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TOBACCO COMPOSITION AND SMOKABLE UNIT CONTAINING MATERIAL FOR REMOVING DELETERIOUS MATTER

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This invention relates to a smoking composition and to smokable articles including an acid activated kaolin clay material intimately associated with tobacco.

There exists strong clinical, statistical and pathological evidence that certain constituents of tobacco smoke condensate are potentially carcinogenic when deposited on human tissue. These carcinogens have been reported to be mixtures of essentially neutral high-molecular weight polycyclic aromatic hydrocarbons, the carcinogenic potency of any mixture depending on the species present. Several general types of polycyclic aromatic hydrocarbons are present in a carcinogenic fraction of such smoke condensate, or tar, and undoubtedly many individual molecular species are hence present. In general, known carcinogens contain at least four condensed benzene rings per molecule and may include derivatives and substitution products of benzanthracene, phenanthrene, pyrene, chrysene and other types of condensed-ring aromatic hydrocarbons. Certain of these highly carcinogenic compounds such as 1,2,5,6-dibenzanthracene and derivatives of 3,4-benzpyrene have been identified, whereas others are as yet unidentified or have not been resolved into individual molecular species. Such deleterious polycyclic hydrocarbons are undoubtedly of pyrogenic origin since they cannot be isolated from raw tobacco prior to the pyrolysis thereof. It is believed that the precursor of these polycyclic hydrocarbons is the wax naturally resident in the cutin and rays of the tobacco leaf, such wax being essentially paraffinic and containing hentriacontane as a principal constituent.

The generation of these allegedly carcinogenic compounds is favored by high temperatures; hence they are present in greater quantity in the tars from a cigarette than in the tars from a cigar or pipe, which when smoked develop peak temperatures several hundred degrees lower than that of a cigarette. At the peak temperature of $1623 \pm 30°$ F. developed in a conventional cigarette when puffed such carcinogens are generated in potentially dangerous quantities.

Such 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. Aside from their alleged pathological effect it appears that certain constituents of tars are in part responsible for a sharp, irritating taste imparted to smoke from a cigarette or the like.

It has been suggested to distribute in the column of tobacco in a cigarette or the like sufficient sorptive material of natural or synthetic origin to effectively retain all deleterious matter produced during the burning of tobacco thereby preventing the exit of such matter in the smoke. However, at the elevated temperatures encountered near the flame-front of a cigarette the sorptivity of such materials is very low as compared with their sorptivity at room temperature and considerable release of volatilizable material contacting such sorptive materials occurrs. Hence the volatilizable material including

high-molecular weight noxious polycyclic aromatic substances can bypass the sorptive material unless the sorptive material is used in very large quantities with attendant difficulty in keeping the cigarette lit. Furthermore, such sorptive materials having this ability unfortunately inherently possess the capacity to remove the desirable volatile aroma-producing components produced during smoking.

The desiderata for an ideal agent for eliminating high-molecular weight undesirable components from the effluent smoke of the tobacco in a cigarette or the like are:

(1) The ability to substantially eliminate these components without alteration of the agent itself and without reduction in the quantity of lower-molecular weight, desirable aromatic compounds generated;

(2) Residual sorptivity, so that any undestroyed high-molecular weight component will be sorbed rather than be carried into the effluent smoke;

(3) Thermal stability at the elevated temperatures encountered at and proximate to the flame-front;

(4) Non-toxic;

(5) Ability to retain physical structure in the presence of moisture.

Accordingly, it is a principal object of the instant invention to provide a composition including tobacco which incorporates a novel treating agent to eliminate substantially and otherwise prevent the exit in the effluent smoke of deleterious high-molecular weight compounds, particularly polycyclic aromatic compounds, pyrogenetically or otherwise produced.

Another object of the invention is the provision of a composition including tobacco intimately associated with a novel combination of treating agents which are capable of acting inter se to promote the formation of relatively low molecular weight compounds of pleasant aroma to the exclusion of the formation of higher-molecular weight, potentially carcinogenic substances.

It is another object of the invention to provide smoking units including such compositions which inherently possess the ability to improve simultaneously the aroma of the smoke from the tobacco and prevent substantially the generation of deleterious high-molecular weight polycyclic compounds.

Other objects will be apparent from an inspection of the following description of the invention.

We have discovered that the presence in smoking tobacco of certain naturally-occurring acid-activated siliceous clays having a high alumina content is associated with the generation of a mild tasting smoke having enhanced aroma. Furthermore, we have found that free moisture carried by our acid activating kaolin clay particles in a tobacco composition cooperates to improve not only the ability of the acid activated kaolin clay to adhere to the tobacco particles but also the smoking characteristics of the composition.

