United States Patent [19]

Reynolds et al.

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[54] **TOBACCO SMOKE FILTER**

[75] Inventors: **Martin Lance Reynolds**, Anchorage, Ky., **James R. Hammersmith**, Jeffersonville, Ind.

[73] Assignee: **Brown & Williamson Tobacco Corporation**, Louisville, Ky.

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[21] Appl. No.: **202,148**

Related U.S. Application Data

[63] Continuation-in-part of Ser. No. 33,621, May 1, 1970, abandoned. Continuation-in-part of Ser. No. 849,384, Aug. 12, 1969, abandoned.

[52] **U.S. Cl.** **131/267**, 131/268

[51] **Int. Cl.** **A24d 01/04**

[58] **Field of Search** 131/261–269, 131/10

[56] **References Cited**

UNITED STATES PATENTS

3,329,544	7/1967	Smith et al.	131/269 X
3,528,433	9/1970	Johnson et al.	131/10.5
3,574,803	4/1971	Allen et al.	131/267
3,358,695	12/1967	Blakey et al.	131/269
3,393,120	7/1968	Tovey et al.	131/267 UX
2,542,527	2/1951	Honey et al.	131/269 UX

FOREIGN PATENTS OR APPLICATIONS

3,445,578	5/1960	Switzerland	131/268

Primary Examiner—Melvin D. Rein

Attorney, Agent, or Firm—Finnegan, Henderson, Farabow & Garrett

[57] **ABSTRACT**

An improved tobacco smoke filter is provided for cigarettes and the like formed of a bed or rod of shreds of a porous cellulose ester material having a low packing density and an increased surface area which is readily accessible to the smoke aerosol for deposition of smoke particles. The filter draw resistance comprises a pressure drop at or below 2.5 inches of water. The cellulose ester material in shred form has a surface area of from about 0.6 to 3.0 square meters per gram, a mean pore diameter of from about 2 to 20 microns, and a porosity of between 65 percent and 90 percent. The width and thickness of the shreds should be between about 0.25–2.0 mm and 0.025–0.25 mm, respectively. The shreds are formed by casting a dope of the cellulose ester dissolved in solvent and plasticizer in which starch and saline solution are present. The cast sheet is washed to remove the salt and enzymatically treated to remove the starch and thus form the porous structure.

4 Claims, 3 Drawing Figures

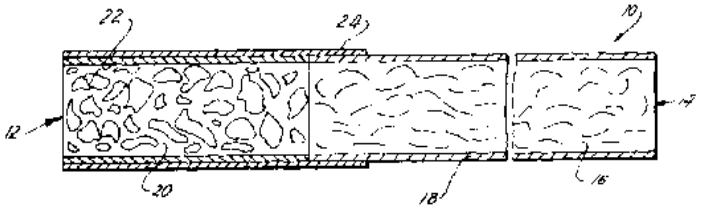
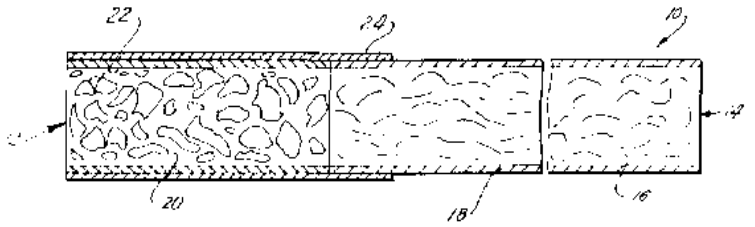


FIG. 1



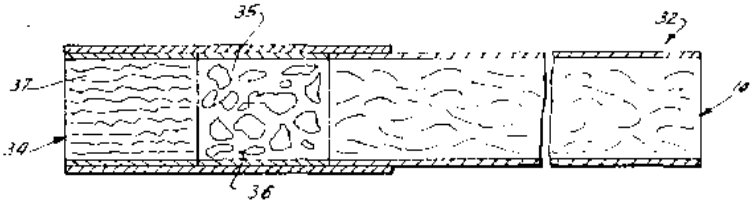


FIG. 2

FIG. 3

TOBACCO SMOKE FILTER

CROSS REFERENCE TO RELATED APPLICATION

This application is a Continuation-in-Part of our United States patent application Ser. No. 33,621 filed May 1, 1970, now abandoned which was a Continuation-in-Part of our United States patent application Ser. No. 849,384 filed Aug. 12, 1969, entitled "Tobacco Smoke Filter," now abandoned

BACKGROUND OF THE INVENTION

The reduction in tar delivery which can be achieved with conventional cigarette filters is limited by the necessity of maintaining an acceptable cigarette pressure drop while, at the same time, maintaining the quality of the smoke. A continuing search has been made both within, and outside, the tobacco industry for more efficient cigarette filters.

