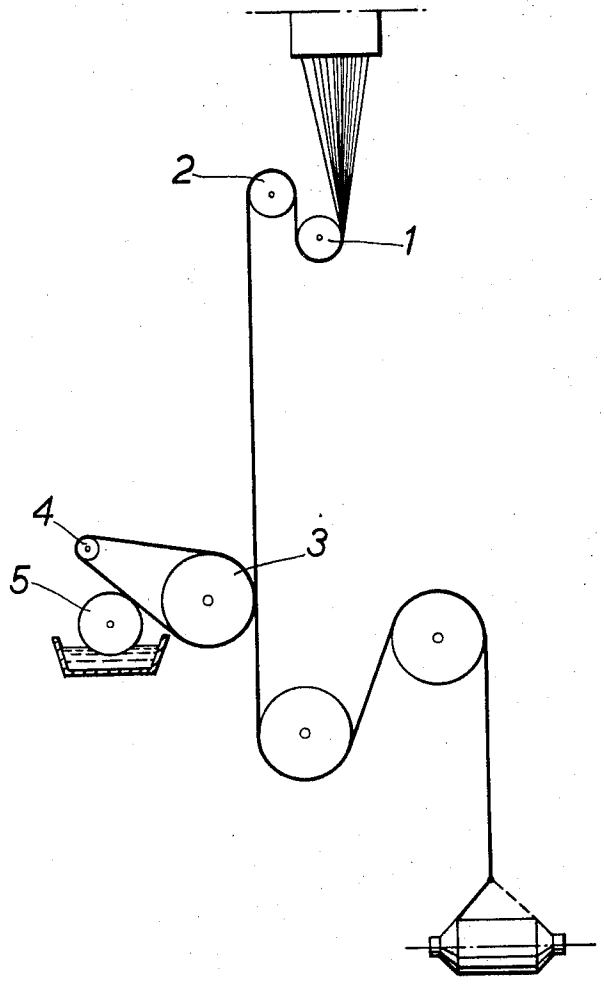
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PROCESS FOR THE PRODUCTION OF ORIENTATED SYNTHETIC FILAMENTS

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PROCESS FOR THE PRODUCTION OF ORIENTATED SYNTHETIC FILAMENTS

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ABSTRACT OF THE DISCLOSURE

The invention provides a process for the preparation of orientated synthetic filaments wherein continuous filaments extruded from a molten mass of linear synthetic polymer, e.g. polyhexamethylene adipamide or polyethylene terephthalate, are cooled to a temperature above 50° C. and at least 5° C. above the second order transition temperature of the polymer, but not above the initial orientation temperature; positively driven at a speed above the extrusion speed; cooled from this to a lower temperature which is still not less than ambient temperature and positively driven at a still higher speed. The process is easily controllable and enables filaments of uniform and desirable tensile properties to be obtained.

This is a continuation of Ser. No. 736,229, filed June 11, 1968 and now abandoned.

This invention relates to orientated synthetic filaments and their production.

In order that filaments based upon linear synthetic polymers and obtained by melt extrusion may have good mechanical properties, it is necessary that the polymer in these filaments should have a certain degree of crystallinity and orientation. This orientation is produced by stretching, at a temperature below the melting point of the polymer, either an already crystalline filament or an amorphous filaments. In the latter case, the orientation generally produces an incipient crystallisation.

In the processes at present most widely employed in industry, the extruded filaments are wound after cooling and subjected, in a subsequent phase, to the stretching operation, which is carried out with or without heating, depending upon the polymers and the nature of the filaments to be obtained.

Various single stage extrusion-stretching processes have been described, wherein the stretching may be effected by various means. For example, the extruded and solidified filaments may be taken up at a very high speed, in such manner that a sufficient tension is imparted to the filaments for bringing about an orientation.

This tension necessary for the orientation may also be obtained by means of the frictional forces imparted to the filaments as they pass continuously, after extrusion, through a liquid bath of appropriate density and at appropriate temperature.

While these two types of processes afford various advantages, they do not make it possible either to control accurately the extent of stretch imparted to the filament or to obtain filaments having a high degree of orientation.

