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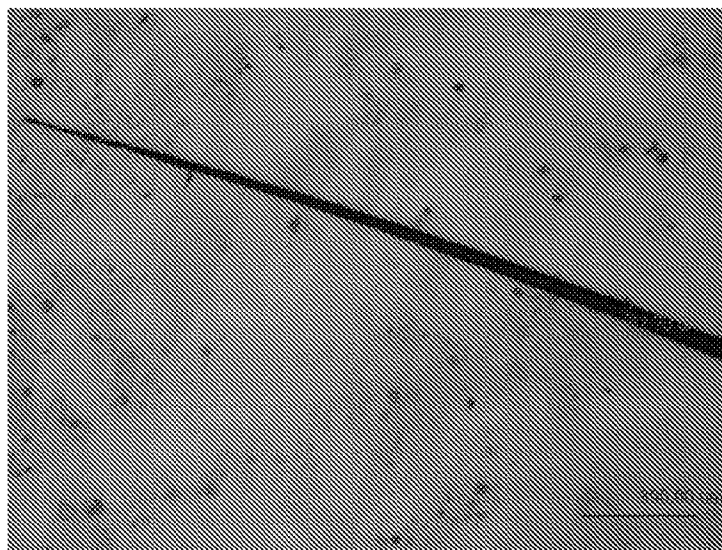


Fig. 1

(57) Abstract: The present invention relates to a tapered filament prepared from bio-based biologic-based material and a method for producing same. The present invention generally provides a method for producing a tapered filament, comprising the following steps of: a) providing filaments; b) providing a treating solution to treating tank; c) immersing the filaments in step a) into the treating solution, using auxiliary means such as mechanical stirring and/or cycling, or ultrasound energy for treating agent for enhancing effects of tapering, in which during step c), the depth of the filaments immersed in the treating agent varies with time.

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## **TAPERED FILAMENTS FROM BIO-BASED MATERIALS AND METHODS FOR PREPARING SAME**

5

### **FIELD OF THE INVENTION**

The present application relates to a tapered filament as well as a method for preparing same. More particularly disclosed herein is a tapered filament obtained from a bio-based material and a method for tapering using a multi-step process to achieve uniform tapering within a  
10 bundle of filaments.

### **CROSS-REFERENCE TO RELATED APPLICATIONS**

The present application claims priority to patent application filed in China on February 2, 2008 having the application number  
15 200810005818.7.

### **BACKGROUND OF THE INVENTION**

Generally, filaments for brushes are made of synthetic materials or animal hair. For example, nylon 612 and nylon 610 are typically used to  
20 prepare filaments for toothbrushes. In addition, petroleum-based polyesters such as polybutylene terephthalate (PBT), polyethylene terephthalate (PET) and polytrimethylene terephthalate (PTT) are also used to prepare filaments of one-off toothbrushes. Typically, the filaments of toothbrushes have a flat end, which cannot remove tartar embedded  
25 between teeth during tooth brushing. Consequently, these flat end filaments are unable to clean each position around teeth. Furthermore, filaments having flat ends are hard, and can be harmful to gums.

In addition, synthetic materials used for filaments are mainly produced from petroleum products, which are not bio-based, which does not address concerns relating to energy sources and the environment.

The paint brush, cosmetic brush, hair pencil and brush pen are often  
5 made of animal hair such as pig hair, fell, squirrel hair and weasel hair. They have naturally tapered ends, however, compared to synthetic materials they are expensive. At the same time, they may cause diffusion and cross-infection of some animal diseases and may lead to more and more protest from worldwide animal protection associations.

10 US6673444 has disclosed a filament for brushes, made of petroleum-based polyesters such as polytrimethylene terephthalate (PTT) or blends of polyesters with other materials. However, the filaments made of polyesters or blends of polyesters with other materials cannot satisfy all requirements. For example, with respect to nylon, polyester has a higher  
15 rigidity and therefore lower applicability. Therefore, the filaments made of polyesters are typically used to prepare cheap one-off toothbrush.

In order to solve the above problem, US2006/0088711 has disclosed a tapered polyester filament, which has one or two tapered ends formed by a chemical tapering process. The chemical tapering process comprises  
20 treating filaments by immersing the filament into strong acid or strong base solution under a temperature of 120 to 180°C. The mechanism of chemical tapering process is to corrupt polyester such as PTT or polybutylene terephthalate (PBT) gradually by chemical capillarity. However, conventional chemical tapering processes cannot produce uniform tapered  
25 filaments, is very rigorous, and the process conditions are harsh. These conventional processes also have a problem of high cost for maintenance of equipment.

Disclosed herein is a method to provide a tapered filament obtained from bio-based polyester using a chemical tapering process which has a mild treating condition over the prior art. The present invention discloses tapered polyester monofilament, demonstrated using a bio-based  
5 polyester (Sorona® polymer), wherein said tapered filament is produced using ultrasonic energy to achieve improved taper ratio/consistency under milder conditions than achieved using conventional methods.

### **SUMMARY OF THE INVENTION**

Disclosed herein is a method for producing a tapered filament,  
10 comprising the following steps of: a) providing filaments; b) providing a treating solution in a tank, wherein said treating solution comprises NaOH solution, KOH solution, LiOH solution, combinations of basic solutions thereof, sulfuric acid, hydrochloric acid, phosphoric acid solution, or combinations of acidic solutions thereof; c) immersing the filaments in step  
15 a) into the treating solution, d) using an auxiliary means for keeping the concentration of the treating solution uniform during the treatment, wherein during step c), the depth of the filaments immersed in the treating solution is varied with time.

