United States Patent

[11] 3,584,630

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New York, N.Y.
Continuation of application Ser. No. 603,419, Dec. 21, 1966, now abandoned.

[54] TOBACCO PRODUCT HAVING LOW NICOTINE CONTENT ASSOCIATED WITH A RELEASE AGENT HAVING NICOTINE WEAKLY ABSORBED THEREON

1 Claim, No Drawings

[52] U.S. Cl. 131/9, 131/267, 131/10.9
[51] Int. Cl. A24d 01/06
[50] Field of Search ..... 131/9, 10, 10.5, 10.7, 261—269, 23, 10.9

[56] References Cited

UNITED STATES PATENTS

2,063,014	12/1936	Allen	131/17 X
3,236,244	2/1966	Irby et al	131/10
3,280,823	10/1966	Bavley et al.	131/10

FOREIGN PATENTS

1,185,164	9/1965	Germany	131/261 UX

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ABSTRACT: This disclosure relates to a tobacco product containing a nicotine-releasing agent and to a method for releasing a controlled amount of nicotine into tobacco smoke. The nicotine-releasing agent comprises a finely divided, weak absorbent of gas furnace or channel carbon black, the said carbon black having on its surface weakly adsorbed per part of weak adsorbent. The nicotine is released from the weak adsorbent during smoking to bring the nicotine content in the smoke generated by a low nicotine tobacco with which it is associated, to a total level of 0.1—3.0 mg.

TOBACCO PRODUCT HAVING LOW NICOTINE CONTENT ASSOCIATED WITH A RELEASE AGENT HAVING NICOTINE WEAKLY ABSORBED THEREON

This application is a continuation of Ser. No. 603,419 filed Dec. 21, 1966, now abandoned.

This invention relates to a tobacco product containing a nicotine-releasing agent and to a method for releasing a controlled amount of nicotine into tobacco smoke. More particularly, the invention relates to tobacco products containing an agent for releasing nicotine into tobacco smoke which agent permits storage of tobacco products containing said agent for prolonged periods of time, without substantial loss of the nicotine associated with said agent. The agent provides for the release of nicotine in controlled amounts, when tobacco smoke is passed in contact with the nicotine-containing agent.

It has long been known in the tobacco industry that in order to provide a satisfying smoke, it is desirable to maintain the nicotine content of tobacco products at a uniform level. However, it is difficult to accomplish this result since the nicotine content of tobacco varies widely, depending on the type of tobacco and the conditions under which the tobacco is grown.

Among the factors affecting the nicotine content of any variety of tobacco are the conditions which exist during the growth of the tobacco, for example, the moisture conditions, the type of soil, the fertilizers that are employed, the number of tobacco plants per acre and the care which is given to the plants during their growth. The nicotine content also varies widely, depending on the variety of tobacco. Many of the newer varieties of tobacco plants yield tobacco which is low in nicotine. Furthermore, methods of preparing tobacco products frequently remove some or all of the nicotine that is naturally present in the tobacco. In addition, modern technology has made it possible to utilize portions of the tobacco plant other than the leaf for smoking and some of these portions, such as the petioles, are low in nicotine content.

Maintaining the nicotine content at a sufficiently high level to provide the desired physiological activity, taste, and odor which this material imparts to the smoke, without raising the nicotine content to an undesirably high level, can thus be seen to be a significant problem in the tobacco art. The addition of nicotine to tobacco in such a way that it remains inert and stable in the product and yet is released in a controlled amount into the smoke aerosol when the tobacco is pyrolyzed, is a result which is greatly desirable.

The present invention provides a solution to this longstanding problem and results in accurate control of the nicotine which is released in tobacco smoke. By employing the nicotine-releasing agents and methods of the present invention, it is possible to incorporate exact amounts of nicotine in a tobacco composition, which will remain constant over extended periods of time and which will ultimately yield a smoke containing a controlled amount of nicotine.

The present invention may also be used to incorporate flavorants into tobacco smoke.

Often flavor or flavors which are incorporated in tobacco, are lost or altered during subsequent manufacturing steps, or storage. It can also be difficult to control the amount of flavor released during the smoking of a tobacco product to insure uniformity of tobacco flavor during the entire smoking process.

While many efforts have been made to introduce flavors into tobacco smoke, no completely satisfactory method has been found. For example, a method has been employed, whereby flavoring gases have been incorporated into the tobacco in the form of stable esters which decompose upon pyrolysis. Such a method, however, is only useful for certain types of flavorants and also may result in the incorporation into the smoke of breakdown products of the pyrolysis of the esters.