The simple combination of a conventional device for extruding the filament with cooling and of a stretching device merely makes it possible to obtain filaments of comparable quality to the filaments obtained in two separate stages. Moreover, filaments which must be hot-stretched must be supplied with calories as in the conventional method. Whether this supply of calories be effected by passing them over a heated surface or through an atmosphere at appropriate temperature, it results in a heating of the yarn and of the filaments which is not uniform from the centre to the periphery and consequently produces a transverse heterogeneity of the stretched yarn and filaments.

The present invention provides a new process for the production of orientated synthetic filaments, wherein continuous filaments extruded from a molten mass of linear synthetic polymer are:

(a) cooled to a temperature of T_1 which is above 50° C., and at least 5° C. above the second order transition temperature of the polymer, but not greater than the initial orientation temperature;

(b) positively driven at a speed V_1 higher than the extrusion speed;

(c) brought to a temperature T_2 which is between T_1 and ambient temperature; and

(d) positively driven at a speed V_2 which is greater than V_1.

The initial orientation temperature, to which the temperature of the thread must fall before it undergoes the first positive drive is measured as follows:

A polymer in the form of a filament is extruded and wound in conventional manner after solidification; at the same time, the temperature along this filament between the spinneret and the winding point is measured by means of a temperature detector for a moving filament; the curve of this temperature is plotted as a function of the distance from the spinneret. The filament is then cut at the level of the spinneret and there is extracted from this point, a length equal to the distance between the spinneret and the winding point. After it has been verified that the variation of the cross-section of the thread over this length is identical to that existing in the course of the spinning between the spinneret and the winding, the birefringence is measured point-by-point commencing at the end corresponding to the spinneret, and the distance from which a measurable birefringence appears is noted. By plotting the value of this distance on the temperature/distance from the spinneret curve, the initial orientation temperature T_O is determined.

The second order transition temperature of the polymer T_G is measured for the purpose of the invention on a moisture-free non-orientated thread based upon the polymer under consideration.

This transition temperature of the second order T_G may be measured by dilatometry, by differential thermal analysis or by differential enthalpic analysis.

As will be understood, the extrusion temperature will depend on the particular polymer employed, suitable temperatures for the various polymers concerned being well known.

It is well known that the speed of extrusion is a function of the cross-section of the extrusion orifices, of their number and of the rate of flow of molten polymer. It does not constitute a critical characteristic of the invention and may vary within very wide limits.

The cooling of the filaments to a temperature at most equal to the initial orientation temperature T_O is influenced by a number of factors, and notably by: the count of the filaments, their speed of travel, the temperature of the atmosphere through which the filaments pass, and the length of the zone traversed.

The means by which the cooling of the thread can be most readily controlled is the temperature of the atmosphere through which it passes. The other factors, i.e. count, speed of the filaments and length of the zone through which they pass, are generally determined by technological requirements and considerations of production. An atmosphere at ambient temperature may be employed without disadvantage, as also a gas at higher temperature, but the latter is at most equal to T_1.

When the filaments have been cooled to a temperature T_1 as defined above, they are positively driven at a speed V_1 above the extrusion speed V_E. The upper limit of the ratio $V_1:V_E$ is determined essentially by the rheological parameters of the polymer.

The filaments are thereafter cooled to the temperature T_2. This temperature T_2 is preferably a few degrees lower than T_1. It is generally made lower than T_G, notably in the case of condensation polymers.

The length of this second cooling zone and the temperature of the atmosphere obtaining in this zone are the two factors which can be most readily varied for operating under the characteristic temperature conditions of the process according to the invention.

It is preferable to neutralise the static electricity on the filaments and this can be done, while cooling them, by passing them through a solution which simultaneously cools them and provides them with a coating of antistatic agent.

After having reached the temperature T_2, the filaments are driven at a speed V_2 higher than V_1. The ratio $V_2:V_1$ is chosen as a function of the extent of stretch which it is desired to impart to the filaments. It is generally between 3:1 and 6:1.

The time for which the filaments are kept in the orientation zone at temperatures above T_G influences the degree of crystallinity of the filaments.

An additional crystallisation may optionally be produced by thermal treatment of the orientated yarn. This thermal treatment may be carried out in various ways: after the filaments have reached the temperature T_2, they may again be brought to a temperature above T_G either continuously or in the course of a separate operation. The thermal treatment may be carried out under conditions such that the yarn contracts, retains its length or undergoes re-stretching.

