March 13, 1962 E. C. WHITE 3,025,130

WET SPINNING OF LOW DENSITY CELLULOSE ACETATE FILAMENTS

Filed Feb. 3, 1959

2 Sheets-Sheet 1

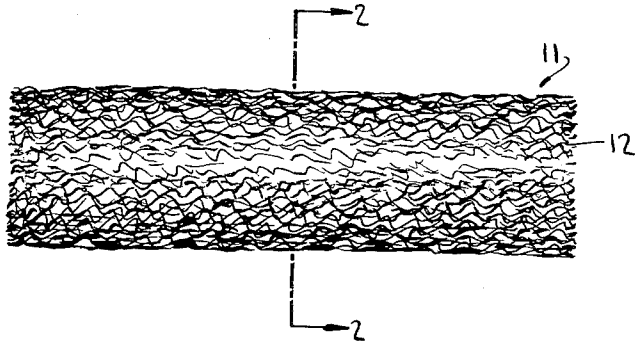


Fig. 1.

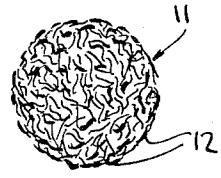


Fig. 2.

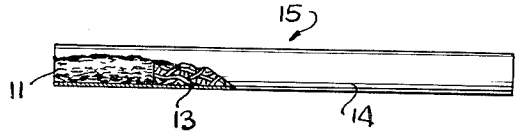


Fig. 3.

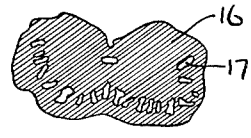


Fig. 4.

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WET SPINNING OF LOW DENSITY CELLULOSE ACETATE FILAMENTS

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2 Sheets-Sheet 2

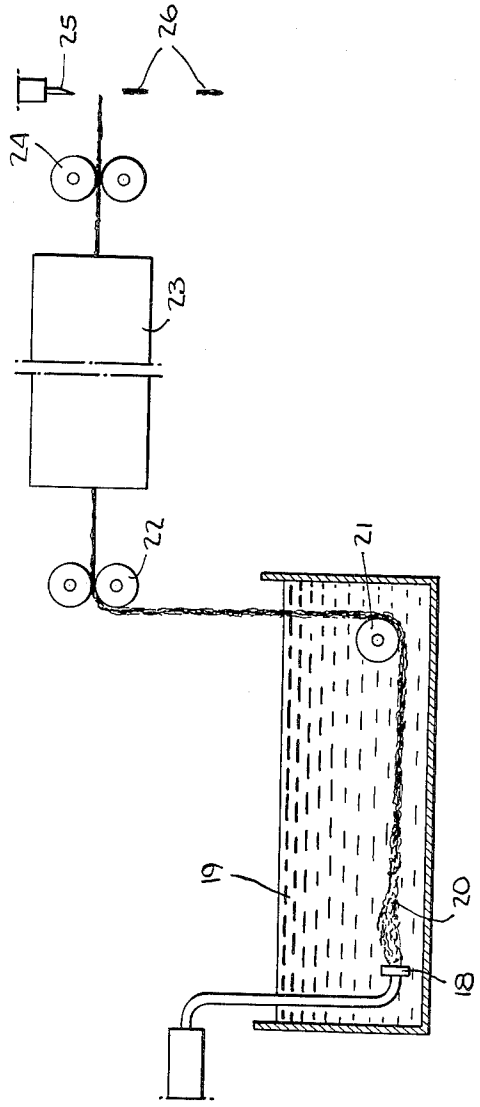


Fig. 1

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WET SPINNING OF LOW DENSITY CELLULOSE ACETATE FILAMENTS

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8 Claims. (Cl. 18—54)

This invention relates to the production of improved fibrous materials of low density which are especially suited for use as cigarette smoke filters.

In an earlier filed application, Serial No. 537,998, filed October 3, 1955, and now abandoned there is disclosed a process in which a solution of cellulose acetate in acetone is extruded through a spinning jet provided with many orifices into water as a coagulant to form filaments. By suitably controlling the orifice arrangement, the orifice dimensions, and the relative rates of extrusion and take-up, fibrous products of low density are produced, either as continuous fibers or as masses of entangled filaments.

