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

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This invention relates to smoking compositions and to smokable units including therein a novel combination of particulate inorganic material intimately associated with tobacco, the additives being capable inter se of substantially reducing the quantity of tars normally generated during the burning of the tobacco and improving the aroma of the 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. One method involves the commingling with smoking tobacco of particulate solids having high sorptivity by virtue of their large specific surface, internal and external. However, at the high temperatures encountered in the combustion zone of tobacco mixtures the sorptivity of materials such as, for example, certain sorptive clays, silica gel, alumina gel and the like, is substantially reduced. The tobacco industry has failed to find that the inclusion of such materials in tobacco compositions is of sufficient value to warrant their use in commercial smoking mixtures. The use of filter-tips has, on the other hand, met widespread acceptance both by the cigarette industry and the consumer. Filter-tips as well as cigarette holders are not, however, without disadvantage. A primary disadvantage of a filter-tip is that its presence interferes with the ease of draw that is characteristic of a well-made cigarette. Another significant disadvantage of a filter-tip is that its presence encourages the smoker to smoke more completely the cigarette at the butt end than he would smoke the cigarette in the absence of the filter-tip. This is so because the filter-tip provides a finger hold for a cigarette so that the intense heat of the advancing combustion zone does not

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force the smoker to extinguish the cigarette when smoked too close to the butt end. It is known that during the course of smoking the sorption of tars towards the cigarette butt by unconsumed tobacco causes relatively heavy deposition of tars proximate the butt end of a cigarette. Hence it is desirable that smoking of a cigarette be ceased prior to the complete pyrolysis of the cigarette.

It is thus 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. Furthermore, substantial benefits result when such an additive inherently possesses the ability to eliminate or substantially reduce the percentage of noxious constituents of the total tar.

Accordingly, it is a principal object of the instant invention to provide a composition including tobacco which incorporates a novel combination of treating agents to reduce substantially the quantity of tars in the effluent smoke of said tobacco.

Another important object of the invention is the provision of smoking units including a novel combination of particulate additives which act synergistically to reduce the quantity of tar generated during smoking of that unit.

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 substantially reduce the quantity of tar generated during smoking of that unit.

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

Briefly, the instant invention is a result of my discovery that an improved smoking composition is provided by the intimate association with tobacco of a mixture of the combination of an acid-activated clay hereafter set forth and hydrated lime. The resultant composition is distinguished by its ability to burn with substantially reduced production of tar per unit of tobacco. A copending U.S. patent application of Serial No. 700,266, filed December 3, 1957, by Frank A. Howard has to do with smoking compositions including tobacco intimately associated with hydrated lime. Said applicant found that tar production of smoked cigarettes was markedly curtailed by the inclusion of hydrated lime in smoking compositions. I have found that the ability of hydrated lime to function in this unique manner is markedly improved by the cooperative effect of certain finely divided acid-activated clays dispersed in the tobacco along with said lime.

More specifically, I have discovered that the addition to tobacco of the combination of certain acid-activated clays and hydrated lime in combined amount of from about 1 to 20% (based on the weight of the tobacco) and preferably from about 2 to 10%, same basis, produces outstanding benefits. However, the optimum ratio of total additive to tobacco to effect the maximum tar reduction consistent with satisfactory ease of draw, ability of the mixture to sustain burning, preservation of redolence and the like, is best determined experimentally for a specific tobacco composition. This is so because the quantity of tars generated during smoking varies considerably, inter alia, with tobacco specie and presence of tobacco treating agents (exemplified by humectants, invert sugar, cocoa, essential oils, chocolate and licorice). Furthermore, commercial smoking mixtures usually comprise a plurality of tobacco species. Suitable ratios of acid-activated clay to hydrated lime lie within the range of from about 1:5 to 5:1. Within the range of about 1:1.5 to 1.5 to 1 marked synergism between additives is usually manifest with respect to total tar reduction when said additives are used with popular smoking mixtures. My invention is not, however, in its broadest aspect restricted to hydrated lime acid-activated clay combinations in which there is synergism with respect to total

tar reduction capacity. Another signal advantage resulting from the combined additive in tobacco is the improved aroma and palatability of smoke effluent from tobacco mixtures including the additive.