The process according to the invention is applicable notably to: condensation polymers such as polyamides and copolyamides obtained by condensation of diacids and diamines, e.g. polyhexamethylene adipamide, or aminoacids, e.g. polyundecanamide, or by polymerisation of lactams, e.g. polycapronamide; polyesters such as polyethylene terephthalate and copolyesters comprising at least 75% of ethylene terephthalate units in the chain; and to addition polymers such as polypropylene.

For carrying out the process of the invention it is desirable that the means employed to drive the filaments at the speed V_1 should not suddenly change the temperature of the filaments at this point.

Preferably, rollers maintained at the same temperature as the filaments are employed. However, the temperature of the rollers employed influences the properties of the filaments, and more particularly the contraction properties, and it may be desirable to utilise this factor in the preparation of filaments having particular characteristics.

The preferred means for driving the filaments at the speed V_2 are also rollers which may be either at the temperature T_2 or at a higher temperature, depending on whether or not it is desired to subject the filaments to a thermal treatment at this point.

In the process of the invention, the dynamometric properties of the threads which it is desired to obtain can be precisely controlled, and products having high regularity of count and a good structural homogeneity can be continuously prepared.

The process according to the invention affords the possibility of stretching very uniformly the various filaments of a multi-filament product, since the distance between the spinneret and the first driving rollers may be sufficiently low for the filaments to be disposed on this roller without difficulty and stretched in the form of a tow rather than assembled in compact form, so that all the filaments undergo exactly the same extent of stretch.

On the other hand a regular stretch could not be effected if the filaments were driven at the speed V_1 before being cooled to a temperature not greater than T_O, because under these conditions the fibres would adhere to the driving rollers. On the other hands, if the filaments were cooled to a temperature below T_G they would slip on the rollers and it would no longer be possible to effect a regular stretch.

In the following examples, which illustrate the invention, the tenacity and the elongation are measured on the Instron dynamometer, the contraction is measured on "skeins" after remaining in hot air at 150° C. for 30 minutes, the value of the modulus is given by the slope of the tangent to the origin of the dynamometric curve corresponding to a specimen of a length of 50 cm. and to a break time of 20 to 30 seconds and the birefringence is measured on filaments without tension.

EXAMPLES 1–2

A polyhexamethylene adipamide having a relative viscosity of 36 measured at 25° C. on an 8.4% by weight solution in 90% formic acid has a transition point of the second order at 50° C. and its intial orientation temperature is 100° C.

This polymer is extruded at 292° C. through a spinneret having 23 holes of a diameter of 0.34 mm.

The extruded filaments pass through a calm atmosphere maintained at ambient temperature.

When these filaments have reached a temperature of 75° C., i.e. at a distance of 54 cm. from the spinneret, they are driven at 295 m./min. by a supply system consisting of 2 rollers heated at 75° C. and rotating in opposite directions, which are diagrammatically illustrated at **1** and **2** in the accompanying FIG. 1.

The filaments thereafter pass through a zone 65 cm. long, in which a calm atmosphere at ambient temperature also obtains and in which they are cooled to this temperature. They are then driven by a stretching roller **3** associated with an idler **4** maintained at 25° C. Disposed in the passage of the filaments between the stretching roller **3** and the idler **4** is a batching device **5**. The filaments are thereafter wound in known manner. The following results are obtained for various degrees of stretch:

Example	1	2
Peripheral velocity of the stretching rollers, m./min	1,240	1,300
Temperature of the filaments at the level of the stretching rollers, ° C	25	25
Count:		
dtex	100	96.7
d	90	87
Number of filaments	23	23
Tenacity:		
g./tex	36	39.6
g./d	4.0	4.4
Elongation, percent	33	29.6

EXAMPLES 3–4

A polyamide identical to that of Example 1 is extruded under the same conditions with a rate of flow of 0.36 g./min. per hole, and then orientated under the conditions set out in the following table:

Example	3	4
Distance between spinneret and supply roller, cm	55	47
Temperature of the filaments at the level of the supply rollers, ° C	55	65
Temperature of the supply rollers, ° C	55	65
Peripheral velocity of the supply rollers, m./min	300	300
Distance between the supply and stretching rollers, cm	60	70
Peripheral velocity of the stretching rollers, m./min	1,260	1,200
Temperature of the filaments at the level of the supply rollers, ° C	25	25
Count:		
dtex	73	78
d	66	70
Tenacity:		
g./tex	43.2	37.8
g./d	4.8	4.2
Elongation, percent	35.5	37.6
Modulus:		
g./d	30	27
g./tex	270	248
Contraction, percent	6.2	5.9

These tests show that, in the temperature range between 55° and 65° C., the temperature of the thread at the level of the supply rollers only slightly affects the strength and the elongation, which are above all influenced by the value of the extent of stretch.