While the fibrous products so produced are suitable for many purposes, it has been found that there is a practical limit to the ratio of filtration efficiency to filter weight which can be achieved when using such materials as filters on cigarettes. In an attempt to increase this ratio by increasing the available surface area and thus the filtration efficiency, more spinning orifices of smaller size have been provided to produce finer denier filaments. Very small openings, however, are easily blocked so spinning is not stable. Furthermore, the increasing cost of producing extremely small accurately dimensioned orifices also acts as a deterrent to decrease in orifice size.

The denier of the filaments can be decreased by decreasing the concentration of filament-forming material. This is disadvantageous in that it requires additional solvent and larger equipment to handle a given amount of filament-forming material. The solvent recovery system would also have to be enlarged and special measures would have to be taken to prevent excessive increase in the concentration of solvent in the coagulating bath.

In addition, as spinning speed was increased it resulted in flattening of the filament bundle, spinning instability, discontinuities, excessive coalescence and excessive pressure build-up behind the extrusion spinnerette.

One object of the invention is to increase the production rate in processes of the type described.

A further object is to improve the uniformity of the filamentary product resulting from the wet spinning process.

An additional object is to provide a combination of dope composition and coagulating medium composition which produces a filamentary product with enhanced filtration efficiency per unit weight of the filamentary product.

Still another object is to produce a novel filter of low density and enhanced filtration efficiency.

These and other objects will become more apparent from the following description in which a preferred embodiment of the invention has been set forth by way of illustration. All parts are by weight unless otherwise specified.

In accordance with one aspect of the present invention cellulose acetate is dissolved in a solvent comprising acetone and in excess of about 20% by weight of water and this highly aqueous solution is extruded into an aqueous coagulant to produce filaments. The coagulant may also include alcohol or other non-solvents for the cellulose acetate or even solvents for the cellulose acetate present in insufficient amount to prevent coagulation; it may also include surface active agents, etc. The resulting filaments are drawn through and out of the coagulating liquid at a linear speed ("take-up speed") less than half the linear speed at which the solution is extruded ("extrusion speed"); preferably the ratio of the take-up speed to the extrusion speed is below 1:3, and especially between 1:5 and 1:12 or 1:15.

After leaving the coagulating bath the fibrous materials may at once be wound up or otherwise collected, or they may be subjected to a treatment with a solvent or swelling agent for the cellulose acetate or other filament-forming substance, or with a suitable adhesive, in order to bond the filaments together at some or all of their crossing places. For example the material may be drawn from the coagulating bath by means of a roller which is heated or on which the material is treated with a solvent or swelling agent or an adhesive. Thus when a cellulose acetate material is being made the roller may be kept wet with acetone or the material as it passes over it may be subjected to a fine spray of acetone. A certain amount of bonding may occur in the coagulating bath.

The fibrous materials as initially formed may if desired be modified by compressing them, e.g. by passing them between suitably spaced rollers of which at least one is positively driven. The rollers may be at the ambient temperature, or they may be heated so as to promote mutual adhesion or interfusion between the individual filaments; alternatively or in addition acetone or another solvent or swelling agent may be applied to the rollers or to the material before it passes between the rollers. The rollers may be so positioned that the material passes between them immediately after leaving the coagulating bath. They have the effect not only of increasing the density, tightness of packing, and (under the appropriate conditions) the degree of bonding of the materials, but also of expelling from them water taken up from the coagulating bath.

The products are made up of a large number of continuous filaments which are highly curled or crimped and are so entangled with each other as to form a unitary structure having large air spaces and in consequence a low density. The fibrous materials may have many forms; for example they may be more or less narrow flat sheets or ribbon-like, rod-like or tube-like structures, or they may resemble a ribbon or sheet which is both corrugated and folded one or more times longitudinally.

With respect to the process variables, the solvent for the filament-forming material comprises in excess of about 20% by weight of water and preferably about 24 to 35%. The maximum water content is that which will permit the filament-forming material to remain dissolved at the prevailing temperature; thus, it will vary inversely with the concentration of the filament-forming material and directly with the temperature.

The filament-forming material comprises acetone-soluble cellulose acetate and is dissolved to constitute about 15 to 30% and preferably about 20 to 24% of the weight of the solution.

The temperature of the solution when extruded may range from about 20 to 80° C. and room temperature is quite satisfactory. The coagulant is conveniently at about room temperature although higher or lower temperatures are permissible. While slow take-up speeds are permissible, it is an advantage of the present invention that satisfactory products are obtained even when the take-up speed exceeds 3 meters per minute. It may be as high as 5 meters per minute or even more.