Also disclosed herein is tapered filament obtained by the above  
20 method.

Another disclosure herein is use of tapered filament prepared as disclosed above in toothbrush, paint brush, cosmetic brush, hair pencil and brush pen applications.

### **BRIEF DESCRIPTION OF THE FIGURES**

Figure 1 is a micro-photograph of a filament bundle of example 1.

Figure 2 is a partial-enlarged view of a filament bundle of example 1.

Figure 3 is a micro-photograph of a filament in the center of the bundle of example 2.

Figure 4 is a micro-photograph of a filament in the periphery of the bundle of example 2.

5        Figure 5 is a micro-photograph of a filament in the center of the bundle of example 3.

Figure 6 is a partial-enlarged view of a filament in the periphery of the bundle of example 3.

10

### **DETAILED DESCRIPTION OF THE INVENTION**

Filament prepared from bio-materials and thereby is bio-based and has better applicability, such as better softness. Disclosed herein is a bio-based polyester filament having tapered ends, and the process for making same.

15

Also disclosed herein is a tapered filament obtained by a method wherein there is uniform taper length and taper ratio from the center of the tapered filament bundle to the peripheral outer edge of the filament bundle.

20

The process disclosed herein is a multi-step process that results in tapered ratio/consistency under relatively mild process conditions.

25

Chemically tapered polyester, such as Sorona® polymer (having a main component of PTT), is prepared by bio-based material, wherein said tapering treatment can be practiced under mild condition by dividing the one-step conventional chemical treatment used for tapering into a multi-step chemical treatment. Further, an auxiliary means to maintain concentration uniformity of the treating solutions during the tapering process is disclosed. The resultant filaments have better applicability and

more uniformly tapered length.

One embodiment of the present invention is a method for producing a tapered filament, comprising the following steps of: a) providing filaments; b) providing a treating solution for tapering filament; and c) immersing the  
5 filament of treating solution, using auxiliary means for keeping the concentration of the treating solution uniform during the treatment wherein during step c), the depth of the filaments impregnated in the treating solution varies with time.

In a preferred embodiment, the filaments comprise a bio-based  
10 polymer, such as Sorona® polymer, or blends of bio-based polymers combined with other polymers.

In yet another preferred embodiment, during step c), the initial depth of the filaments immersed into the treating solution is from 2 to 20 mm. The final depth of the filaments immersed into the treating solution is from 0 to  
15 10 mm, and the total time for treatment is from 5 to 60 min. Alternatively, the initial depth of the filaments immersed in the treating solution is from 0 to 10 mm. The final depth of the filaments immersed into the treating solution is from 2 to 20 mm, and the total time for treatment is from 5 to 60 min.

20 In the process disclosed herein, the filament used in the disclosed process has a length of from 16 to 65 mm. The treating solution comprises NaOH solution, KOH solution, LiOH solution, sulfuric acid, hydrochloric acid, phosphoric acid solution, or their combinations and step c) is carried out at a temperature of from 60 to 140°C.

25 In one preferred embodiment, auxiliary means is used to mechanically stirring and/or circulating the treating solution to maintain a uniform concentration of the solution.

It is also preferred that the filament(s) of the present invention comprise bio-based polymer. It is more preferred that the filament of the present invention comprises bio-based polyester. It is most preferred that the filament of the present invention comprise bio-based Sorona® polymer,  
5 which comprises PTT, or blends comprising Sorona® polymer.

In the present invention, the filament is conventional in the art. In one preferred embodiment, the filament can be prepared from polyesters, such as polybutylene terephthalate (PBT), polyethylene terephthalate (PET), polytrimethylene terephthalate (PTT) and their copolyesters, or their  
10 mixtures. In another preferred embodiment, the filament is prepared from Sorona® or its mixture with the above-mentioned materials.

Sorona® is a bio-based polymer provided by DuPont Company having a main component of polytrimethylene terephthalate (PTT). Sorona® is mainly obtained from fermentation of crop such as corn, therefore it is  
15 bio-based.

PTT useful in the invention is of the type made by polycondensation of terephthalic acid or acid equivalent and 1,3-propanediol; with the 1,3-propane diol preferably being of the type that is obtained biochemically from a renewable source, that is "biologically-derived" 1,3-propanediol.

20 As indicated above, the PTT resin composition comprises a predominant amount of a poly(trimethylene terephthalate).

Poly(trimethylene terephthalate) suitable for use in the invention are well known in the art, and conveniently prepared by polycondensation of 1,3-propane diol with terephthalic acid or terephthalic acid equivalent.

25 By "terephthalic acid equivalent" is meant compounds that perform substantially like terephthalic acids in reaction with polymeric glycols and diols, as would be generally recognized by a person of ordinary skill in the

relevant art. Terephthalic acid equivalents include, for example, esters (such as dimethyl terephthalate), and ester-forming derivatives such as acid halides (e.g., acid chlorides) and anhydrides.

Preferred are terephthalic acid and terephthalic acid esters, more preferably the dimethyl ester. Methods for preparation of PTT are discussed, for example in US6277947, US6326456, US6657044, US6353062, US6538076, US2003/0220465A1 and commonly owned U.S. Patent Application No. 11/638919 (filed 14 December 2006, entitled "Continuous Process for Producing Poly(trimethylene Terephthalate)").