The increasingly widespread use of filter tip cigarettes, has further complicated the efforts to solve the above-mentioned longstanding problems. The various filters are efficient absorbers which extract from the smoke nicotine, as well as many other flavor components, and thus can deprive the smoke of the qualities which are desired by smokers.

However, while the present invention can be employed to incorporate flavors into tobacco smoke, it is particularly useful for the maintenance of the proper amount of nicotine in tobacco smoke. Thus, the discussion which follows and the examples are directed essentially to the use of a nicotine-containing agent in a tobacco product.

With regard to the above-discussed problems of maintaining the nicotine content of tobacco products to the proper level to provide the desired qualities in tobacco smoke, previous efforts have been made to add nicotine to tobacco products wherein the nicotine level in the tobacco was undesirably low.

Such efforts have included adding nicotine per se to the tobacco. However, it has not been found feasible to add nicotine per se to tobacco products. For one thing, the nicotine can be absorbed through intact skin and is, thus, difficult and hazardous to handle in processing operations. In addition, free nicotine is a very volatile material and will volatilize readily at room conditions. Therefore, the addition of nicotine into a tobacco product as the free material could readily result in a substantial loss of the nicotine during storage of the tobacco product. Even though the nicotine content of tobacco products could, by the addition of nicotine content of tobacco products could, by the addition of nicotine under conditions involving considerable effort, be made initially uniform, the volatization losses attending storage of the product would not provide smoke containing a uniform amount of nicotine.

In U.S. Pat. No. 3,109,436, a solution to the problem of adding nicotine to tobacco products has been set forth. In that patent, nicotine-ion exchange resins are incorporated in the tobacco. The nicotine-ion exchange resins decompose upon smoking of the tobacco to result in incorporation of the nicotine in the smoke. The present invention provides even greater improvements than those obtained in accordance with that invention, however, in that there are no ion exchange breakdown products introduced into the smoke.

In U.S. Pat. No. 3,280,823, a further solution to the problem has been provided. In that patent, the nicotine-ion exchange resins are incorporated in filters for tobacco smoke. The invention involved in that patent provides an improvement over the invention set forth in U.S. Pat. No. 3,109,436, in that the use of the ion exchange resin in the filter eliminates the incorporation in the tobacco smoke of ion exchange breakdown products. The present invention provides even greater improvements than those obtained in accordance with the invention set forth in U.S. Pat. No. 3,280,823.

When the present agents are incorporated in tobacco, they provide an efficient release of the nicotine into the tobacco smoke and do not involve incorporation into the tobacco smoke of any undesirable breakdown products. Thus, the present invention overcomes the disadvantages of the method described in U.S. Pat. No. 3,109,436. When the present agents are incorporated in the filter for tobacco smoke, superior results to those obtained by following the teachings of U.S. Pat. No. 3,280,823 are obtained, in that they provide more efficient transfer of nicotine from the agent to the tobacco smoke. They also provide a more economical method of incorporating nicotine into a tobacco product, since they are more easily prepared and are less expensive than the nicotine-ion exchange resins.

It is an object of the invention to provide a method for treating tobacco smoke which overcomes the above-mentioned disadvantages of the prior art.

It is another object of the invention to provide an agent for the treatment of tobacco smoke whereby nicotine is easily released thereinto in controlled amounts.

In accordance with the present invention, an agent for incorporation in a tobacco product is provided which comprises a finely divided weak absorbent having nicotine disposed on its surfaces.

As used herein the term "weak adsorbents" is intended to include "primary weak adsorbents" and "secondary weak adsorbents" and includes adsorbents composed of particles having the following characteristics:

1. The surface of the particles is principally formed by the exterior of the particles, in contrast to the surfaces formed by the pores and fissures, if any, of the particles or the particles are hydrophilic in nature and, thus, hold moisture in a manner whereby the nicotine is readily displaceable by the moisture present in tobacco smoke coming in contact therewith;
2. The particles do not tend to form chemical bonds with nicotine; and
3. The particles absorb substantially no nicotine.

The term "weak adsorbents" if intended to include materials having a specific surface greater than one square meter per gram and the "primary weak adsorbents" and "secondary weak adsorbents" relate, respectively, to:

a. adsorbents, the surface of which is principally formed by the exterior of the particles and
b. particles which are hydrophilic in nature, as set forth in (1) above and possessing the other properties which are set forth above.

The weak adsorbent, as it is employed in accordance with the present invention, holds the nicotine on its surface during storage of the tobacco product and releases the nicotine into the tobacco smoke which forms when the tobacco product is smoked.