More specifically we have discovered that certain acid-activated kaolin clays, particularly when adjusted to a volatile matter content of from about 30 to 40 percent, fulfill the desiderata of an ideal agent for the purposes set forth. By "volatile matter" is meant that weight of the clay expressed on a percentage basis, which would be eliminated by heating said clay at a temperature of about 1700° F. to substantially constant weight. As used herein, the terms "kaolin" or "kaolin clay" are those clays which include in the raw state as the principal clay mineral constituent, kaolinite, halloysite, indianite, dickite, nacrite or anauxite. All of these clay minerals are hydrous aluminum silicates prior to calcination and are represented by the formula $Al_2O_3.2SiO_2.nH_2O$, n usually being 2. The weight ratio of silica to alumina is about 1.0 to 1.5, and most generally about 1.18. The alumina content of those acid-activated clays useful in practicing

the instant invention is from 40 to 50 percent by volatile-free weight, and usually about 46 percent, the weight of the clay without its volatile matter being referred to as the volatile-free weight of said clay.

It has been found that the addition to tobacco of this specific acid-activated kaolin clay in amount from about 0.5 to 15 percent (based on the volatile-free weight of acid-activated kaolin clay and preferably from about 2 to 10 percent, same basis, produces outstanding benefits. The specific amount will vary with the type and quality of tobacco used and the moisture content of the composition and is best ascertained by examination of the smoking characteristics of the composition, such as the ease of draw, ability to sustain burning, analysis of combustion product distribution and of taste and aroma. The improvement in redolence of the smoke produced by burning of tobacco compositions including the acid-activated kaolin clay over that of smoke generated in the absence of such material results from an alteration of the character of the products of combustion. Specifically, it is felt that in lieu of those high-molecular weight polycyclic hydrocarbons of a carcinogenic character which would normally be present in the tobacco smoke condensate, the smoke from tobacco containing the acid-activated kaolin clay includes not only the quantity of desirable volatile aromatic compounds normally formed but additional quantities attributable to the elimination of formation of high-molecular weight polycyclic compounds. Moisture and acid-activated kaolin clay appear to act inter se to eliminate the formation of high-molecular weight polycyclic hydrocarbons and promote the formation of the more desirable lower-molecular weight aromatic hydrocarbons. Since the acid-activated kaolin clay of the specified character is highly sorptive tars which form and might pass out in the smoke are sorbed and upon the approach of the flame-front the tars are revolatilized and undergo additional change. The extent of the results in these respects depends largely on the quantity of acid-activated kaolin clay present with the tobacco.

The moisture serves the additional important function of affixing the finely-divided clay particles to the tobacco and aids in keeping the tobacco fresh.

We have found that a suitable acid-activated kaolin clay for the purpose above set forth is necessarily one which is not deactivated by sintering or by collapse of the internal pore structure thereof at the elevated temperatures of the approaching flame-front, which is usually at a temperature of from about 1500 to 1650° F. For this reason the composition of the material is critical. High silica content materials, such as those synthetically produced by precipitation of silica or co-precipitation of silica with other metallic oxides or by the acid-activation of magnesium-aluminum silicate clay materials, such as bentonite, are unsatisfactory. Likewise, kaolin clay processed to remove a substantial proportion of the aluminum content thereof by leaching, or the like, to remove soluble salts is unsatisfactory for the purpose. All of the synthetically produced catalysts and the high silica content catalysts derived from naturally-occurring clays have the serious disadvantage of sintering at temperatures above about 1000° F. with attendant loss in catalytic and sorptive properties. To our knowledge only the high-alumina content materials derived from the acid-activation of kaolin clays possess the requisite thermal stability for the instant application. It will be understood that the tobacco may be in the form of leaf or shredded or otherwise particulated tobacco which is derived from whole leaf tobacco. Whole leaf tobacco is distinguished from so called "homogenized tobacco," homogenized tobacco being powdered whole leaf tobacco formed into a coherent mass with a combustible binder. Likewise benefits are imparted by addition of the specific acid-activated kaolin clay to homogenized leaf tobacco or to shredded or otherwise particulated tobacco prepared therefrom.

The acid-activated kaolin clay is preferably incorporated in the cigarette tobacco in a very finely-divided form, usually less than about 10 microns and preferably having a substantial portion between about 0.5 and 2 microns. In general, the more finely-divided the material the greater the active surface available to combat deleterious substances and the greater the adhesion of the material to the tobacco particles. It has been found that coarser particles, for example particles approximately 325-mesh, tend to be drawn into the mouth of the smoker upon inhalation whereas finer particles are more resistant to inspiration. Of course the particular particle size used in a composition will depend on such factors as moisture content, presence of tacky humectants, such as glycerol or sorbitol, mode of application to the tobacco, size of tobacco particles, presence of and nature of filter-device adapted to be placed in smoker's mouth, quantity of acid-activated kaolin clay used and locus of placement.