10 A particularly preferred source of 1,3-propanediol is via a fermentation process using a renewable biological source. As an illustrative example of a starting material from a renewable source, biochemical routes to 1,3-propanediol (PDO) have been described that utilize feedstocks produced from biological and renewable resources such as corn feed  
15 stock. For example, bacterial strains able to convert glycerol into 1,3-propanediol are found in the species *Klebsiella*, *Citrobacter*, *Clostridium*, and *Lactobacillus*. The technique is disclosed in several publications, including previously incorporated US5633362, US5686276 and US5821092. US5821092 discloses, inter alia, a process for the  
20 biological production of 1,3-propanediol from glycerol using recombinant organisms. The process incorporates *E. coli* bacteria, transformed with a heterologous pdu diol dehydratase gene, having specificity for 1,2-propanediol. The transformed *E. coli* is grown in the presence of glycerol as a carbon source and 1,3-propanediol is isolated from the  
25 growth media. Since both bacteria and yeasts can convert glucose (e.g., corn sugar) or other carbohydrates to glycerol, the processes disclosed in these publications provide a rapid, inexpensive and environmentally



responsible source of 1,3-propanediol monomer.

The biologically-derived 1,3-propanediol, such as produced by the processes described and referenced above, contains carbon from the atmospheric carbon dioxide incorporated by plants, which compose the feedstock for the production of the 1,3-propanediol. In this way, the biologically-derived 1,3-propanediol preferred for use in the context of the present invention contains only renewable carbon, and not fossil fuel-based or petroleum-based carbon. The PTT based thereon utilizing the biologically-derived 1,3-propanediol, therefore, has less impact on the environment as the 1,3-propanediol used does not deplete diminishing fossil fuels and, upon degradation, releases carbon back to the atmosphere for use by plants once again. Thus, the compositions of the present invention can be characterized as more natural and having less environmental impact than similar compositions comprising petroleum based diols.

The biologically-derived 1,3-propanediol, and PTT based thereon, may be distinguished from similar compounds produced from a petrochemical source or from fossil fuel carbon by dual carbon-isotopic finger printing. This method usefully distinguishes chemically-identical materials, and apportions carbon material by source (and possibly year) of growth of the biospheric (plant) component. The isotopes,  $^{14}\text{C}$  and  $^{13}\text{C}$ , bring complementary information to this problem. The radiocarbon dating isotope ( $^{14}\text{C}$ ), with its nuclear half life of 5730 years, clearly allows one to apportion specimen carbon between fossil ("dead") and biospheric ("alive") feedstocks (Currie, L. A. "Source Apportionment of Atmospheric Particles," Characterization of Environmental Particles, J. Buffle and H.P. van Leeuwen, Eds., 1 of Vol.I of the IUPAC Environmental Analytical Chemistry

Series (Lewis Publishers, Inc) (1992) 3-74). The basic assumption in radiocarbon dating is that the constancy of  $^{14}\text{C}$  concentration in the atmosphere leads to the constancy of  $^{14}\text{C}$  in living organisms. When dealing with an isolated sample, the age of a sample can be deduced approximately by the relationship:

$$t = (-5730/0.693)\ln(A/A_0)$$

wherein  $t$  = age, 5730 years is the half-life of radiocarbon, and  $A$  and  $A_0$  are the specific  $^{14}\text{C}$  activity of the sample and of the modern standard, respectively (Hsieh, Y., Soil Sci. Soc. Am J., 56, 460, (1992)). However, because of atmospheric nuclear testing since 1950 and the burning of fossil fuel since 1850,  $^{14}\text{C}$  has acquired a second, geochemical time characteristic. Its concentration in atmospheric  $\text{CO}_2$ , and hence in the living biosphere, approximately doubled at the peak of nuclear testing, in the mid-1960s. It has since been gradually returning to the steady-state cosmogenic (atmospheric) baseline isotope rate ( $^{14}\text{C}/^{12}\text{C}$ ) of ca.  $1.2 \times 10^{-12}$ , with an approximate relaxation "half-life" of 7-10 years. This latter half-life must not be taken literally; rather, one must use the detailed atmospheric nuclear input/decay function to trace the variation of atmospheric and biospheric  $^{14}\text{C}$  since the onset of the nuclear age. It is this latter biospheric  $^{14}\text{C}$  time characteristic that holds out the promise of annual dating of recent biospheric carbon.  $^{14}\text{C}$  can be measured by accelerator mass spectrometry (AMS), with results given in units of "fraction of modern carbon" (fM). "fM" is defined by National Institute of Standards and Technology (NIST) Standard Reference Materials (SRMs) 4990B and 4990C, known as oxalic acids standards HOxI and HOxII, respectively. The fundamental definition relates to 0.95 times the  $^{14}\text{C}/^{12}\text{C}$  isotope ratio HOxI (referenced to AD 1950). This is roughly

equivalent to decay-corrected pre-Industrial Revolution wood. For the current living biosphere (plant material),  $\delta M \approx 1.1$ .