On the other hand, if the filaments are cooled to a temperature of 45° C. before being passed over supply rollers at the same temperature, a regular stretch is no longer possible: slipping occurs on the supply rollers and the degree of stretch cannot be maintained at a constant value.

If the temperature of the filaments at the supply rollers is above 100° C., the filaments stick together and to the rollers: the stretching cannot be effected under good conditions.

EXAMPLE 5–10

A polyhexamethylene adipamide identical to that of Example 2 is extruded under the same conditions. When the filaments have reached a temperature of 60° C., i.e. at 51 cm. from the spinneret, they are driven at 300 m./min. by supply rollers maintained at 60° C.

After having passed through a calm atmosphere over a distance of 60 cm., maintained at ambient temperature, the filaments are driven by stretching rollers under the conditions indicated in the following table, in which the characteristics of the filaments obtained are also indicated:

Example	5	6	7	8	9	10
Speed of the stretching rollers, m./min	450	600	750	900	1,050	1,200
Temperature of the stretching rollers, ° C	25	25	25	25	25	25
Count:						
dtex	170	130	104	88	75	68
d	155	118	95	80	68	61
Tenacity:						
g./tex	13.5	18.9	27	29.7	37.8	46
g./d	1.5	2.1	3	3.3	4.2	5.1
Elongation, percent	266	165	106	66	42	28.5
Contraction, percent	1.6	0.6	1.2	1.9	3.4	5.5
Modulus of elasticity:						
g./tex		77.4	116	172	215	306
g./d		8.6	12.9	19.1	24	27.6
Birefringence	0.025	0.041	0.048	0.051	0.053	0.057

It will be seen from these examples that it is possible to obtain by the process of the invention polyamide filaments having high elongation and low contraction. Filaments stretched by conventional methods and having characteristics of tenacity and elongation substantially identical to those of Examples 5 to 8 have substantially higher contractions of the order of 8–12%.

It is found that in the process of the invention the tenacity and the elongation of the filaments vary continuously as a function of the extent of stretch, while the values of the contraction pass through a minimum.

EXAMPLE 11

A copolyamide obtained from 95 parts by weight of hexamethylene diamine adipate and 5% by weight of caprolactam, having a relative viscosity of 36, is extruded through a spinneret having 23 holes of a diameter of 0.34 mm. at a rate of flow of 0.36 g./min./hole. At 55 cm. from the spinneret, when the filaments have reached a temperature of 55° C., they are driven at 300 m./min. by a system of supply rollers maintained at 55° C. After having travelled 60 cm. through a calm atmosphere maintained at ambient temperature, the filaments are driven at 1380 m./min. by a system of stretching rollers at 25° C.

The filaments obtained have a count of 69 d./tex (62 d.), a tenacity of 45 g./tex (5 g./d.), an elongation of 24.7% and a contraction of 8%.

EXAMPLES 12–13

Polyethylene terephthalate having an intrinsic viscosity of 0.67 has a transition point of the second order at a temperature T_G of 70° C. and an initial orientation at a temperature T_O of 130° C.

This polymer is extruded at 285° C. through a spinneret having 30 holes of a diameter of 0.34 mm. in an atmosphere maintained at ambient temperature.

At 53 cm. from the spinneret, the filaments have reached a temperature of 90° C. and are driven at 300 m./min. by a system of supply rollers maintained at 90° C. After having travelled a distance of 1 m. through an atmosphere maintained at ambient temperature and having been passed over a batching device, the filaments, which have reached a temperature of 25° C., are driven at 1500 m./min. by a system of stretching rollers heated at the temperature indicated in the following table, in which the properties of the filaments obtained are also set out:

Example	12	13
Temperature of the stretching rollers, ° C	150	170
Count:		
dtex	167	167
d	150	150
Tenacity:		
g./tex	41.5	41.5
g./d	4.6	4.6
Elongation, percent	20	19
Modulus:		
g./tex	1,080	950
g./d	120	104
Contraction, percent	12	9.7

It will be seen from these examples that when the temperature of the stretching rollers increases, the tenacity and the elongation of the filaments are only slightly modified, while the modulus and the contraction decrease.

