United States Patent Office 3,424,173 Patented Jan. 28, 1969

3,424,173

FILTER ELEMENT FOR SELECTIVELY REMOVING NICOTINE FROM TOBACCO SMOKE

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No Drawing. Filed Sept. 14, 1967, Ser. No. 667,650
U.S. Cl. 131—267 4 Claims
Int. Cl. A24f 7/04; A24d 1/06

ABSTRACT OF THE DISCLOSURE

A filter element containing 1,2,4-butanetriol for selectively removing nicotine from tobacco smoke.

This invention relates to a tobacco smoke filtering material for selectively removing deleterious materials from tobacco smoke without at the same time removing those desirable smoke vapors which contribute aroma and taste to the smoke. More particularly, this invention concerns a novel cigarette filter tow and filters made therefrom, as well as the method for their manufacture, which will selectively remove nicotine from tobacco smoke.

The harmful physiological effects of certain constituents contained in tobacco smoke has long been recognized. It is well known, for example, that tobacco smoke contains certain solid tar constituents and health-affecting materials of the nicotine class which tend to physiologically irritate the smoker. In the past, various attempts have been made to obviate these harmful materials from tobacco smoke by either using various types of tobacco smoke filters attached to the smoking device, or incorporating certain preventive compounds into the tobacco being smoked. As of the present time, the use of a tobacco smoke filter element placed on the tip of the smoking device is the method or device most commonly used for removing these undesirable components from tobacco smoke. These filters, which normally consist of a bundle of cellulose acetate, convoluted crepe paper, cotton or combinations of these products formed into a cylindrical plug, are designed to and do remove varying proportions of the liquid-solid particles passing through them thereby greatly reducing the amount of undesirable materials reaching the smoker's mouth. This liquid-solid particle filtering action is accomplished by a combination of diffusional, impactive, and direct collision of the particles with the filter material. Upon collision the particles are retained in the filter by the surface attraction between the extremely small particles and the relatively large filter material. Thus, filters of this type are capable of removing varying percentages of tar and nicotine from cigarette smoke depending on the amounts of fibrous material compacted into them, their length, their resistance to draw, the surface characteristics of the fibers, and other factors.

However, these prior known fibrous filters show no selectivity for the removal of nicotine from the smoke of a cigarette, particularly when the tobacco involved is the conventional type used on domestic cigarettes. This type usually consists of a blend of bright, burley, and Turkish tobaccos with the bright ("flue cured") tobacco constituting the major portion of the blend. Filters of cotton, paper or cellulose acetate fibers, when attached to such domestic cigarettes, always remove the same percentage of nicotine from the smoke as they do tar. For example, if one of these filters removes 25 percent of the tar it also removes 25 percent of the nicotine; if is removes 40 percent of the tar it also removes 40 percent of the nicotine. Therefore, it can be said that these filters show no selectivity for removing nicotine over tar.

It has been suggested that one way to make a fibrous

filter of cellulose acetate, paper or cotton remove a higher percentage of nicotine than tar from cigarette smoke is to coat the fibers in the filter with acids and particularly with nontoxic, nonvolatile organic acids such as tannic acid, citric acid, glutaric acid and the like. However, such a technique leaves something to be desired from the standpoint of the taste of the filter. Also, in the case of filters of cellulose acetate fibers, the addition of an acid can cause hydrolytic degradation of the fibers on prolonged contact. As a result, acetic acid can be released from the filter giving it an objectionable odor and taste.

Therefore, it is readily apparent that an improvement in the tobacco smoke filter art which would permit the selective removal of nicotine from tobacco smoke without at the same time adversely affecting the flavor and aroma of the smoke would represent a substantial stride forward. Ideally this improvement should use a material and/or method that is inexpensive, reliable and commercially available. Furthermore, the filter should be chemically stable and be capable of selectively removing nicotine permanently so that it will not be eluted at a later date as the hot burning coal approaches the filter during the last few puffs on the cigarette. The process whereby the filter is produced must also be compatible with existing manufacturing equipment.

According to the present invention a convenient and effective method has been found by which a tobacco smoke filter can be constructed for the selective removal of nicotine from tobacco smoke. This method consists of coating or otherwise dispersing 1,2-4-butanetriol onto the filtering material from which the tobacco smoke filter element is formed.