The stable carbon isotope ratio ( $^{13}\text{C}/^{12}\text{C}$ ) provides a complementary route to source discrimination and apportionment. The  $^{13}\text{C}/^{12}\text{C}$  ratio in a  
5 given biosourced material is a consequence of the  $^{13}\text{C}/^{12}\text{C}$  ratio in atmospheric carbon dioxide at the time the carbon dioxide is fixed and also reflects the precise metabolic pathway. Regional variations also occur. Petroleum,  $\text{C}_3$  plants (the broadleaf),  $\text{C}_4$  plants (the grasses), and marine carbonates all show significant differences in  $^{13}\text{C}/^{12}\text{C}$  and the  
10 corresponding  $\delta^{13}\text{C}$  values. Furthermore, lipid matter of  $\text{C}_3$  and  $\text{C}_4$  plants analyze differently than materials derived from the carbohydrate components of the same plants as a consequence of the metabolic pathway. Within the precision of measurement,  $^{13}\text{C}$  shows large variations due to isotopic fractionation effects, the most significant of which  
15 for the instant invention is the photosynthetic mechanism. The major cause of differences in the carbon isotope ratio in plants is closely associated with differences in the pathway of photosynthetic carbon metabolism in the plants, particularly the reaction occurring during the primary carboxylation, i.e., the initial fixation of atmospheric  $\text{CO}_2$ . Two  
20 large classes of vegetation are those that incorporate the " $\text{C}_3$ " (or Calvin-Benson) photosynthetic cycle and those that incorporate the " $\text{C}_4$ " (or Hatch-Slack) photosynthetic cycle.  $\text{C}_3$  plants, such as hardwoods and conifers, are dominant in the temperate climate zones. In  $\text{C}_3$  plants, the primary  $\text{CO}_2$  fixation or carboxylation reaction involves the enzyme  
25 ribulose-1,5-diphosphate carboxylase and the first stable product is a 3-carbon compound.  $\text{C}_4$  plants, on the other hand, include such plants as tropical grasses, corn and sugar cane. In  $\text{C}_4$  plants, an additional

carboxylation reaction involving another enzyme, phosphoenol-pyruvate carboxylase, is the primary carboxylation reaction. The first stable carbon compound is a 4-carbon acid, which is subsequently decarboxylated. The CO<sub>2</sub> thus released is refixed by the C<sub>3</sub> cycle.

5 Both C<sub>4</sub> and C<sub>3</sub> plants exhibit a range of <sup>13</sup>C/<sup>12</sup>C isotopic ratios, but typical values are ca. -10 to -14 per mil (C<sub>4</sub>) and -21 to -26 per mil (C<sub>3</sub>) (Weber et al., J. Agric. Food Chem., 45, 2942 (1997)). Coal and petroleum fall generally in this latter range. The <sup>13</sup>C measurement scale was originally defined by a zero set by pee dee belemnite (PDB) limestone,  
10 where values are given in parts per thousand deviations from this material. The “δ<sup>13</sup>C” values are in parts per thousand (per mil), abbreviated ‰, and are calculated as follows:

$$\delta^{13}\text{C} = \frac{(\text{^{13}C/^{12}C})_{\text{sample}} - (\text{^{13}C/^{12}C})_{\text{standard}}}{(\text{^{13}C/^{12}C})_{\text{standard}}} \times 1000\text{‰}$$

15 Since the PDB reference material (RM) has been exhausted, a series of alternative RMs have been developed in cooperation with the IAEA, USGS, NIST, and other selected international isotope laboratories. Notations for the per mil deviations from PDB is δ<sup>13</sup>C. Measurements are made on CO<sub>2</sub> by high precision stable ratio mass spectrometry (IRMS)  
20 on molecular ions of masses 44, 45 and 46.

Biologically-derived 1,3-propanediol, and compositions comprising biologically-derived 1,3-propanediol, therefore, may be completely distinguished from their petrochemical derived counterparts on the basis of <sup>14</sup>C (fM) and dual carbon-isotopic fingerprinting, indicating new  
25 compositions of matter. The ability to distinguish these products is beneficial in tracking these materials in commerce. For example, products comprising both “new” and “old” carbon isotope profiles may be

distinguished from products made only of "old" materials. Hence, the instant materials may be followed in commerce on the basis of their unique profile and for the purposes of defining competition, for determining shelf life, and especially for assessing environmental impact.

- 5 Preferably the 1,3-propanediol used as a reactant or as a component of the reactant in making PTT will have a purity of greater than about 99%, and more preferably greater than about 99.9%, by weight as determined by gas chromatographic analysis. Particularly preferred are the purified 1,3-propanediols as disclosed in US7038092, US7098368, US7084311  
10 and US20050069997A1.

The purified 1,3-propanediol preferably has the following characteristics:

- (1) an ultraviolet absorption at 220 nm of less than about 0.200, and at 250 nm of less than about 0.075, and at 275 nm of less than about 0.075;  
15 and/or
- (2) a composition having a CIELAB "b\*" color value of less than about 0.15 (ASTM D6290), and an absorbance at 270 nm of less than about 0.075; and/or
- (3) a peroxide composition of less than about 10 ppm; and/or  
20 (4) a concentration of total organic impurities (organic compounds other than 1,3-propanediol) of less than about 400 ppm, more preferably less than about 300 ppm, and still more preferably less than about 150 ppm, as measured by gas chromatography.