Tobacco products into which the present agent may be incorporated include filter tip and nonfilter cigarettes, cigars and cigarillos, as well as pipe tobacco, pipe filters and other products through which tobacco smoke passes on the way to the smoker's mouth. The agent can be incorporated directly in the tobacco or can be incorporated in the filter for tobacco smoke. Preferably, the agent is employed in a filter, since this method removes any possibility of decomposition of the nicotine during combustion of the tobacco and since the method provides for the most accurate control of the amount of nicotine which is incorporated in the smoke.

In accordance with the present invention, it has been found that not all adsorbent materials will provide a base for nicotine, which base will hold the nicotine during storage but from which nicotine can be easily removed into a passing smoke stream. It has been found that the adsorbent, to be effective in accordance with this invention should be a finely divided, weak adsorbent, of limited adsorptive ability. For example the weak adsorbent may be a primary weak adsorbent, such as carbon black, lampblack, gas furnace black, channel black bentonite, titania, calcium or magnesium silicate, hydrated alumina, and the like. Other primary weak adsorbents which may be employed include kaolin, montmorillonite, attapulgite, and finely divided polyacrylonitrile, microcrystalline cellulose, polyvinylidene chloride and similar nonabsorbent polymers.

Somewhat stronger adsorbents than those set forth above may also be used. However, when such adsorbents are employed, they should have a high affinity for water, and can be categorized as hydrophilic adsorbents. Such hydrophilic adsorbents are herein designed as "secondary weak adsorbents" and include materials such as silica gel, alumina, aluminosilicates, activated alumina, large pore zeolites and the like. The high moisture content of tobacco smoke causes the hydrophilic adsorbents to release the nicotine or other tobacco additive deposited on their surface, while they tend to adsorb, due to their affinity to water, the moisture content of the smoke. The proportion of smoke moisture so adsorbed, however, is small.

In accordance with a preferred embodiment of the invention, the tobacco treating agent forms a part of a filter tip, or a portion of a filter tip, for a smoking article such as a cigarette. The nicotine is deposited on the finely divided particles of the mild adsorbent or hydrophilic adsorbent carrier which has the capability of holding the nicotine in readiness to be dislodged by contact with the smoke. The coated carrier can be placed into a chamber of the filter tip, but preferably, the carrier, with its additive coating, is applied onto a tow or web filter substrate, to be less compact and more readily permit the passage of smoke therethrough. This latter combination can form part of the filter tip.

The nicotine can be deposited on to the surface of the adsorbent carrier of the invention by any method which permits relatively uniform distribution of the nicotine on the surface of the carrier. Well-known methods for this purpose include impregnation of the carrier by soaking it in a nicotine solution, spraying it with a nicotine solution, adsorption on the carrier of nicotine from a nicotine solution, condensation of nicotine from vapor phase on the carrier, and the like. For treating the weak adsorbents such as carbon black, the nicotine solution can be added to the adsorbent and the resulting mixture stirred or tumbled in order to thoroughly blend the same. In the case of weak adsorbents of the hydrophilic adsorbent-type, preferably these materials are soaked with a solution of nicotine and a relatively nonpolar solvent, followed by filtration of the mixture and evaporation of any solvent retained on the nicotine-containing adsorbent.

The proportion of nicotine to dry adsorbent will generally be between 1:20 and 1:1, on a weight basis, respectively, and will preferably be between 1:10 and 1:2, respectively.

The amount of nicotine (in nicotine-adsorbent form) added to a filter, on a cigarette basis, may be from 2.0 to 35.0 milligrams per cigarette and preferably from 4.0 to 12.0 milligrams per cigarette, depending on the amount of nicotine in the tobacco. Only sufficient nicotine should be added to the tobacco product to result in a nicotine content in the tobacco smoke approximating that of a conventional tobacco product.

The amount of nicotine-adsorbent that is added to the tobacco will vary depending upon the nicotine content originally present in the tobacco and upon the nicotine content desired in the smoke. Generally, the nicotine content of the tobacco is importantly to a level whereby there is 0.1—3.0 mg. of nicotine in the smoke per cigarette.

One method of determining how much of a particular nicotine adsorbent to add to a particular tobacco product is to analyze the smoke from the tobacco product in which the nicotine-adsorbent agent is incorporated, which can be done by conventional methods, such as is described in the "Journal of the Association of Official Agricultural Chemists" (vol. 42) (Nov. 2, 1959) on pages 424—429. In accordance with this method, an aqueous solution of smoke particulate phase is steam distilled under appropriate conditions; this is followed by spectrophotometric examination of the resulting distillate in the ultraviolet spectral region.

