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TREATMENT OF SMOKING TOBACCO WITH CHLORATE SALTS

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ABSTRACT OF THE DISCLOSURE

The amount of fluorescent polynuclear aromatic hydrocarbons in tobacco smoke is reduced by adding to the tobacco a metal salt of chloric acid, e.g., alkali metal or alkaline earth metal chlorates.

This invention relates to additives for smoking tobacco and to smoking devices such as, for example, cigarettes made with tobacco containing such additives. More particularly, this invention concerns the addition of a chemical additive to smoking tobacco for reducing certain noxious constituents normally produced when the tobacco is smoked.

Among the undesirable components found in tobacco smoke are the polynuclear aromatic hydrocarbons. These specific components (polynuclear aromatic hydrocarbons) of tobacco smoke merit special attention inasmuch as the ones that are present in cigarette smoke condensates are believed to be carcinogens (cancer producing substances) and have been shown to be at least partially responsible for the tumorigenicity of cigarette smoke condensate upon animal testing [E. L. Wynder and D. Hoffmann, Acta. Path. Microbiol. Scand., 52, 119 (1961)]. From this data, workers in this area of research have concluded that these compounds should be eliminated from cigarette smoke [Smoking and Health, U.S. Public Health Service Publication No. 1103, Ch. 9, p. 142 and p. 146 (1963)]. Therefore, any method or improvement for decreasing the amount of polynuclear aromatic hydrocarbons in cigarette smoke would appear to be highly beneficial.

In attempting to lower the polynuclear aromatic hydrocarbon content of tobacco smoke, various treatments of the tobacco materials have been proposed and tried by the tobacco industry. Among the materials and treatments proposed for use in or with the tobacco was a number of well-known nitrates such as potassium and copper nitrates. Such nitrate-containing tobacco is effective to some extent in reducing polynuclear aromatic hydrocarbons such as 3,4-benzopyrene from the cigarette smoke, but their use has some definite disadvantages. For example, nitrate or nitrite salts have the serious disadvantage of producing large amounts of highly toxic oxides of nitrogen in the smoke. Sodium dichromate and potassium permanaganate have also been suggested for use as a polynuclear aromatic hydrocarbon reducing agent. However, as a result of tobacco having a tendency to rapidly reduce these materials, they are ineffective for this purpose. Furthermore, these materials produce very undesirable by-products when subjected to high temperatures.

Therefore, it is readily apparent that some improvement in the tobacco art which would permit tobacco to be smoked with less polynuclear aromatic hydrocarbons being released would represent a substantial stride forward. Ideally this improvement should be obtained using a common, readily obtainable material or materials that can be applied or used in a continuous process compatible with existing manufacturing techniques thereby adding no appreciable cost to the manufacturing expense of the tobacco product. Furthermore, and perhaps most importantly, the new tobacco product should not adversely affect the flavor or aroma of the tobacco smoke.

It is known that polynuclear aromatic compounds are highly fluorescent [J. Natl. Can. Inst., 21, No. 1, 1 (1958)]. Advantage is taken of that fact in analyzing particulate matter from cigarette smoke since a decrease in fluorescence concentration shows a decrease in polynuclear aromatic hydrocarbon concentration.

According to the present invention it has been found that a substantial portion or percentage of the polynuclear aromatic hydrocarbons normally found in the smoke of a natural leaf smoking material can be eliminated therefrom by adding certain quantities of salts of chloric acid to the material. The chlorate salts act upon the smoking material such as, for example, tobacco to change its burning characteristics thereby preventing the initial formation of the deleterious hydrocarbons.

Therefore, an object of this invention is to provide a chemical additive for natural leaf materials which will substantially eliminate certain deleterious substances normally produced when the material is burned.

Another object of this invention is to disclose a tobacco mixture containing a chemical additive for reducing the polynuclear aromatic hydrocarbons and other deleterious fluorescent materials produced when the tobacco is smoked.

Still another object of this invention is to disclose a nontoxic chemical additive for tobacco which will alter the burning characteristics of the tobacco in a manner that will decrease the amounts of carcinogenic polynuclear aromatic hydrocarbons to be found in the tars of the smoked tobacco, yet will not produce toxic materials themselves while reducing the harmful hydrocarbons in the combustion products of the tobacco.

Yet still another object of this invention is to disclose a cigarette which produces less of polynuclear aromatic hydrocarbons when smoked.

A further object of this invention is to disclose a combination of tobacco with chlorate salts formed from metals of Groups I*a*, I*b*, II*a*, II*b*, III*a*, III*b*, IV*a*, IV*b*, V*a* and the transition metals for reducing the polynuclear aromatic hydrocarbons contained in the total particulate matter produced when the material is smoked.

Yet still another object of this invention is to disclose a method of treating shredded tobacco with a chemical additive for decreasing the amounts of certain deleterious products produced when the tobacco is smoked.