The acid-activated kaolin clay 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. When an unsmoked cigarette is first ignited the effluent smoke contains a smaller amount of tars than when the smoke issues from the more completely smoked cigarette. Any tars produced by the burning of the tobacco are conveyed towards the mouth of the smoker. The elevation of the burning temperature due to the presence of tars promotes the generation of deleterious compounds. Hence, the greatest load on the acid-activated kaolin clay is near that portion of the smoking unit, in the case of a cigarette in particular, adapted to be placed in the smoker's mouth.

A filter, either of the well-known type which is integral with the smoking unit or of the holder type including a filter, is preferably employed in a smoking unit when tobacco is treated by the process herein taught. The filter may be fibrous and/or include adsorbents such as silica, clays or the like. The purpose of the filter is to prevent any inspiration of any finely-divided 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 mixture and the smoker's mouth and imprisons any particles 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.

A cigarette displaying enhanced aroma, outstanding mildness and ease of draw was prepared by thoroughly mixing 1.0 gram of conventional cigarette tobacco with 0.05 gram of a finely-divided acid-activated kaolin clay previously moistened with water to a volatile matter content of about 30 percent and packing this mixture into a paper cylinder having a cork-type tip provided at an end. The acid activated kaolin clay used was "Kaosorb," made by Minerals & Chemicals Philipp Corporation, which was ground to a powder having a maximum particle size of 10 microns and including 50 percent of material finer than 2 microns. "Kaosorb" is an alumina-silica material having an alumina content of about 47.4 percent.

The preparation of the "Kaosorb" is described in the copending U.S. application Serial No. 490,128, filed February 23, 1955, by Alfred J. Robinson et al. In accordance with that invention an acid-activatable clay, preferably kaolin type clay, is mixed with sulfuric acid to an appearance of homogeneity and the homogeneous mixture is reacted at elevated temperature to bring about substantial reaction between the acid and alumina of the clay. The reacted mixture, without any of the water soluble reactants in the mixture washed out, is subjected

to calcination under time and temperature conditions such as to decompose the aluminum sulfate therein and render the product substantially sulfate free. It will be understood, however, that kaolin clay acid-activated by other "dry processes" in which soluble salts are not leached from the clay-acid reaction product may be used in practicing the instant invention.

Furthermore, the invention is not restricted to the method of intermingling the acid activated kaolin clay particles with the tobacco particles by simple mixing of components. For example, a slurry of acid activated kaolin clay particles may be added to leaf or particulated tobacco. This may be done by spraying an aqueous slurry including up to about 40 percent solids on the tobacco, the liquid of the slurry being water, fugitive oxygenated humectant or mixtures thereof. Likewise, a slurry of the particles may be added to so called "homogenized tobacco leaf" or particles of tobacco cut therefrom. Alternatively, the acid activated kaolin clay can be directly incorporated into the homogenized leaf tobacco by forming the comminuted tobacco and additive into a coherent sheet-like mass. In another method the high sorptivity of the additive may be exploited by intimately commingling it with the tobacco, preferably at a somewhat elevated temperature to adsorb wax from the tobacco. Acid activated kaolin clay in excess of the amount called for by the ultimate formulation may be used and the excess acid activated kaolin clay therefrom discarded or processed to remove adsorbed wax and alkaloids. The resultant tobacco with adhering additive particles is then processed according to practices well-known in the art.

The acid activated kaolin clay may be used to replace at least part of the filler present in the paper encasing the column of tobacco. When the additive is used in relatively high loadings the overall appearance of the cigarette may be improved by drably coloring the additive particles by means of a drably colored dye, pigment or material of vegetable origin.

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.

We claim:

1. A smoking mixture comprising shreds of tobacco commingled with a small amount of kaolin clay which has been reacted with sulfuric acid and then thermally desulfated without leaching of water-soluble reaction products therefrom.

2. A smoking mixture comprising shreds of tobacco commingled with a small amount of finely divided kaolin clay which has been reacted with sulfuric acid and then thermally desulfated without leaching of water-soluble reaction products therefrom, said acid-treated clay being further characterized by having an alumina content of 40% to 50% by weight, volatile free basis.

3. A cigarette comprising a paper wrapper enveloping the smoking mixture of claim 1.

4. A cigarette comprising a paper wrapper enveloping the smoking mixture of claim 2.

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