Therefore, one object of this invention is to disclose nonacid containing tobacco smoke filters which remove a higher percentage of nicotine than tar from tobacco smoke.

Another object of this invention is to disclose a nonacidic additive for a tobacco smoke filter which imparts to the filter an improved capacity for removing nicotine from tobacco smoke.

These and other objects and advantages of this invention will be more apparent upon reference to the following description, appended claims and specific working examples.

As set out hereinabove, this invention involves the use of 1,2,4-butanetriol as an additive in tobacco smoke filters for selectively removing nicotine from effluent tobacco smoke. Since the 1,2,4-butanetriol acts through physical contact to adsorb the nicotine, it is very important that as much surface area of the 1,2,4-butanetriol be exposed for contact with the tobacco smoke vapors as possible. This desired surface exposure can be obtained, for example, by adding the 1,2,4-butanetriol to any general type of cigarette "filtering material" such as fibrous products of cotton, paper, regenerated cellulose, cellulose acetate, polyolefins or any other suitable fibrous carrier medium having large surface areas that can be coated. The 1,2,4-butanetriol can also be coated on granular particles such as flake acetate, carbon, rice starch, or any other similar material which has a large surface area and this coated granular material can then be placed in the filter element in a conventional manner. However, the preferred carrier media for the 1,2,4-butanetriol are paper in a creped form and cellulose acetate fibers in the form of a crimped tow.

The addition of the 1,2,4-butanetriol to the fibrous filtering material is accomplished by treating the fibrous filtering material with the liquid polyol either in its undiluted form or dissolved in a solvent. Obviously the undiluted form is preferred. This treating of the fibrous filtering material with the 1,2,4-butanetriol can be accomplished through a spraying, dipping, sponging, sprinkling

or other suitable method whereby the polyol is uniformly spread throughout the filter. As is apparent, this treatment of a fibrous filter material with the liquid 1,2,4-butanetriol or a solution thereof can best be performed while the material is in the form of a spread-out or "bloomed" form. However, the additive can be applied at other points in the processing of the material into a filter. In fact, the incorporation of the additive may take place after the final packaging of the tobacco smoke filter. To accomplish this, the 1,2,4-butanetriol is encapsulated in easily breakable plastic capsules and the capsules incorporaated into the filter. Upon mild pressure, these capsules within the filter could then be easily crushed releasing the 1,2,4-butanetriol onto the fiber surfaces.

The amount of the 1,2,4-butanetriol needed to selectively remove a significant amount of the nicotine from the effluent stream of tobacco smoke has been found to be rather critical with best results being obtained by adding enough 1,2,4-butanetriol to the carrier material such as paper, cotton, or cellulose acetate fibers, to give filters containing from 5–15 percent by weight of the 1,2,4-butanetriol. An amount less than 5 percent will not impart any significant increase in the filter's capacity to remove nicotine selectively over "tar" and an amount above 15 percent can cause the material to wet the conventional paper wrapper used to encase the filter material thus causing the wrapper to rupture. The use of excess 1,2,4-butanetriol could also prevent the paper wrapper from being sealed by the conventional adhesives used in the cigarette and filter making industry. However, probably the most damaging effect of too much 1,2,4-butanetriol is the creation of channels within the filter which cause the smoke to escape filtration. These channels, in the case of fibrous filters, are caused by a matting of adjacent fibers by the 1,2,4-butanetriol which is somewhat viscous. Extensive tests indicate that the preferred amount of 1,2,4-butanetriol additive is within the range of 8 and 12 percent.

As will be appreciated, the addition of 1,2,4-butanetriol to a filter does not preclude the possibility that other liquid additives can be added to it for other purposes. For example, in the case of filters made from a crimped tow of cellulose acetate fibers it is highly desirable that this material be treated with certain high boiling plasticizers prior to the final rod formation to impart rigidity to the filter rod. Thus, it is common to use such room temperature bonding agents as glyceryl triacetate or polyethylene glycol diacetates for this purpose. In such a case the 1,2,4-butanetriol may be applied to the opened tow during the manufacture of the filter rod either before or after the application of the bonding plasticizer. The addition of the plasticizer has no effect on the filter in regard to its efficiency for selectively removing nicotine from the tobacco smoke.