The activated clays I employ may contain from 1 to 50% alumina, on a volatile-free weight basis (volatile-free weight being the weight after being heated to substantially constant weight at about 1700° F.) Suitable products are prepared by acid-activation of naturally occurring clays such as kaolin, halloysite and montmorillonite clays. However, because of its outstanding resistance to sintering at the elevated temperatures encountered during the smoking of tobacco, the preferred porous aluminosilicate is one prepared by the acid-activation of kaolin clay and having an alumina content of 40% to 50%, volatile-free weight basis. The preparation of the preferred acid-activated kaolin clay is described in the copending U.S. patent application of 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, 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, and without washing out any of the water soluble products of the reaction, calcining the reactant mixture to eliminate substantially the sulfate therefrom. 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



n usually being 2. The ratio of silica to alumina is about 1.0 to 1.5, and most generally about 1.18. The alumina content of the acid-activated clays is from about 40 to 50% by volatile-free weight, and usually about 46%.

The acid-activated clay commingled with hydrated lime is preferably incorporated in the tobacco of a cigarette 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 additive the greater the surface available to combat deleterious substances and the greater the adhesion of the acid-activated clay to the tobacco particles. Of course it will be understood that the particular particle size used in a tobacco composition will depend on such factors as moisture content, quantity of humectant, size of tobacco particle and quantity of acid-activated clay and hydrated lime used. The specific amount will vary principally with the type and quality of tobacco used and the optimum amount is best ascertained by the smoking characteristics of the composition, such as ease of draw, ability to sustain burning, analysis of combustion products, taste and aroma characteristics.

The hydrated lime is preferably incorporated in the smoking tobacco in finely-divided form, usually less than about 10 microns and preferably having a substantial portion between about 0.5 and 2.0 microns. Dry hydrated lime is commercially available as a fine powder which has good adhesion to the tobacco. It has been found that coarse or agglomerated particles, for example 44 micron particles, are more prone than more finely-divided particles to pass during smoking into the effluent smoke. Of course the particular size used will depend on such factors as moisture content, presence of tacky humectants, presence of an agent to bind the mineral to the tobacco surface, mode of application to the tobacco, size of tobacco particles, presence of and nature of filter-tip in the smoking unit, quantity of particulate additives used and locus of additive placement.

Hydrated lime is in its pure form calcium hydroxide, $\text{Ca}(\text{OH})_2$, a dry powder obtained by treating lime (CaO ,

or, as it is variously termed "quicklime," "burnt lime," "caustic lime" or "live lime") with water. Lime is a highly infusible solid obtained by calcining limestone, shells or other forms of calcium carbonate. Commercial limes commonly contain impurities such as magnesia, alumina, iron oxide and silica. High-calcium limes or "fat limes" contain less than about 5% of magnesia whereas "lean limes" contain in excess of about 5% magnesia. Hence hydrated lime is seldom pure $\text{Ca}(\text{OH})_2$ but is rather $\text{Ca}(\text{OH})_2$ associated with minor amounts of at least one of the following: magnesia, alumina, iron oxide and silica. Accordingly, the term "hydrated lime" as used herein and in the claims encompasses pure $\text{Ca}(\text{OH})_2$ and $\text{Ca}(\text{OH})_2$ associated with minor amounts of such impurities such as are listed above. Furthermore, synthesized $\text{Ca}(\text{OH})_2$ is embraced within the term "hydrated lime."

The additives may be commingled and distributed substantially uniformly throughout the body of the cigarette or, pursuant to an embodiment of the invention, in combination they 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 additives 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 cigarette or of the holder type including a filter, is preferably employed in a smoking unit comprising tobacco mixtures of my invention. The 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 any finely-divided acid-activated clay and/or hydrated lime particles which may occur if the cigarette is subjected to dry, warm weather for prolonged periods. The filter interposes a bed of porous material, or the like, between the tobacco acid-activated clay mixture and the smoker's mouth and imprisons any particle which might otherwise be drawn into the smoker's mouth. However, by proper selection of moisture content and, in some cases inclusion of humectant in the composition, the filter may be omitted.

When the adhesion of a particulate additive to the tobacco is inadequate a binder, such as sodium silicate or a pectic substance, suitably pectin derived from tobacco leaf, may be used to improve adhesion between the tobacco and that additive.

Although the description of the invention has been made with particular reference to cigarettes, it will be understood that the compositions herein provided are applicable to other smoking units.

The following example is given only for the sake of further illustrating my invention and is not to be construed as limiting the scope thereof.

In the example of my invention which follows, 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.