Heretofore attempts to incorporate porous materials into cigarette filters to increase the filter's efficiency have met with only limited success in that they have not proven highly efficient in tar removal at acceptable pressure drops. Such prior art filter materials include, for example, activated charcoal in granular form, as a self-sustaining bonded rod or incorporated on fibers or web material, activated alumina, silica gel and various types of synthetic resins in granular form.

SUMMARY OF THE INVENTION

It is the principal object of the present invention to provide an improved cigarette filter cellulose ester material capable of filtering a smoke aerosol at acceptable pressure drops of at or below 2.5 inches of water with a high degree of efficiency so as to remove at least 60% of the tar from the aerosol without adversely affecting the tobacco flavor.

Another object is to provide such a cellulose ester material which can be produced at a sufficiently low cost so as to enable the same to be competitive with existing filter materials.

These and other beneficial objects and advantages are attained in accordance with the present invention by providing a filter constituent formed of porous cellulose ester shreds have a surface area between 0.6 and 3.0 square meters per gram, a porosity of between 65 percent and 90 percent and a mean pore diameter of from about 2 to 20 microns. The width and thickness of the shreds should preferably be between about 0.25–2.0 mm and 0.025–0.25 mm, respectively. The individual filter shreds are packed in abutting proximity with one another to form a bed or a rod so as to provide a tortuous path for the smoke aerosol.

BRIEF DESCRIPTION OF THE DRAWINGS

In the accompanying drawing.

FIG. 1 is a cross-sectional view of a cigarette provided with a filter incorporating the porous material of the present invention.

FIG. 2, which is a view similar to FIG. 1, depicts a cigarette filter employing the porous material of the present invention with a conventional mouthpiece.

FIG. 3 is a scanning electron microscope photograph at 500X magnification showing the porous nature of the material.

DESCRIPTION OF THE PREFERRED EMBODIMENTS

In FIG. 1 a cigarette 10 is provided with a filter 12 utilizing the porous cellulose ester shreds of the present invention. The cigarette includes a tobacco rod 14 comprising tobacco material 16 contained within a paper tube 18 to which the filter 12 is secured. The filter 12 contains the porous cellulose ester shreds 20 contained within a wrap 22. An outer wrap 24, which may be formed of paper or cork, overlies the entire filter and extends somewhat beyond the tabacco rod-filter interface. If desired, the shreds may be bonded to one another at random points with a conventional plasticizer such as, for example, triethylene glycol

The porous cellulose ester shreds 20 are formed having physical characteristics defined by a surface area of between about 0.6 square meters per gram and about 3.0 square meters per gram, a porosity of between 65 percent and 90 percent, and a mean pore diameter of from about 2 to 20 microns. It is preferred, however, that the surface area range from about 1 to 3 sq. meters per gram. The porosity of the cellulose ester material is the percentage of the total volume occupied by voids contained within each particle related to the total volume of the particle. The approximate relationship between pore volume, pore diameter, and surface area is defined by the following equation (assuming cylindrical pores):

$$\text{Volume of Pores (in cc/gm)}/\text{Surface area (cm}^2\text{/gm)} = 0.25 \text{ Pore Diameter (in cm)}$$

The lengths of the individual shreds should be at lenst 0.5 mm and should vary from about 0.5 mm to 20 mm; the width of the individual shreds should vary from about 0.25 mm to 2 mm; and the thickness of the individual shreds should vary from about 0.025 to 0.25 mm. Good results were obtained using porous shreds of cellulose acetate having a mean thickness of 0.03 mm and a mean width of 1.0 mm. The cross-sectional configuration of the individual shreds is not critical, although ribbon-shaped shreds are preferred.

The filter efficiency and draw resistance of the filter bed is also, of course, related to the bulk density of shreds within the bed and in this connection the shred bulk density should lie between 0.05 and 0.40 grams per cc and preferably between 0.09 and 0.30 grams per cc to provide pressure drops across the filter section of approximately 2 to 2.5 inches of water. The shreds are packed into abutting proximity with one another and, thus, unlike granular filter material, have a high degree of self-cohesiveness.

Various cellulose esters may also be employed, as, for example, porous cellulose propionate, porous cellulose butyrate or the like. Porous cellulose acetate is the preferred cellulose ester because consumers have become accustomed to the taste of smoke drawn through a cellulose acetate filter.

FIG. 2 depicts a cigarette 32 provided with a filter 34 formed of a shredded porous cellulose ester 36 in accordance with an alternate embodiment of the present invention. The filter 34 comprises two sections, tobacco section 35 formed of shredded porous cellulose ester 36 abutting the tobacco rod 14 and mouthpiece section 37. The mouthpiece section may be made from conventional fibrous material such as cellulose acetate tow. The porous material 36 of the tobacco section 35

of the filter possesses the same physical characteristics set forth previously.