- PTT useful in this invention can be PTT homopolymers (derived  
25 substantially from 1,3-propane diol and terephthalic acid and/or equivalent) and copolymers, by themselves or in blends. PTT used in the invention preferably contain about 70 mole % or more of repeat units derived from

1,3-propane diol and terephthalic acid (and/or an equivalent thereof, such as dimethyl terephthalate).

In one embodiment the initial poly(trimethylene terephthalate) resin useful in the process further comprises 0.1 to 30 mole % repeat units, other than poly(trimethylene terephthalate), made from monomers selected from the group consisting of: terephthalic acid, isophthalic acid, 1,4-cyclohexane dicarboxylic acid, 2,6-naphthalene dicarboxylic acid, 1,3-cyclohexane dicarboxylic acid, succinic acid, glutaric acid, adipic acid, sebacic acid, 1,12-dodecane dioic acid, and the derivatives thereof such as the dimethyl, diethyl, or dipropyl esters of these dicarboxylic acids; and diols ethylene glycol, 1,3-propane diol, 1,4-butane diol, 1,2-propanediol, diethylene glycol, triethylene glycol, 1,3-butane diol, 1,5-pentane diol, 1,6-hexane diol, 1,2-, 1,3- and 1,4-cyclohexane dimethanol.

More preferably, the PTT resin composition contains at least about 80 mole %, or at least about 90 mole %, or at least about 95 mole %, or at least about 99 mole %, of repeat units derived from 1,3-propane diol and terephthalic acid (or equivalent). The most preferred polymer is poly(trimethylene terephthalate) homopolymer (polymer of substantially only 1,3-propane diol and terephthalic acid or equivalent).

The filaments of the present invention are useful in toothbrush, paint brush, cosmetic brush, hair pencil and brush pen applications. These filaments have advantages over the prior art in that there is provided a tapered filament prepared from a bio-based material, having a rounded or rough tapered end, and uniform tapered length. When used for toothbrushes, these filaments perform better to remove tartar between teeth. When used for paint brush, cosmetic brush, hair pencil and brush pen, applications, it can reduce cost, extend the life of brush and avoid

diffusion, and cross-infection of animal diseases.

In the present invention, the method for producing filament is conventional in the art, and a skilled person in the art can directly determine which method can be used to form a filament. In one preferred  
5 embodiment, the method comprises solution spinning, melt spinning, dry spinning, wet spinning and the like.

In step (a) of the present invention, the filament can be provided individually or can be provided in a form of filament bundle. In the present invention, the length of a filament is conventional in the art, and a  
10 skilled person in the art can adjust its length according to actual usage. Generally, the filament has a length of from 16 to 65mm, preferably from 20 to 60mm, more preferably from 30 to 50mm and most preferably 40mm.

In the present invention, there are no limitations on the shape of the  
15 cross area of the filament, as long as it can be applied to toothbrush, paint brush, cosmetic brush, hair pencil and brush pen. Generally, the shape may be, but is not limited to, circular, oval, square, rectangle, triangle, diamond, and the like.

In the present invention, the treating solution is conventional, and a  
20 skilled person in the art can directly determine which treating solution can be used. In one preferred embodiment, the treating solution comprises an acidic treating solution or a basic treating solution. In another preferred embodiment, the basic treating solution comprises NaOH solution, KOH solution, LiOH solution; and the acidic treating solution comprises sulfuric  
25 acid, hydrochloric acid, or phosphoric acid solution.

In the present invention, one may treat one or two ends of a filament. In one preferred embodiment, the treating temperature is from 60 to 140°C,

preferably from 80°C to 140°C, more preferably from 100°C to 140°C, and most preferably 120°C. The treating time, or the time that the filament is immersed in the treating solution, is from 10 minutes to 4 hours, preferably from 30 minutes to 3 hours, more preferably from 30 minutes to 2 hours,  
5 and most preferably from 30 minutes to 1.5 hours.

There are several key differences, individually and in combination with other differences, between the tapered filament according to the present invention and the conventional filament. The filaments disclosed herein are bio-based, rather than petroleum based. Also, rather than being  
10 prepared by one step treatment (i.e., the filament is immersed into a treating solution in a given depth at one time for a given time), the filaments disclosed herein are obtained from a bio-based product which is and is prepared by a method comprising the step of immersing filament into a treating solution successively for carrying out a multi-step treatment  
15 process (i.e., the depth of the filament into the treating solution varies with time). Further, the treating solution used in the process of the present invention is milder than solutions used in conventional tapering of filament using caustic solutions. There is also the option of incorporating auxiliary means for agitating the treating solution during treatment to maintain  
20 uniform concentration of the treating solution, and achieve uniformity in the tapering.

In the conventional chemically tapering processes, since the treating solution substantively remains non-flowing during treatment, the treating solution has different concentrations at different positions within the  
25 treating solution. The solution concentration gets depleted as the treating solution makes contact with the immersed filament. Consequently, concentration of the solution near the periphery bundle of filaments is low,



while the concentration away from the bundle of filaments is higher. In turn, treatment for tapering of filament in center of the bundle is not as effective as in periphery of the bundle.

The present invention provides an approach of incorporating auxiliary means for maintaining the uniformity of concentration in the treating solution during the tapering process. With respect to the specific auxiliary means, a skilled person in the art can select same according to actual requirements, as long as it can obtain the above object. In one preferred embodiment, the auxiliary means comprises mechanically stirring and circulating the treating solution. The mechanically stirring and circulating the treating solution are conventional in the art, respectively, and a skilled person in the art can determine their specific modes according to the disclosure of the present description. For example, the auxiliary means comprises, but not be limited to, a stirring rod, a magnetic stirring; and the circulation of the treating solution comprises use of a circulating bump. A preferred auxiliary means for maintaining the uniformity of concentration in the treating solution during the tapering process involves the use of ultrasound energy.