A still further object of this invention is to disclose a process for producing a cigarette which when smoked produces a minimum of polynuclear aromatic hydrocarbons.

These and further objects and advantages of this invention will be more apparent upon reference to the accompanying specification, specific examples and claims.

Over the past years various additives have been included into natural leaf smoking products for accomplishing a variety of objectives. Among these objectives are tar reduction, lowering of the burning-zone temperature, reduction of "bite," and improvement of the flavor of the smoke. However, until recently little attention has been directed toward finding an additive for natural leaf smoking materials which would prevent or greatly reduce the formation of polynuclear aromatic hydrocarbon components in the particulate phase of the smoke. In accordance with the present invention it has been found that a definite and specific quantity of metal salts of chloric acid added to tobacco, for example, substantially reduces the percentage of the carcinogenic hydrocarbons that are produced when the tobacco is smoked in a cigarette. This improvement in the quality of the tobacco smoke was accomplished without sacrificing any of the

flavor or aroma of the smoke. The discovery that chlorate salts of this type would reduce the hydrocarbon content of tobacco smoke as well as the extent of the reduction was quite unexpected since various compounds that are similar to chlorate salts have normally been thought to be ineffective for this purpose, or, if effective, to give highly toxic by-products when heated.

The chlorate salt additives contemplated by this invention can be incorporated into the tobacco in any desirable manner. For example, solutions of the chlorate salt additives in a suitable solvent, such as water, may be applied to the tobacco as by spraying, soaking, sprinkling or the like after which the solvent is driven off as a vapor leaving the additive thoroughly incorporated with the tobacco. The chlorate salt additives may also be applied as a finely-divided solid material through a dusting, shaking or dispersing medium of any suitable tyre which will uniformly disperse the additive over the tobacco. The incorporation of the additives may take place at any time prior to the final packaging of the tobacco product. In the case of cigarette tobacco it may be incorparted before or after blending of the various tobaccos if, in fact, blended tobacco is employed, and the additive may be applied to one or all of the blend constituents.

The amount of additive in the final product contemplated by this invention is quite small but the specific amount employed is rather critical. Generally speaking, however, desirable reduction of polynuclear aromatic hydrocarbons can be obtained if the metal salt of chloric acid is incorporated into the final tobacco product in amounts between about 2.0 and 10 percent by weight. It has been found that at least 2.0% metal chlorate is necessary in order to realize a significant decrease in fluorescence per weight of "tar" and therefore a significant decrease in the concentration of polyaromatic hydrocarbon in the "tar." Since the cigarettes burn slightly faster with 5–6% metal chlorate added it is likely that more than 10% metal chlorate added will result in a prohibitively rapid burning rate. Thus, the limitations of this invention are between 2.0 and 10% metal chlorate added to tobacco. However, optimum reduction in the polyaromatic hydrocarbons in the tobacco smoke is obtained when about 6 percent of chlorate salt additives is used.

As mentioned briefly hereinabove, the chlorate salts that have been found to be useful as additives to tobacco to produce cigarettes which, upon smoking, produce less polyaromatic hydrocarbons and other fluorescent materials than cigarettes made from tobacco which does not contain these salts are salts including mteals from Groups Ia, Ib, IIa, IIb, IIIa, IIIb, IVa, IVb Va and the transition metals. The preferred salts are the alkali metal and alkaline earth salts such as, for example, the chlorates of sodium, potassium, barium and magnesium.

The explanation of how metal chlorates operate to decrease the concentration of fluorescent material in cigarette smoke is not known. However, one theory appears particularly plausible. Nitrate salts have been shown to decrease the concentration of fluorescent material in cigarette smoke. Sodium nitrate is particularly effective and this compound decomposes at about 400° C. to yield oxygen, nitric oxide and sodium oxide. It has not been proved but it is quite possible that sodium nitrate functions by releasing oxygen at the correct temperature and position with respect to the burning coal. In addition, polynuclear aromatic hydrocarbons or their precursors may react with sodium nitrate at about 400° and thus be oxidized. Therefore, potassium chlorate and other like chlorate salts probably also function in one of these ways since they are oxidizing agents and are known to release oxygen at about 400° C. However, the chlorate salts do not oxidize the tobacco at or near room temperature, unlike the cases of other oxidizing agents previously suggested. The lack of non-oxious by-products of the combustion of chlorates and the stability of chlorates in contact with tobacco at ordinary temperatures makes the chlorates an unobvious and unique tobacco additive for the purposes mentioned herein.

A further understanding of the invention will be had from a consideration of the following examples that may be used in actual commercial practice and are set forth to illustrate certain preferred embodiments.