The spinning jet employed to form the filaments preferably contains at least 100, and for most purposes at least 200, orifices; for example it may contain 400–2500 or more orifices. These orifices should be closely spaced;

thus they may be less than 2 millimeters and preferably less than 1.5 millimeters (between centers) apart. The diameter of the orifices and their arrangement will depend on the precise nature of the product desired. Diameters between about 0.025 and 0.125 millimeter are generally preferred.

The arrangement of the orifices determines the form of the fibrous material produced. Thus if a sheet- or ribbon-shaped product is desired, the orifices may be arranged in a straight row, or better in two or more parallel straight rows. If a rod-like structure is desired, the orifices may be distributed over substantially the whole of a circular area, while if the product is to have a tube-like structure, the center of the circle may be left undrilled and the orifices distributed over the remaining annulus. By extruding the solution through orifices arranged in one or more closely spaced concentric circles around the outside of the circular jet face, leaving a comparatively large undrilled central area, a fairly flat, longitudinally corrugated and folded structure is formed, which may be described as a collapsed tube. Other structures may be obtained by using other arrangements of the orifices. While the orifices will usually be of circular cross-section for the sake of convenience and ease of formation, orifices of non-circular cross-section, e.g. of an elliptical, rectangular or slot-like cross-section, or of a cross-section having re-entrant angles, may be used.

The orifices in the spinning jet need not all be identical. In particular, materials of increased longitudinal strength can be obtained if the jet contains some orifices, for example from 10–40% of the total number, designed to produce more or less straight filaments running through the tangled mass of curled or crimped filaments. For this purpose it is necessary that the spinning solution should flow more slowly throug the "straight filament" orifices than through the remainder, so that in their case the ratio of the take-up speed to the extrusion speed is above about 0.8:1, and preferably in the neighborhood of 1:1 or higher. This may often be achieved simply by making these orifices with a bigger cross-sectional area, although in some instances it may be necessary or advisable also to increase the length of the channels leading to them or to adopt some other means of suitably restricting the amount of spinning solution passing through them. Thus depressions in the inner face of the jet from which the orifice channels lead may be shallower for the "straight filament" orifices than for the remainder. Alternatively the "straight filament" orifices may be fed with spinning solution from a different source, e.g. under a lower pressure, or preformed straight filaments may be led through the coagulating bath in such a way that the tangled mass of curled or crimped filaments is formed round them, but these methods are more complicated.

For forming cigarette filters, advantageously there is employed a spinning jet having about 600 to 3000 orifices and preferably about 1200 to 2000 orifices to produce an equal number of filaments. Each filament advantageously is about 4 to 20 and preferably about 8 to 12 denier. Preferably the total denier of the filter is about 60,000 to 80,000 in the curled or crimped state, each filament having about 40 to 80 crimps per inch of tensioned length. The filters are more bulky, more resilient, have a softer hand and are more effective than those produced under conditions identical except for a much lower water content in the extrusion dope. When incorporated into cigarettes they are cut into plugs about 15 millimeters long and 25 millimeters in circumference. Their density ranges from about 0.2 to 0.4 gram per cubic centimeter. Their filtration efficiencies equal those of similar filters of greater weight produced by extruding less aqueous dopes. The lower density of the new filters, i.e. the lower weight for a filter of a given size, is due partially to better disposition of the component filaments relative to one another and partially to the lower density of the individual filaments, i.e. the novel filaments are characterized by voids or holes which become visible in inspecting the filament cross-section.

In addition, the present invention permits faster take-up than when the water content is 5% or less. Thus, the present invention permits satisfactory filters to be produced at take-up speeds in excess of 1.7 meters per minute and even in excess of 3.0 meters per minute.

The novel products in accordance with the invention are illustrated in the accompanying drawing wherein:

FIG. 1 is an elevational view on an enlarged scale of a cigarette filter produced in accordance with the invention;

FIG. 2 is a cross-section through the filter of FIG. 1 taken along line 2—2;

FIG. 3 is an elevational view of a cigarette, with the paper partly torn away, having one of the novel filters incorporated therein;

FIG. 4 is a cross-section, on a highly magnified scale, through a filament produced in accordance with the invention; and

FIG. 5 is a schematic illustration of the process for producing the product of FIG. 1.