FIG. 3 illustrates pictorially the porous nature of the shredded material which here is cellulose acetate. The photograph is at 500X magnification.

Porous materials in accordance with the present invention may be manufactured from cellulose esters by encasing a starch with the ester, swelling the starch and then removing the starch by enzymatic and/or chemical hydrolysis or by solubilizing the starch by cooking in water and draining off the solution. This process provides porous cellulose esters having the desired surface area and mean pore diameter. The porous cellulose esters are made as shreds of the desired width and thickness and, thus, may be formed into filter elements by employing standard cigarette making machinery.

For a better description and understanding of the method for making synthetic porous cellulose esters having the characteristics of the invention herein, the following examples are provided. In each example, the starch utilized was one which, upon swelling, produced a pore of from 2 to 20 microns. Since the swelling rates of the starches utilized was of the order of 100 percent, the initial starch particle size was from 1 to 10 microns. Also, in each of the following examples, the enzyme used was an alpha-amylase of bacterial origin commercially available under the trade name TENASE from Miles Laboratories. The "parts" of the examples are parts by weight

EXAMPLE I

One hundred parts of cellulose acetate were dissolved in a solution of 800 parts acetone and 20 parts plasticizer (6 parts triethylene glycol diacetate, 3 parts polyethylene having a molecular weight of 600), this amount being 20 percent of the cellulose acetate weight. The resulting dope was stirred for 30–40 minutes until all the cellulose acetate had been dissolved. A warm salt solution of about 80° C. comprising 30 parts sodium chloride and 90 parts water was slowly mixed into the dope before blending in 300 parts corn starch. The resulting mixture was stirred for about one hour, after which the mixture was rolled into a sheet at thickness 80–85 percent of that desired for the final product.

The dried sheet was cut into shreds about 29 cuts per inch, using a guillotine cutter similar to the type employed for making tobacco shreds from the tobacco leaf. The irregular, flaked and powdered material was separated from the shredded material by screening. The shredded material, 1 part by weight, was suspended in 4 parts of 0.02 molar calcium chloride, and the resulting slurry adjusted to pH 7 with sodium hydroxide to approximately 0.02M. The slurry was heated in a pressurized vessel at about 120° C. and 15 psi for about 30 minutes to expand the starch and to rupture the pore walls. The slurry was then cooled to 88°–95° C. Since the slurry was previously adjusted for enzymatic conditions, 0.003 parts of a concentrated alpha-amylose (TENASE - Miles Laboratories) solution was added for solubilization of the starch. To effect solubilization of the starch, the temperature was maintained at between 88° and 92° C. with occasional agitation. Solubilization of the starch was substantially completed after 50 to 60 minutes. The solubles were drained from the shredded material which was then thoroughly washed with hot tap water, then spread out to dry under ambient conditions. The dry shredded porous cellulose acetate was air fluffed and screened to break up roped material and long strands. The mean shred size obtained after selectively sieving the dry material was found to be 0.88 mm in width and 0.0305 mm in thickness. The surface area was found to be about 1.2 M^2/gm with a porosity of about 80 percent.

Sufficient shreds were packed into a filter tube 25 mm long and 24.8 mm in circumference to provide a bulk density of about 0.122 gms/cm^3 and a pressure drop of 2.3 inches. The filter removed 71 percent of the tar when attached to a conventional cigarette.

EXAMPLE II

The relationship between pore volume, surface area and pore size has been previously discussed. Thus, with the mean pore diameter ranging between 8 to 12 microns the effect of the total volume and the surface area may be examined where starch to acetate ratio in the preparation of the porous cellulose acetate shreds are fixed in 1:1, 2:1, 3:1, and 4:1 ratios as set forth in Table I below:

TABLE I

Starch/Acetate Ratio Used in Preparation	Porosity %	Surface Area M^2/g	Average Filtration Efficiency %
1/1	53	0.3	30
2/1	66	0.6	50
3/1	82	1.2	68
4/1	90	2.7	70

The filters employing the materials indicated in the table above were all 25 mm in length and packed with sufficient porous shreds to provide a pressure drop of 2.4 inches.

From the examples presented herein with regard to the porous cellulose acetate shreds, other cellulose ester materials such as cellulose propionate or cellulose butyrate may be employed in place of the porous cellulose acetate ester of the examples herein.

To compare the teachings of the prior art and the advances made by the applicants herein, applicants' porous cellulose ester made in accordance with this invention is compared with the teachings of the prior art as set forth in U.S. Pat. No. 3,364,938 granted to Mumpower II, et al, Jan. 23, 1968. This patent discloses microporous polyolefin floc type of powder which is applied to the surface of crimped cellulose acetate filaments which is then formed into a filter element for cigarettes.