The directions for the details of this procedure include the following:

Collect the smoke particulate phase from 10 successive cigarettes, using a standard robot smoking procedure (35 ml. puff volume taken over a 2 second interval, once per minute) upon a glass wool plug or an equivalent collection medium suitable for separation of 0.1 μ particles and larger from a gas particulate phase mixture. Strip the nicotine alkaloids from the collection medium with four 10 ml. portions of 0.05 N HCl. Combine the separate eluates and dilute to exactly 50 ml. with 0.05 N HCl. Transfer a 10 ml. aliquot of this solution to the port of the distillation unit. Distill approximately 100 ml., and discard. Make the sample in the unit alkaline by adding a size 00 capsule of NaCl and a size 00 capsule of Na0 H, in that order. Repeat the distillation and collect a second 100 ml. portion, as follows: distill approximately 98 ml. into a 100 ml. volumetric flask to which 5 ml. 3 N H_2S0_4 has previously been added. Dilute the distillate exactly to mark with distilled water, mix well, and examine spectrophotometrically between 230 and 300 μ. Correct the absorbance of the unknown solution at approximately 260 μ by a baseline selected on the basis of the curve. Draw a line from the lowest point on both sides of the maximum absorbance. The difference between the baseline and peak at the wavelength of the peak is taken

as the absorbance of the sample. Compare this corrected absorbance to the absorbance of the standard nicotine solution corrected in the same manner, and calculate the nicotine content of the sample directly from this comparison, using standard spectrophotometric technique.

Mg. nicotine alkaloid =(A/A')X(mg. known per ml. X dilution factor/no. cigarettes in sample); where A is corrected absorbance of unknown, and A' is corrected absorbance of known.

The nicotine-adsorbent content for the tobacco can, by making tests as set forth above, be adjusted to bring the nicotine in the smoke to within the range which is desired.

The invention, accordingly, comprises the several steps and the relation of one or more of such steps with respect to each of the others, and the article possessing the features, properties, and the relation of elements, which are exemplified in the following detailed disclosure.

Example 1

A mixture of 1 gram nicotine sold by the Eastman Chemical Company, under the designation "White Label" was blended by manual stirring with 5 grams of gas furnace carbon black sold by the Cabot Corporation under the trade name "Sterling R." The mixture was set aside overnight and was then weighed. The weight loss was found to be 0.3 percent. A 0.6 gram portion of this mixture was spread evenly over a thin cotton web. The coated web was rolled transversely and set into a hollow paper tubing having an inside diameter of 7.8 mm. and the resulting rod was cut into 1 cm. lengths. The sections, corresponding to the diameter of a conventional cigarette, had a resistance-to-draw of 1.5—2 inches of water, as determined by a "Magnehelic" gauge connected to the line in which the filter sections were inserted and through which air was drawn at the rate of 600 cc./minute.

The sections were found to contain 0.054 grams of the mixture. Cigarettes were prepared, with the sections prepared in accordance with the foregoing, attached to the tobacco rod, followed by a 5 mm. long section of a conventional cellulose acetate backup filter, with a resistance-to-draw, as determined above, of 1.2 inch of water. The smoke of the cigarettes prepared in accordance with this example was found by the method describe above to contain 1.61 mg. nicotine per cigarette. Control cigarettes made with conventional filters, but without the implement of the invention, having the same resistance-to-draw, delivered only 1.14 mg. nicotine.

Example 2

A solution of 2 grams nicotine of the type used in Example 1, in 20 ml. *n*-hexane was applied to 2 grams of dry silica gel sold by the Davison Chemical Division of W. R. Grace & Company under the trade designation "Grade 62," and having a particle size of 60X200 mesh. The solution was applied to the silica gel carrier by manual stirring. After 3 hours resting, the solid was separated by filtration, and air-dried for 2 hours. The weight of the silica gel increased by 26.5 percent and was found, by the method described above to contain 18.8 percent total alkaloids, which represented 23.1 percent of the weight of the silica gel.