The synergistic effect of the addition of hydrated lime and acid-activated kaolin clay on smoking tobacco with respect to reduction of tar yield per unit is shown in the example. Samples of cigarettes were prepared each comprising 0.90 gram of a popular-brand shredded tobacco blend packed in conventional cigarette paper. The moisture content of the tobacco blend was approximately 12%. Control cigarettes including no particulate additive were prepared. Other cigarettes were made including comminuted hydrated lime in amounts of 5 and 10% based on the weight of the tobacco. They were made up by homogeneously dispersing the dry additive with the tobacco shreds prior to packing. Similarly, a comminuted high alumina content acid-activated kaolin clay made in accordance with the method of said copending Robinson et al. application was dispersed throughout another fraction of tobacco prior to incorporating said tobacco into a cigarette. To another fraction of the tobacco shreds 5% each of acid-activated kaolin clay and hydrated lime were added and homogeneously distributed therethrough. Tar yields obtained by analysis of the condensate of mechanically smoked cigarettes are tabulated below.

Cigarette	Weight of tobacco and Additive per Cigarette (grams)	Tar Yield Per Cigarette
1. Control cigarette	0.90 gram tobacco	35.3±1.3 milligrams (average of 6 runs).
2. Control plus 5% hydrated lime.	0.90 gram tobacco and 0.045 gram hydrated lime.	(30.1) 30.1 milligrams.
3. Control plus 10% hydrated lime.	0.90 gram tobacco and 0.090 gram hydrated lime.	(24.9) 23.7 milligrams.
4. Control plus 5% acid activated kaolin clay.	0.90 gram tobacco and 0.045 gram acid-activated kaolin clay.	34.2±1.3 milligrams (average of 6 runs).
5. Control plus 5% hydrated lime plus 5% acid-activated kaolin clay.	0.90 gram tobacco and 0.045 gram hydrated lime, .45 gram acid-activated kaolin clay.	(23.6) 24.4 milligrams.

The results illustrate the outstanding benefits accruing from the combined addition of hydrated lime and catalyst to cigarette tobacco with respect to total tar reduction. Furthermore, the in vivo smoking characteristics of the cigarette including a composition of my invention was distinguished by outstanding mildness and freedom from bite or harshness.

It will be understood the invention is not restricted to the method of commingling the tobacco and additives described in the example, any suitable means for intimately associating the components being satisfactory. For example, a slurry of additive particles may be added to leaf or particulated tobacco, suitably by spraying a slurry of the particles onto the tobacco. Likewise the additives can be incorporated into so-called "homogenized leaf tobacco" by forming comminuted tobacco and additives into a coherent sheet-like mass in the presence of a suitable binder.

Obviously many modifications and variations of the invention as hereinabove 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.

I claim:

1. A smoking mixture comprising tobacco having commingled therewith a small amount of a mixture of hy-

drated lime and an acid-activated clay selected from the group consisting of kaolin clay, halloysite clay and montmorillonite clay.

2. A smoking mixture comprising tobacco having commingled therewith a small amount of a mixture of hydrated lime and acid-activated kaolin clay.

3. A smoking mixture comprising tobacco having commingled therewith a small amount of a mixture of hydrated lime and acid-activated kaolin clay having an alumina content of 40% to 50% on a volatile-free weight basis.

4. A smoking mixture comprising tobacco having commingled therewith a small amount of a mixture of hydrated lime and sulfuric acid-treated kaolin clay which has been thermally desulfated and which has an alumina content of 40% to 50% on a volatile-free weight basis.

5. A smoking mixture comprising shreds of cigarette tobacco having commingled therewith a small amount of a mixture of hydrated lime and sulfuric acid-treated kaolin clay which has been thermally desulfated and which has an alumina content of 40% to 50% on a volatile-free weight basis.

6. A smoking mixture comprising shreds of tobacco having commingled therewith a small amount of a mixture of equal proportions of hydrated lime and acid-activated kaolin clay having an alumina content of 40% to 50% on a volatile-free weight basis.

7. A smoking mixture comprising tobacco shreds having commingled therewith about 10% by weight of a finely divided mixture of equal proportions of hydrated lime and sulfuric acid-treated kaolin clay which has been thermally desulfated and which has an alumina content of 40% to 50% on a volatile-free weight basis.

8. A cigarette comprising a paper wrapper enveloping shreds of tobacco, said shreds being commingled with a small amount of a finely divided mixture of hydrated lime and an acid-activated clay selected from the group consisting of kaolin clay, halloysite clay and montmorillonite clay.

9. A cigarette comprising a paper wrapper enveloping shreds of tobacco, and mixed with said shreds a small amount of a mixture of hydrated lime and acid-activated kaolin clay having an alumina content of 40% to 50% on a volatile-free weight basis.

10. A cigarette comprising a paper wrapper enveloping shreds of tobacco, and mixed with said shreds about 10% by weight of a mixture of hydrated lime and acid-activated kaolin clay having an alumina content of 40% to 50% on a volatile-free weight basis.

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