Applicants examined the results set forth in Examples 1–4 of U.S. Pat. No. 3,364,938. All of the filters described therein were 17 mm in length. Applicants then prepared filtered cigarettes employing porous cellulose acetate shreds having a filter element 25 mm in length.

The comparative characteristics of the filter material, the filter pressure drop and efficiency are set forth in the Tables II and III.

TABLE II (U.S. Pat. No. 3,364,938)

Ex.	Material[a]	Floc Size[b]	Filter P.D.[c]	Efficiency[c]
1	21PF 5/10,000	20–50	2.7	53
2	17PE 5/12,000	40–70	3.0	55
3	15PE 5/9,000	80–120	2.7	50
4	25PP 5/10,000	20–50	2.6	55

Notes

a. 21PE 5/10,000 describes filter containing 21% polyethylene floc dusted onto 5 denier/filament cellulose acetate tow containing 10,000

filaments PP is polypropylene
b Floc size refers to standard sieve series mesh numbers
c Efficiency % of the filter for removing tar

TABLE III (Porous Cellulose Acetate Shreds)

Ex.	Shred Size*(mm)	Filter P [2 "	Average Efficiency
1	1.6×0.08	2.4	70
2	0.6×0.12	2.5	65
3	0.6×0.035	2.3	71
4	0.9×0.03	2.3	71

a. Shreds were 1.5 to 2.5 centimeters in length

As can clearly be seen from reviewing the data in Tables II and III, the filtration efficiency of the material of the present invention is about 40 percent higher than that of the material of the prior art. Surprisingly, the significant increase in efficiency is obtained with a pressure drop which is lower than that of the prior art. Thus, the efficiency increase is obtained with no loss in drawability.

In order to properly compare the intrinsic performance of the two different filter types, it is necessary to calculate the filtration coefficients which allow for differences in length and pressure drop. By using the equation

$$\log (1 - E/100) = -\mu PL$$

Where E = Filter efficiency percent

P = Filter pressure drop, inches w.g.

= Filter length, cm, the coefficient μ which describes the inherent filtering capability of the material is obtained.

From the formula above the filtration coefficients were calculated for Examples 1 through 4 inclusive for U.S. Pat. No. 3,364,938 and applicants' Examples 1 through 4 inclusive.

TABLE IV

(Comparison of filtration coefficients, μ)

Example	U.S. Pat. No. 3,364,938	Porous Cellulose Acetate Shreds
1	0.072	0.087
2	0.064	0.073
3	0.066	0.094
4	0.079	0.094
Average	0.071	0.085

Thus, for a filter 21 mm in length, which is the average of the filter in U.S. Pat. No. 3,364,938 and the one employed in this disclosure, the filtration efficiency would be significantly greater using material of the present invention

Not only do filters made in accordance with the present invention provide an increase in efficiency, but the desirable taste qualities are retained. Additionally, this increase in efficiency is obtained with no increase in pressure drop. In fact, the pressure drop is described while obtaining this increase in efficiency. This is surprising in view of the prior art attempts to increase efficiency which have generally resulted in an increase in the pressure drop

The shredded shape and bulk density characteristics also afford a high degree of self-cohesiveness. Therefore, it is apparent that variations and changes will be obvious to one skilled in the art after a reading of the present disclosure. Such variations and changes are meant to be included within the scope of the invention as delineated by the following appended claims.

Having thus described the invention, what is claimed is:

1. A tobacco smoke filter comprising:

a section formed of dried shreds of porous cellulose ester selected from the group consisting of cellulose acetate, cellulose propionate, and cellulose butyrate,

said shreds having a width of about 0.25 to 2.0 mm. and a thickness of about 0.025 and 0.25 mm., a porosity greater than 65 percent and up to about 90 percent, a filtration efficiency of at least 65 percent up to about 71 percent, and

a surface area of between 0.6 and 3.0 square meters per gram,

said shreds being made by forming a dope of cellulose ester dissolved in solvent and plasticizer and in which there is starch and saline solution whereby a sheet cast from said dope is shredded, washed to remove the salt, and enzymatically treated to remove said starch to form porous shreds, a ratio of said starch to said cellulose ester being above 2:1 and up to 4:1.

2. The invention in accordance with claim 1 wherein the bulk density of said materials is from about 0.05 to 0.40 grams per cubic centimeter.

3. The invention of claim 1 wherein said constituent shreds are characterized by a mean width of about 1.0 mm and a mean thickness of 0.03 mm.

4. The invention in accordance with claim 1 wherein said porous constituent shreds are bonded to one another at random points.

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