As mentioned briefly hereinabove, it was highly unexpected to find that, out of the large number of diols tested as additives to tobacco smoke filters, only 1,2,4-butanetriol was effective in rendering the filter capable of removing more nicotine than tar. Such compounds as ethylene glycol, diethylene glycol, polyethylene glycol, propylene glycol (1,2- or 1,3-), butylene glycols, and sorbitol, when applied to a filter material at concentrations below 15%, i.e., at concentrations which did not destroy the effectiveness of the filter for removing tar or which did not wet the paper wrapper around the filter, gave no selective removal of nicotine. Such treated filters removed the same percentages of nicotine and tar and were equal in this respect to the untreated filters.

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

EXAMPLE 1

Four sheets of weighed soft tissue paper known by the trade name "Kleenex," is spread out on a flat surface and exposed to a fine mist of 1,2,4-butanetriol created by a spray gun held about 3 feet above the paper. After this treatment the sheets are reweighed and numbered. The first sheet is treated so that it contains 5 percent 1,2,4-butanetriol while the second, third, and fourth sheets contained 7, 10, and 20 percent 1,2,4-butanetriol, respectively. Each of these sheets are folded to form a rod which is then wrapped with the conventional paper used for wrapping cigarette filter rods. The rod containing the paper with 20 percent additive cannot be used for conversion into filter tips because the additive will penetrate the paper and prevent it from being sealed with the water based glue commercially used for wrapping filter rod materials. However, the other rods are easily sealed into the paper wrapper with the glue. These rods are then cut into 20-mm. filter tip lengths (25-mm. circumference) and attached to 85-mm. standard cigarettes which had been shortened by 20 mm. to compensate for the length of the filters. A control filter rod of the tissue paper without the additive is also prepared in an identical fashion, cut into 20-mm. filter tip lengths, and these filters are attached to the shortened standard cigarettes.

All of the 85-mm. filter cigarettes are automatically smoked to 30-mm. butt lengths by means of a machine which drew a 35-ml. puff of 2 seconds duration at the rate of 1 puff/minute. The main stream smoke issuing from these cigarettes (5 cigarettes/run) is analyzed for its tar content by the photofluorometric procedure. Its nicotine content was analyzed by the silicotungstic acid procedure. The amounts of nicotine and tar on the filters are also determined using the same procedures. The results listed in Table I show that the control paper filter will remove the same percentage of tar as it does nicotine whereas those filters containing 1,2,4-butanetriol will remove a higher percentage of nicotine than tar even though all the filters have essentially the same pressure drop.

TABLE I.—FILTERS OF PAPER WITH AND WITHOUT 1,2,4-BUTANETRIOL

Type 20-mm. filter	Filter pressure drop (in.)	Percent tar removed by the filter	Percent nicotine removed by the filter
Paper, no additive	2.1	36	37
Paper with 5.0 percent 1,2,4-butanetriol	2.2	37	41
Paper with 7.0 percent 1,2,4-butanetriol	2.1	38	48
Paper with 10.1 percent 1,2,4-butanetriol	2.6	40	51

In all of these examples, the "tar" values are obtained by a photofluorometric procedure described by McConnell, Mumpower, and Touey in Tobacco Science, 4, 55–61 (1960). The nicotine values are obtained by the silicotungstic acid procedure described by Avens and Pearce in Ind. and Eng. Chem., 11 (No. 9), pp. 505–508 (1939). The filter pressure drops, expressed as inches of water at an air flow rate of 17.5 ml./sec. are obtained with the apparatus described by Touey in Analytical Chemistry, 27 (No. 11), pp. 1788–1790 (1955). The percentage of tar removed by the filters are obtained as follows: the total mg. of tar delivered by 5 filter cigarettes is measured. Then the total mg. of tar trapped by their filters is determined and the values are substituted in the formula

$$\text{Percent tar removed by filter} = \frac{F}{F+D} \times 100$$

F=mg. of tar found on the filters of the 5 filter cigarettes
D=mg. of tar delivered by the 5 filter cigarettes

The percentage of nicotine removed by the filters is obtained in a similar manner.