Total treating time is from 10 minutes to 4 hours, preferably from 30 minutes to 3 hours, more preferably from 30 minutes to 2 hours, and most preferably from 30 minutes to 1.5 hours.

In one preferred embodiment in the method according to the present invention, the initial depth of a filament immersed into a treating solution is from 2 to 20 mm, preferably from 1 to 15 mm, more preferably from 7 to 15 mm, and the final depth of a filament immersed into a treating solution is from 0 to 10 mm, preferably from 0 to 8 mm, more preferably from 0 to 5 mm, and most preferably 0 mm. The total treating time is from 5 to 60

minutes.

In another preferred embodiment, the initial depth of a filament immersed into a treating solution is 0 to 10 mm, preferably from 0 to 8 mm, more preferably from 0 to 5 mm, and most preferably 0 mm. The final  
5 depth of a filament immersed into treating solution is from 2 to 20 mm, preferably from 5 to 15 mm, more preferably from 7 to 15 mm. Total treating time is from 5 to 60 minutes.

In the method according to the present invention, the method of varying depth of immersion with time is conventional, and a skilled person  
10 in the art can determine directly this method in detail. Other preferred embodiments of the disclosed method comprise:

- (1) immersing filament into treating solution at a certain depth and treating for a time, then immersing at different depth and treating for a time, and repeating until a desired tapered filament is achieved;
- 15 (2) immersing filaments into a treating solution at a certain depth and treating for a time, then adding treating solution to increase the depth of the filaments that are immersed and treating the filaments again for a time; and repeating until repeating until a desired tapered filament is achieved;
- (3) immersing filament into a treating solution at a fixed rate such that  
20 the depth of filaments immersed into the treating solution reaches the final depth after a certain given total treatment time; or
- (4) immersing filaments into a treating solution in a certain depth, then raising the filaments at a fixed rate such that the filaments can leave the treating solution after a given total treatment time.

25 Although the present invention mentions the above methods for varying the immersion depth with time, the present invention will not be limited to these methods. A skilled person in the art can make, according

to his expertise, any modification and amendments to the above methods, and these modification and amendments will not depart from the scope of the method disclosed herein.

Since a process of multi-step tapering is used, the tapered end of the  
5 filament will contain different tapered sections having different tapering slopes such that the tapered end of the filament may have better applicability, such as better softness. At the same time, since the one-step tapering is replaced with the multi-step tapering and an auxiliary means is incorporated therein, the treating solution may be milder than conventional  
10 solutions and the filament may have more uniform tapered length.

In the present invention, the filament may have one tapered end or two tapered ends. If it has one tapered end, any end of the filament can be tapered. If it has two tapered ends, the two ends of the filament can be treated in the same or similar manner. The tapered filament disclosed  
15 herein has uniform taper length and taper ratio from the center of the tapered filament bundle to the peripheral outer edge of the filament bundle.

In the present invention, the method for preparing tapered filaments may further comprise a step of washing. The method for washing filaments  
20 is conventional in the art, and can be carried out by a skilled person in the art. Generally, the step of washing comprises rinsing filaments with water until the pH of the washings is neutral (near or at pH of 7).

In the present invention, the method for preparing tapered filaments can further comprise a step of drying. The method for drying filaments is  
25 conventional in the art, and can be carried out by a skilled person in the art. Generally, the step of drying comprises drying in an oven.

Another aspect of the present invention is a tapered, filament prepared

according to the present invention.

A preferred embodiment of the present invention is tapered filament made from Sorona® polymer, or its mixture.

5

### **EXAMPLES**

Tapered filament obtained from a bio-based material and the method for preparing same is also described according to the following examples. The unit used in the examples is percentage by weight. These examples are only exemplified and will not limit the scope of the present invention.

10

#### **Example 1**

1. Sorona® polymer was melt spun to form filaments and the formed filaments were thermally molded; bundles were formed by wrapping the resultant filaments through a pre-wrapping film.

15      2. The resultant bundles were cut into a length of 50 mm using a cutting knife.

3. An aqueous solution of 40% NaOH was added into a molding tank as treating solution; then the treating solution was heated to 120°C and the above bundles were immersed into the treating solution at a depth of  
20      10 mm. Mechanical stirring was carried out near the bundles. After 10 minutes, the bundles were raised 2mm such that the filaments were immersed in a depth of 8mm. After treatment of another 10 minutes, the bundles were raised another 2mm. The above steps were repeating until the bundles were completely above the level of treating solution.

25      4. The treating solution remaining on the surface of the tapered filaments was rinsed using fresh water. After dehydration in a spin-drier, the filaments were dried in the sun.

**Example 2**

Example 2 was carried out in the same manner as that in Example 1, except that in step 3 of Example 2, an aqueous solution of 40% NaOH was added into the molding tank as treating solution for tapering, and then the treating solution was heated to 120°C. The bundles were immersed into the treating solution at a depth of 4mm one time and a mechanically stir was carried out near the bundles. The treating time was 40 minutes to obtain tapered filaments.