EXAMPLE 14

Filaments of polyethylene terephthalate which have reached a temperature of 25° C., and which have been extruded under the same conditions as in Example 13 and are driven at 280 m./min. by a system of supply rollers maintained at 90° C., are passed over a first train of stretching rollers maintained at 200° C. and rotating at a peripheral velocity of 1400 m./min. The filaments are thereafter continuously conducted to a second train of stretching rollers maintained at 25° C., which is situated at 30 cm. from the first stretching train and which drives these filaments at 1680 m./min.

The yarn obtained, of 150 d./tex (135 d.), 30 filaments, has a tenacity of 60.3 g./tex (6.7 g./d.), an elongation of 9% and a contraction of 13.5%; its modulus is 1080 g./tex (120 g./d.).

EXAMPLE 15

The polyhexamethylene adipamide filament of Example 10 is re-stretched to 1.4 times its length, at 140 m./min., by passing it over a cold finger and a plate heated at 180° C.

The resultant filament has a tenacity of 74 g./tex (8.2 g./d.), an elongation of 12.6%, a modulus of 414 g./tex (46 g./d.) and a contraction of 4.7%.

This test shows that the process according to the invention makes it possible to obtain polyamide filament of very high modulus and low contraction.

EXAMPLE 16

A polyethylene terephthalate identical to that employed in Example 12 is extruded and the filaments obtained are driven under the conditions described in the said Example 12. These filaments are thereafter passed over a system of stretching rollers maintained at 25° C. and rotating at a peripheral velocity of 1500 m./min.

The filament obtained is re-stretched to 1.3 times its length, at 130 m./min., by passing it over a finger maintained at 90° C. and a plate heated at 230° C. There is finally obtained a filament having a tenacity of 74 g./tex (8.2 g./d.), an elongation of 6.5% and a contraction of 5.5%.

What is claimed is:

1. A process for the production of oriented continuous filaments of polyesters or polyamides of fiber-forming molecular weight by spinning the polymer and orienting the resulting filament by stretching in which the spinning and stretching are carried out as a single continuous process, which comprises as a series of successive steps, one following immediately on one another in the following order:

(a) extruding as continuous filaments the molten polymer into an atmosphere at substantially the ambient temperature;

(b) cooling the filaments so formed directly to a temperature T_1 between 55° and 130° C. which is (i) above 55° C., (ii) at least 5° C. above the second order transition temperature of the polymer, and (iii) not higher than the maximum temperature at which stretching filaments of the polymer orients them;

(c) positively forwarding the filaments when they have reached the said temperature T_1 at a speed V_1 higher than the extrusion speed, whereby the freshly extruded filaments are attenuated without being substantially oriented;

(d) cooling the filaments further to a temperature T_2 below 75° C. which is also between T_1 and the ambient temperature, by causing them to continue to travel through an atmosphere at the ambient temperature; and

(e) positively forwarding the filaments while they are at the said temperature T_2 at a speed V_2 which is greater than V_1, the ratio V_2/V_1 being from 3:1 to 6:1, whereby the filaments so produced are oriented.

2. Process according to claim 1, wherein filaments of polyhexamethylene adipamide are made, the temperature T_1 is between 55° and 100° C., the temperature T_2 is between 55° C. and the ambient temperature, and the filaments are forwarded in steps (c) and (e) by means of rotating surfaces whose temperatures are T_1 and T_2, respectively.

3. Process according to claim 1, wherein filaments of polyethylene terephthalate are made, the temperature T_1 is between 75° and 130° C., the temperature T_2 is between 75° C. and the ambient temperature, and the filaments are forwarded in steps (c) and (e) by means of rotating surfaces whose temperatures are T_1 and T_2, respectively.

4. Process according to claim 2, wherein the temperature T_2 is the ambient temperature.

5. Process according to claim 3, wherein the temperature T_2 is the ambient temperature.

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