sufficiently for conclusions of in vitro experiments to be interpreted in vivo. Tar yields obtained by measurement of the condensate of mechanically smoked cigarettes are tabulated below.

Cigarette compositions

|                                | Wt. of Desulfated Average Tar Yield |                          |                            |
|--------------------------------|-------------------------------------|--------------------------|----------------------------|
|                                | Wt. of Tobacco (grams)              | Aluminum Sulfate (grams) | Per Cigarette (milligrams) |
| 1. Control cigarette.....      | 0.90                                | -----                    | 35.3                       |
| 2. Cigarette of Example I..... | 0.90                                | 0.09                     | 22.7                       |

Obviously, many modifications and variations of the invention as hereinbefore set forth may be made without departing from the spirit and scope thereof and, therefore, only such limitations should be imposed as are indicated in the appended claims. For example, it is within the scope of the invention to include in the smoking mixture or smokable units including the same, small portions of reconstituted tobacco leaf for the purpose

5

of lowering the material cost of the mixture or unit containing the same. Also, it is fully within the scope of my invention to add to tobacco (prior to or subsequent to incorporation of additive to said tobacco) those taste improving tobacco additives commonly employed to improve the aromatic qualities of the unsmoked and/or smoked tobacco, such additives being herein termed "taste-imparting tobacco treating agents." Materials falling within this classification include essential oils, invert sugar, cocoa, chocolate, licorice and the like.

I claim:

1. A smoking mixture comprising a major portion of tobacco and intimately associated with at least a portion of said tobacco finely-divided thermally desulfated sulfate of aluminum.

2. A smoking mixture comprising a major portion of tobacco and intimately associated with at least a portion of said tobacco finely-divided thermally desulfated alunogenite.

3. A smoking mixture comprising a major portion of tobacco and intimately associated with at least a portion of said tobacco finely-divided thermally desulfated aluminite.

4. A smoking mixture comprising a major portion of particulated tobacco and intimately associated with at least a portion of said tobacco finely-divided thermally desulfated aluminum sulfate.

5. The smoking mixture of claim 4 in which said desulfated aluminum sulfate comprises a preponderating amount by weight of particles smaller than 10 microns.

6. The smoking mixture of claim 4 in which said desulfated aluminum sulfate comprises a substantially major weight proportion of particles less than 2 microns and negligible matter greater than 10 microns.

7. A smoking unit comprising a column of particulated tobacco and dispersed throughout at least a portion of said tobacco and intimately associated therewith comminuted microporous substantially totally desulfated sulfate of aluminum.

6

8. A smoking unit comprising a column of particulated tobacco and dispersed therethrough and intimately associated therewith from 1 to 20 percent by weight of finely-divided microporous amorphous substantially totally desulfated aluminum sulfate.

9. A smoking unit comprising a column of particulated tobacco and dispersed therethrough and intimately associated therewith from 2 to 15 percent by weight of comminuted microporous amorphous substantially totally desulfated aluminum sulfate.

10. A smoking unit comprising a column of particulated tobacco, said column terminating at one free end in a filter-tip, and distributed throughout at least that portion of said column contiguous said filter-tip, comminuted microporous amorphous substantially totally desulfated aluminum sulfate.

11. A smoking unit comprising a column of particulated tobacco, said column terminating at one free end thereof in a filter-tip, and distributed throughout said column and intimately associated with the tobacco therein comminuted microporous amorphous totally desulfated aluminum sulfate.

#### References Cited in the file of this patent

##### UNITED STATES PATENTS

|           |                     |               |
|-----------|---------------------|---------------|
| 2,282,922 | Ahlberg et al. .... | May 12, 1942  |
| 2,706,695 | Frankenburg .....   | Apr. 19, 1955 |
| 2,839,065 | Milton .....        | June 17, 1958 |

##### FOREIGN PATENTS

|         |                     |              |
|---------|---------------------|--------------|
| 666,308 | Great Britain ..... | Feb. 6, 1952 |
|---------|---------------------|--------------|

##### OTHER REFERENCES

35 Derr et al.: (Pub.) "Removal of Undesirable Constituents From Tobacco Smoke" from "Industrial and Engineering Chemistry," July 1937, vol. 29, pages 771 to 776, inclusive.