10

**Example 3 (Comparative Example)**

Example 3 was carried out in the same manner as that in Example 1, except that in step 3 of Example 3, an aqueous solution of 40% NaOH was added into a molding tank as treating solution for tapering, and then the treating solution was heated to 120°C. The bundles were immersed into the treating solution in a depth of 4mm one time, without any auxiliary means. The bundles were treated in the treating solution for 40 minutes to obtain tapered filaments.

15

**Example 4**

Example 4 was carried out in the same manner as that in Example 1, except that in step 3 of Example 4, an aqueous solution of 60% NaOH was added into a tank as treating solution for tapering filaments, and then the treating solution was heated to 140°C. The treating solution was circulated by a pump to maintain the treating solution as unchanged. The bundles were immersed into the treating solution in a depth of 2mm. After 2 minutes, additional aqueous solution of 60% NaOH was added such that

20

25

the filaments were immersed at a depth of 4mm. The above steps were repeated. After 10 minutes, the treatment was complete.

### **Example 5**

5        Example 5 was carried out in the same manner as that in Example 1, except that in step 3 of Example 5, the concentration of the treating solution was 60%, the treating solution was heated to 60°C and the bundles were immersed into in a depth of 20mm. After 10 minutes, the bundles were raised for 5mm such that the filaments were immersed at a  
10    depth of 15mm. After another 10 minutes, the bundles were raised again for 5mm. The above steps were repeated until the treating time reached 30 minutes.

### **Example 6**

15        Example 6 was carried out in the same manner as that in Example 1, except that the bundles had a length of 65mm.

### **Observations**

20        From Figures 1 and 2, it is found that the tapered filaments obtained according to the present invention have a tapered end which varies, thereby increasing the softness and applicability.

25        From Figures 5 and 6, it is found that the tapered filaments obtained not according to the present invention have different tapered ends between the filaments in the center of the bundle and those in the periphery of the bundle, thereby reducing qualification rate.

## **CLAIMS**

### **What is claimed is:**

1. A method for producing a tapered filament, comprising the following  
5 steps of:

a) providing filaments;

b) providing a treating solution in a tank, wherein said treating solution  
comprises NaOH solution, KOH solution, LiOH solution, combinations of  
basic solutions thereof, sulfuric acid, hydrochloric acid, phosphoric acid  
10 solution, or combinations of acidic solutions thereof;

c) immersing the filaments in step a) into the treating solution,

d) using an auxiliary means for keeping the concentration of the  
treating solution uniform during the treatment,

wherein during step c), the depth of the filaments immersed in the  
15 treating solution is varied with time.

2. The method according to claim 1, wherein the filaments comprises  
bio-based polyester or blends comprising a bio-based polyester.

20 3. The method according to claim 1 or 2, wherein, during step c), the  
initial depth of the filaments immersed into the treating solution is from 2 to  
20 mm, the final depth of the filaments immersed into the treating solution  
is from 0 to 10 mm, and the total time for treatment is from 5 to 60 min.

25 4. The method according to claim 1 or 2 wherein during step c), the  
initial depth of the filaments immersed into the treating solution is from 0 to  
10 mm, the final depth of the filaments impregnated into the treating

solution is from 2 to 20 mm, and the total time for treatment is from 5 to 60 min.

5        5. The method according to claim 1, wherein the filament has a length of from 16 to 65 mm.

6. The method according to claim 1, wherein the step c) is carried out at a temperature of from 60°C to 140°C .

10       7. The method according to claim 1, wherein the auxiliary means comprises mechanically stirring, circulation the treating solution, or ultrasound energy applied to said treating solution.

15       8. A tapered filament obtained by the method according to claim 1 or 2 with uniform taper length and taper ratio from the center of the tapered filament bundle to the peripheral outer edge of the filament bundle.

20       9. The filament according to claim 8, wherein the filament may be applied in toothbrush, paint brush, cosmetic brush, hair pencil and brush pen.