EXAMPLE 2

A 20-ft. length of crimped tow of three denier per filament cellulose acetate fibers (14,600 filaments) is spread out to a width of 12 in. and sprayed on both sides with glyceryl triacetate until it contains 7 percent of the hardening plasticizer. Three feet sections of this tow are then sprayed with 1,2,4-butanetriol in such a manner that an increasing amount of 1,2,4-butanetriol is applied to each section. One section, representing the control, is not treated with 1,2,4-butanetriol. Immediately after this treatment, the filter tow segments are compacted into a cord and paper wrapped into filter rods 90 mm. in length and 25 mm. in circumference (the circumference of a cigarette). After the filter rods are hardened (about 2 hours) due to the action of the bonding plasticizer they are cut into 20-mm. segments and attached to the standard (65 mm.) cigarettes used in Example 1. These filter cigarettes are then automatically smoked to 30-mm. butt lengths and the efficiencies of the filters for tar and nicotine removal are measured as described in Example 1. As in Example 1, each value given below (Table II) is based on the smoking of 5 filter cigarettes.

TABLE II

Type of 20-mm. filter	Filter pressure drop (in.)	Percent tar removed by filter	Percent nicotine removed by filter	Remarks
Acetate fibers without 1,2,4-butanetriol	2.1	40	41	Rigid filter rods were obtained after 2-hours' curing at room temperature.
Acetate fibers with 5.1 percent 1,2,4-butanetriol	2.1	39	43	Do.
Acetate fibers with 6.3 percent 1,2,4-butanetriol	2.0	40	50	Do.
Acetate fibers with 10.1 percent 1,2,4-butanetriol	2.2	39	51	Filter rods were firm but not rigid after 2-hours' curing at room temperature.
Acetate fibers with 20 percent 1,2,4-butanetriol [1]				Filter rods were very soft after 2-hours' curing at room temperature and could not be used in commercial production.

[1] Due to the excessive amount of 1,2,4-butanetriol on these filter rods, their paper wrapper became wet and the glue seal gradually ruptured.

EXAMPLE 3

A wad of cotton is divided into five sections each weighing 0.090–0.10 gram. One of the sections (control) is not treated. The others are treated with 1,2,4-butanetriol by immersing the wads in 1,2,4-butanetriol dissolved in methanol. Solutions having increased concentrations of the 1,2,4-butanetriol are used to give wads having different percentages of the polyol additive. All of the cotton wad sections are heated to 70° C. for 1 hour to evaporate off the methanol and then they are stored over calcium chloride for 48 hours. Finally each wad is inserted into a cardboard tube of 20-mm. length and having a circumference of 25 mm. These packed tubes are attached to the standard cigarettes of Example 1. The cigarettes are automatically smoked to butt lengths of 30 mm. and the percentage amounts of nicotine and tars removed by the filters are calculated as described in Example 1. The results of these tests are shown in Table III.

TABLE III.—COTTON FILTERS CONTAINING 1,2,4-BUTANETRIOL

Percent 1,2,4-butanetriol on filter	Filter pressure drop (in. H_2O)	Percent tar removal	Percent nicotine removed
0.0	2.0	40	39
4.0	2.0	39	41
5.1	2.1	40	45
7.4	2.3	41	53
10.0	2.4	43	53
17.0	3.8	35	47

It can be seen from these results that when the filter contains at least 5 percent 1,2,4-butanetriol it removes more nicotine than tar. However, the results also illustrate that adding over about 10 percent of the polyol to the cotton impairs its tar removing capacity giving a removal value of 33 percent rather than 40 percent. It can also be seen that the addition of over 10 percent 1,2,4-butanetriol increases the pressure drop (resistance to draw) of the filter excessively yet does not give a corresponding increase in tar removal. For these reasons, therefor, the amounts of 1,2,4-butanetriol on the filter should be kept within a certain critical limit; namely, a minimum of 5 percent and a maximum of 15 percent with the ideal or preferred range being between 8 and 12 percent.

Although the mechanism for this selective removal of nicotine is not clearly understood, the following postulation is offered: the presence of 1,2,4-butanetriol on the surface of the fibers in the filter renders it a better solvent for that portion of the nicotine which is present in the vapor phase of the smoke aerosol as it enters the filter. As a result the nicotine vapor is absorbed or dissolved while the nicotine in the particulate phase (commonly called tar) of the smoke is trapped by the filter by physical impingement. In the case of the filter without the additive the only nicotine capable of being removed is that which comes over in the particulate phase of the smoke (tar). Hence, the filter removes the same percentages of nicotine and tar because in this case the nicotine is a component of the tar particle. By such an explanation it is obvious that most of the nicotine in cigarette smoke is in the particulate phase (aerosol particles). However, there is still a significant amount in the vapor phase and it is this amount that responds to selective filtration by the 1,2,4-butanetriol.