The nicotine-coated silica gel was spread evenly over a thin cotton web which was rolled to fit into paper tubing of cigarette diameter. The resulting rod was cut into 1 cm. lengths, each containing about 31 mg. of the nicotine-coated silica of which 5.9 mg. was nicotine. The resistance-to-draw of the filter plugs was found to be 2.1—2.3 inch of water. The plugs were attached to the ends of cigarette tobacco rods, making thereby the plugs into cigarette filter tips, the outer end of the tip then was backed up by 5 mm. lengths of conventional cellulose acetate filters having a resistance-to-draw of 1.2 inch of water. The analysis of the smoke by the method described above showed 1.47 mg. nicotine, as compared to the smoke obtained for control cigarettes identically made, but without the silica-nicotine and with a cellulose acetate filter tip of slightly lower resistance-to-draw, as control, the smoke of which contained only 1.14 mg. nicotine.

Example 3

A cigarette equipped with auger high-efficiency filter containing in one section 83 mg. of an activated coconut charcoal delivered 0.58 mg. of nicotine in the smoke. When the same cigarettes had nicotine applied to the charcoal in quantities of 9, 15 and 25 mg., the increase in nicotine delivery was 0.16, 0.29 and 0.90 mg., respectively. These represent 1.8, 1.9 and 3.6 percent of the added nicotine, a decidedly lower proportion than that shown in Examples 1 and 2, even for the uneconomically high 25 mg. level. This example illustrates the disadvantage of using a strong but nonhydrophilic adsorbent as a carrier for nicotine.

Example 4

To 5.0 g. of "Monarch" 81 channel carbon black (Cabot Corp.) was added 1.0 g. of nicotine (Eastman Chemical Co., white label) and the mixture was blended by hand. This mixture was blended with the tobacco filler of handmade cigarettes at the rate of 30 mg. per cigarette. Cigarettes were also made up from the same filler without the added nicotine-carbon, both rods having the same weight and resistance-to-draw. The 65 mm. rods were attached to 20 mm. filters of cellulose acetate having high efficiency of particulate removal. The experimental cigarettes delivered an average of 1.4 mg. of nicotine per cigarette; those without the additive delivered 0.78 mg. in the smoke.

Example 5

Nicotine as a 10 percent by weight solution in *n*-hexane was applied to activated alumina.

After standing overnight, the alumina was separated by filtration and air-dried and was found to contain 14.5 percent of nicotine, based on total weight. This material was spread evenly on a thin cotton web which was then rolled and fed into a tube of stiff paper having the diameter of a cigarette; the weight of alumina-nicotine was 65 mg. per cm. of length (or 9.5 mg. nicotine). The filter tube was cut into 1-cm. sections which were attached to cigarette rods together with a 1-cm. backup filter of cellulose acetate. Filters 2-cm. long consisting only of cellulose acetate and having total resistance like the preceding were attached to the same type of cigarette rods to provide controls. Samples with additive delivered 1.6 mg. nicotine to the smoke; the controls delivered 1.1 mg.

It can thus be seen that the objects set forth, and those made apparent from the preceding description, are effectively obtained, and since certain changes may be made in carrying out the above process and in the article set forth without departing from the scope of the invention, it is intended that all matter contained in the above description shall be interpreted as illustrative and not in a limiting sense.

Having described my invention, what I claim as new and desire to secure by Letters Patent is:

1. A tobacco product comprising a tobacco section and a filter section, said filter section including a weak adsorbent agent of limited adsorptive ability therein selected from the group consisting of gas furnace or channel carbon black, said carbon black adsorbent having a specific surface area greater than 1 square meter per gram, and a charge of nicotine weakly adsorbed on the surface said carbon black, the tobacco of the tobacco section being of a low nicotine content, the smoke from which has its nicotine content increased to a total value of 0.1—3.0 mg. of nicotine upon release of the nicotine from the weak carbon black adsorbent during smoking.

UNITED STATES PATENT OFFICE

CERTIFICATE OF CORRECTION

Patent No. 3,584,630 Dated June 15, 1971

Inventor(s) G. Esler Inskeep

It is certified that error appears in the above-identified patent and that said Letters Patent are hereby corrected as shown below:

In abstract, line 4, "ab-" should be "ad-"

In abstract, line 6, after "adsorbed", "nicotine in an amount of from 0.05 to one part by weight" was omitted.

Column 2, line 27 - remove "content of tobacco products could, by the addition of nicotine"

Column 3, line 15 - "if" should be -- is --.

Column 4, line 36 - "importantly" should be -- brought --.

line 70 - "300 μ" should be -- 300 mμ --.

line 72 - "260 μ" should be -- 260 mμ --.

Column 6, line 7 - "auger" should be -- a --.

Signed and sealed this 14th day of March 1972.

(SEAL)
Attest:

EDWARD M.FLETCHER,JR.
Attesting Officer

ROBERT GOTTSCHALK
Commissioner of Patents

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