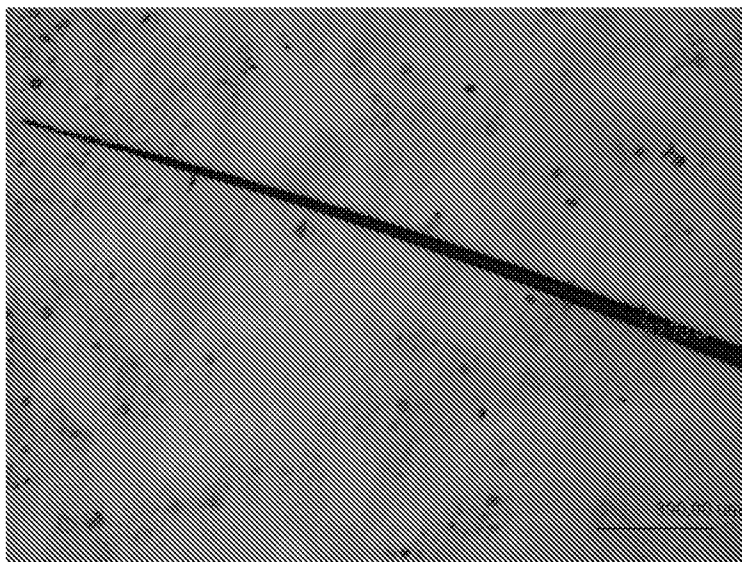


Fig. 1



Fig. 2

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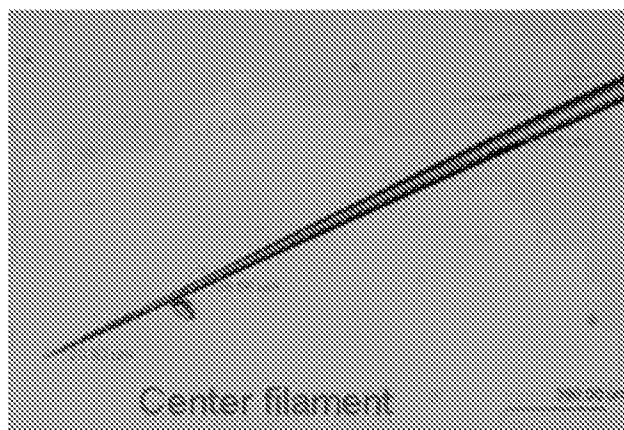


Fig. 3

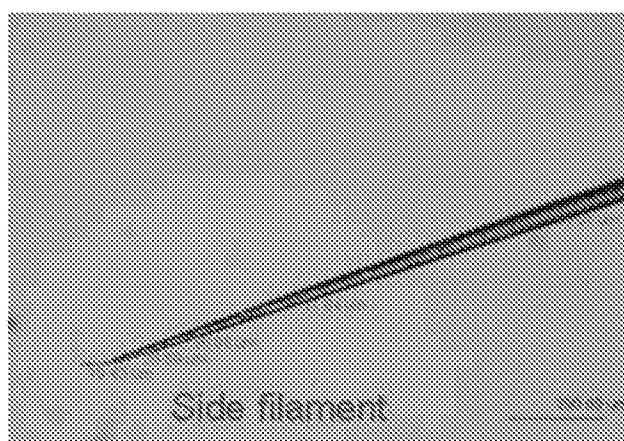


Fig. 4

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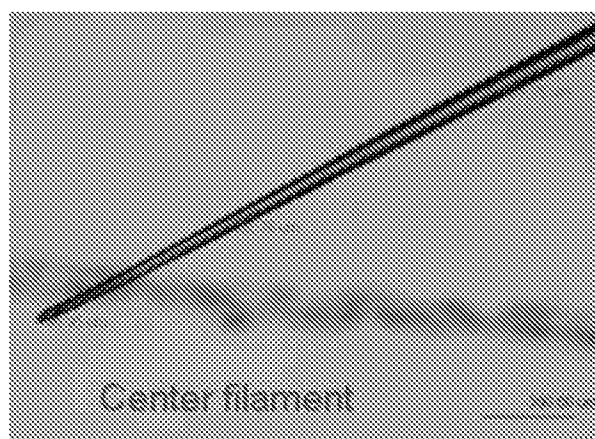


Fig. 5

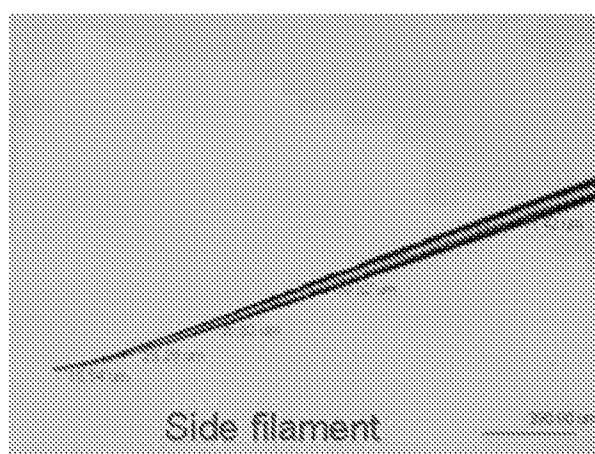


Fig. 6

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/US 09/32804

## A. CLASSIFICATION OF SUBJECT MATTER

IPC(8) - A46D 1/00; D01D 1/00 (2009.01)

USPC - 15/207.2

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)  
15/207.2Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched  
15/207.2Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)  
PubWEST(USPT,PGPB,EPAB,JPAB); Google Scholar; Google Patents  
bristle or filament or fiber, bio-based polymer, biodegradable, recyclable, tapering with chemical process, uniform

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

| Category* | Citation of document, with indication, where appropriate, of the relevant passages      | Relevant to claim No. |
|-----------|-----------------------------------------------------------------------------------------|-----------------------|
| X         | US 4,285,892 A (Betsuda et al.) 25 August 1981 (15.08.1981), entire document especially | 1,7,9                 |
| ---       | Table; col 2, ln 1-7; col 4, ln 6-14                                                    | -----                 |
| Y         |                                                                                         | 2-6,8                 |
| Y         | US 5,437,918 A (Taniguchi et al.) 1 August 1996 (01.08.1996), abstract                  | 2-6,8                 |
| Y         | US 2006/0088711 A1 (Nakamura et al.) 27 April 2006 (27.04.2006), para [0048]            | 3-6                   |
| Y         | US 5,722,106 A (Masterman et al.) 3 March 1998 (03.03.1998), col 5, ln 26 -29           | 8                     |

☐ Further documents are listed in the continuation of Box C.

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"&amp;" document member of the same patent family

Date of the actual completion of the international search

27 February 2009 (27.02.2009)

Date of mailing of the international search report

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