Referring now more particularly to the drawing, there is shown in FIG. 1 a plug **11** comprising a short length of cohered substantially continuous crimped cellulose acetate filaments **12**. In FIG. 2 it can be seen that the plug **11** has an approximately circular cross-section. FIG. 3 shows a plug **11** wrapped together with tobacco **13** in paper **14** to form a filter tip cigarette **15**. FIG. 4 shows a single filament **16** in cross-section, the filament having many voids or holes **17** which contribute to decreased weight, i.e. a filament with voids has the same surface area available for filtration as a filament of identical external contour which is solid and thus heavier.

The invention is further illustrated by the following example.

Example

20 parts of cellulose acetate were dissolved in 80 parts of 72/28 acetone/water. The resulting solution at 45° C., was extruded as shown in FIG. 5 through a spinning jet **18** provided with 1659 circular orifices each 40 microns in diameter. The linear speed of extrusion was 27.9 meters per minute. The coagulant **19** was water supplied at 20° C. After passing 0.6 meter through the coagulant, the bundle of filaments **20** was withdrawn by a take-up roll **21** at a speed of 3 meters per minute, condensed by rolls **22** to about 1.5 centimeters in diameter, dried in a forced circulation hot air dryer **23**, re-condensed by shaping rolls **24** to approximately final size of 25 millimeters in periphery, cut by blade **25** into 90 millimeter lengths **26** and surface-plasticized by rolling under pressure over a hot metal surface. The plugs, which had a density of about 0.3 gram per cubic centimeter, weighed about 55% less than control plugs of equal size manufactured identically except for use of 95/5 acetone/water as the cellulose acetate solvent and utilization of a take-up speed of 1.7 meters per minute. In spite of the difference in weight, the smoke removal efficiency of both the novel plugs and the control plugs was 16%. A cross-section through individual filaments of the novel plugs showed voids as illustrated in FIG. 4 whereas filaments of the control plugs showed no such voids. The individual filaments ranged from about 8 to 12 denier with about 40 to 80 crimps per tensioned inch of length.

The smoke removal efficiency referred to above is determined by preparing filter cigarettes with 15 millimeter filters to be tested. For each test, ten cigarettes are each mechanically smoked employing twelve puffs each of 35 cubic centimeters volume and 2 seconds duration. The smoke leaving the cigarette passes through a trap having on its bottom a sintered glass disk. 2 grams of sifted alpha cellulose are placed on the disk, producing a pressure drop of about 5 centimeters of water. The trap is

immersed halfway in a Dry Ice-acetone bath. The formula for the smoke removal efficiency is as follows:

$$\text{Percent efficiency} = \frac{\Delta \text{ weight of filters}}{\Delta \text{ wt. of filters} + \Delta \text{ wt. of trap}} \times 100$$

It is to be understood that the foregoing detailed description is given merely by way of illustration and that many variations may be made therein without departing from the spirit of my invention.

Having described my invention what I desire to secure by Letters Patent is:

1. In the production of fibrous materials of low density, wherein a solution of cellulose acetate is extruded into water as a coagulant to form a plurality of filaments, and the filaments are taken up as a low density mass, the improvement which comprises employing as the solvent for said cellulose acetate aqueous acetone comprising at least about 20% by weight of water.

2. The process of claim 1 wherein said aqueous acetone comprises about 24 to 35% by weight of water.

3. The process for the production of fibrous materials of low density, which comprises extruding a solution of cellulose acetate in aqueous acetone comprising at least about 20% by weight of water through a plurality of orifices into water as a coagulant to form a plurality of filaments, and taking up said filaments at a speed less than about one-half the linear speed of extrusion through said orifices.

4. The process set forth in claim 3, wherein the filaments are withdrawn from the coagulant at less than about one-third their extrusion speed.

5. The process set forth in claim 3, wherein said cellulose acetate comprises at least about 15% by weight of said solution.

6. The process for the production of fibrous materials of low density, which comprises extruding a solution of about 15 to 30% by weight of cellulose acetate in aqueous acetone comprising about 24 to 35% by weight of water through at least about 100 orifices into water as a coagulant to form a plurality of filaments, and taking up said filaments at a speed less than about one-half the linear speed of extrusion through said orifices.

7. The process for the production of fibrous materials of low density, which comprises extruding a solution of about 20% by weight of cellulose acetate in aqueous acetone comprising about 28% by weight of water through at least about 1200 orifices into water as a coagulant to form a plurality of filaments, and taking up said filaments at a speed less than about one-half the linear speed of extrusion through said orifices.

8. The process set forth in claim 7 wherein said orifices are spaced apart less than about 2 millimeters.

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