As will be appreciated it was highly unexpected to find that 1,2,4-butanetriol exhibits this property of imparting to a fibrous filter the capacity of removing nicotine selectively from cigarette smoke in view of the fact that other aliphatic polyols, including other butanetriols, were ineffective. For example, the following example illustrates the ineffectiveness of other polyols for significantly improving a filter's capacity to remove nicotine selectively.

EXAMPLE 4

A 20-ft. length of crimped tow of 3 denier per filament cellulose acetate fibers (14,600 filaments) is continuously opened and sprayed with triacetin, the bonding agent commonly used in preparing rigid filters of this material. The opened tow is then sprayed with one of the polyols shown in Table IV. This is accomplished by changing the polyols in the spray booth at different intervals during the continuous preparation of paper wrapped cellulose acetate filter rods by a technique similar to that described in United States Patent 3,017,309. In each case the amounts of hardening agent (triacetin) and polyols added separately to the opened moving tow is between 10 and 11 percent, respectively (based on the total weight of the combination without the paper wrapper). The treated tow is then recompacted into a tight bundle which is then drawn through a device that wraps it in thin cigarette paper and cuts it into 90-mm. rods with a circumference (25.3 mm.) equal to that of a cigarette. After about a 2-hour storage the rods will become sufficiently firm so that they can be cut into 20-mm. filter lengths. These filters are tested for their capacity for removing nicotine and tar from cigarette smoke in the same manner as described in Example 1. The results are listed in Table IV.

TABLE IV

Type of glycol (10–11 percent) on the 20-mm. filter	Filter pressure drop (in. H_2O)	Percent tar removed	Percent nicotine removed
None (control)	2.2	41	42
Ethylene glycol	2.1	39	38
Diethylene glycol	2.2	40	39
Triethylene glycol	2.0	40	40
Tetraethylene glycol	2.0	39	40
1,2-propanediol	2.1	40	39
1,3-propanediol	2.0	39	41
1,2-butanediol	2.2	38	39
1,3-butanediol	2.1	40	39
1,6-hexanediol	2.1	39	41

From the foregoing description it is readily apparent that the tobacco smoke filters of this invention offers numerous advantages over those previously known and used in the cigarette industry. For example, the filters are selective i.e. they are capable of removing a higher percentage of nicotine than "tar" from tobacco smoke as it moves through the interstices of the filter. The nicotine removed by the 1,2,4-butanetriol is formed into a harmless complex which cannot later be eluted from the filter. Furthermore, the fact that the 1,2,4-butanetriol additive is a liquid means that it can be coated over the filter tow and this substantially eliminates the masking, sifting-out and house-cleaning problems heretofore so commonly encountered where finely-divided solid additives were used. The method and ease with which the 1,2,4-butanetriol additive can be evenly dispersed throughout a tobacco smoke filter also alleviates to a great extent the need for special equipment, processing steps and skilled personnel that has heretofore been required to produce a filter tow having a solid additive therein. Thus the cost of producing the tow, as well as the filter units themselves, by the method and with the materials of this invention is substantially reduced.

We claim:

1. A tobacco smoke filter for selectively removing nicotine from the effluent stream of tobacco smoke comprising a carrier medium arranged to provide interstices for the passage of smoke therethrough and having dispersed therein an additive of 1,2,4-butanetriol in an amount between about 5 and 15 percent by weight of said filter.

2. A tobacco smoke filter according to claim **1** wherein the 1,2,4-butanetriol is coated on the surface of the carrier medium.

3. A tobacco smoke filter according to claim **2** wherein the carrier medium is a fibrous material selected from the group consisting of cotton, paper, regenerated cellulose, cellulose acetate, polyesters and polyolefins.

4. A tobacco smoke filter according to claim **1** wherein between about 8 and 12 percent by weight of the 1,2,4-butanetriol is dispersed within the filter.

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