EXAMPLE I

Tobacco used in the two following examples was obtained by removing the paper from cigarettes made from a typical domestic blend of tobacco. After the tobacco was removed from the cigarettes, 30 ml. of distilled water was blended dropwise into 50 g. of the tobacco as the tobacco was stirred. The wet tobacco was then air dried to its original weight, tumbled for about 16 hours and fabricated into 85 mm. cigarettes containing 1.0 g. each of tobacco. The cigarettes were equilibrated to approximately 12% moisture and these were used as control cigarettes.

To 47.0 g. of tobacco was added, in the same way, 30 ml. of distilled water containing 3 g. of potassium chlorate. The tobacco plus potassium chlorate was air dried to a weight of 50.0 g. so that the tobacco contained 6% potassium chlorate. The subsequent treatment was the same as for the control cigarettes with 1.0 g. of tobacco plus additive being used to make each cigarette.

The total particulate matter from five of the control cigarettes and five of the potassium chlorate-treated cigarettes was collected on separate, tarred, Cambridge filter assemblies using an automatic smoking machine [described by Mumpower and Touey, Tob. Sci. 5, 31 (1961)] which smoked the cigarettes at a rate of one 35 ml. puff per minute of 2-sec. duration. The cigarettes were smoked to a 30 mm. butt length and the Cambridge filter assemblies reweighed after smoking to furnish the weight of total particulate matter delivered. This method for collecting and weighing total particulate matter delivered by a cigarette has been described by Ogg [J. Assoc. Offic. Agri. Chemiss, 47, 356 (1964)]. After analyzing the two samples of total particulate matter for moisture by the method of Sloan and Sublett [Tob. Sci., 9, 70 (1965)] it was found that the control cigarettes delivered 31.0 mg. of dry total particulate matter per cigarette while the potassium chlorate-treated cigarettes delivered 29.6 mg of dry total particulate matter per cigarette.

The total particulate matter of both the control and potassium chlorate-treated cigarettes was dissolved in methanol and the fluorescence of each sample of total particulate matter was determined on a scale of fluorescence units using the method described by McConnell, Mumpower and Touey [Tob. Sci. 4, 55 (1960)]. This analysis step was employed since the known polynuclear aromatic hydrocarbons in cigarette smoke are highly fluorescent so that a decrease in fluorescence intensity indicates a decrease in polynuclear aromatic hydrocarbon concentration in cigarette smoke condensate. Over the range of concentration of total particulate matter used the relationship between fluorescence and concentration is linear. The fluorescence of the sample from control cigarettes was found to be 2.08 fluorescence units per mg. of total particulate matter. The fluorescence of the sample from the potassium chlorate-treated cigarettes was found to be 1.50 fluorescence units per mg. of total particulate matter. Therefore, the addition of potassium chlorate to tobacco results in a cigarette which burns to produce 28% less fluorescent material than the same tobacco with no potassium chlorate added.

EXAMPLE II

The process of Example I was repeated except that tobacco was used which contained 5% sodium chlorate. Control cigarettes were found to deliver 32.2 mg. of dry total particulate matter per cigarette and cigarettes containing 5% sodium chlorate were found to deliver 31.1 mg. of dry total particulate matter per cigarette. The

fluorescence of the sample from control cigarettes was found to be 2.08 fluorescence units per mg. of total particulate matter. The fluorescence of the sample from cigarettes containing sodium chlorate was found to be 1.60 fluorescence units per mg. of total particulate matter which represents a 23% reduction in fluorescent material.

From the foregoing it will be readily apparent that the use of a chlorate salt tobacco additive of the type and in the manner hereinabove described substantially reduces the carcinogenic polynuclear aromatic hydrocarbons from tobacco smoke. Furthermore, not only is over a20 percent reduction of the polynuclear aromatic hydrocarbons realized by using the chlorate salt additives but the additives are extremely easy to apply to natural leaf smoking materials by known techniques employing equipment already available to the tobacco industry. This coupled with the low cost of the additives themselves permits tobacco products containing the additives to be manufactured at a price competitive with existing commercial tobacco products. The use of the additives is also advantageous since it does not in any way adversely affect the flavor or aroma of the smoke as has heretofore occurred when certain other additives were employed, nor is a special and expensive filtering unit necessary.

What is claimed and desired to be secured by the United States Letters Patent is:

1. As a new article of manufacture, a natural leaf smoking tobacco composition containing a non-toxic additive selected from the group consisting of sodium chlorate and potassium chlorate, said additive constituting from about 4 to about 10 percent by weight of the composition, said additive functioning to at least partially reduce the polynuclear aromatic hydrocarbons normally present in the smoke of said leaf tobacco.

2. The method of treating natural leaf smoking tobacco so as to reduce the polynuclear aromatic hydrocarbons that are normally produced when the material is smoked, comprising applying prior to use, of from 4 to 10 percent by weight of a salt of chloric acid selected from the group consisting of sodium and potassium chlorate.

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