

## SPINNING DEVICE

The invention is concerned with a spinning device for carrying out the amine-oxide process according to the dry/wet-spinning process, wherein a spinneret is used comprising spinning holes for extruding filaments, a container containing spinning bath liquid, a bundling means provided in the spinning bath liquid for bundling the extruded filaments and an air gap defined as the distance of the spinneret to the surface of the spinning bath liquid.

In general terms, the technique of the dry/wet-spinning process consists in that dope is extruded through a forming tool, such as a spinneret, into a medium which is non-precipitating for the dope, such as air or an inert gas, whereby, when a spinneret is used, filaments are produced which are stretched in this medium and subsequently conducted into a spinning bath liquid (precipitation bath), wherein the filaments coagulate.

In general terms, amine-oxide process refers to the production of cellulose moulded bodies using tertiary amine-oxides. In this process, cellulose is dissolved in a mixture of a tertiary amine-oxide and water, the solution is moulded by means of a forming tool and conducted through an aqueous precipitation bath wherein the cellulose is precipitated. As the amine-oxide, primarily N-methylmorpholine-N-oxide (NMMO) is used. Other amine-oxides are described e.g. in EP-A - 0 553 070. A process for the production of mouldable cellulose solutions is known i.e. from EP-A - 0 356 419.

The operation of the amine-oxide process according to the dry/wet-spinning process is known e.g. from DE-A - 29 13 589.

From WO 93/19230 and WO 95/04173 of the applicant, an advantageous embodiment of the amine-oxide process and a device for the production of cellulose fibres are known, whereby a solution of cellulose in a tertiary amine-oxide is moulded in hot state, and the moulded solution is introduced through a gaseous medium (air)

-2-

into the precipitation bath to precipitate the cellulose therein contained, the hot, moulded solution being cooled before introducing it into the precipitation bath. Cooling is carried out immediately after moulding, consisting preferably in that air is blown horizontally to the cellulose moulded body. By means of the process according to that invention the cellulose solution can be spun with a high filament density, preventing the adhesion between individual filaments after being extruded from the spinneret.

DD-A - 218 121 is also concerned with a dry/wet-spinning process for the production of cellulose fibres from cellulose solutions in tertiary amine-oxides. Also according to this process, the cellulose solution is spun into an air gap, i.e. the space between the spinneret and the surface of the spinning bath liquid, stretched and conducted into an aqueous precipitation bath. In DD-A - 218 121 it is mentioned that the air gap may be reduced without any negative effects on the spinning safety, when before spinning a polyalkylene ether is added to the cellulose solution. A small air gap is advantageous, since the risk of an adhesion between the individual fresh extruded filaments is reduced.

In EP-A - 0 574 870, a dry/wet-spinning process for processing solutions of cellulose in tertiary amine-oxides is described, pointing out the advantage of a small air gap. According to the specification of this patent application, this spinning process can be carried out employing a small air gap and a high number of spinning holes per area unit. It is maintained that inspite of these conditions, the adhesion between the individual filaments during the spinning operation can be prevented. It is recommended to carry out the contacting of the spun filaments with the spinning bath liquid in a spinning funnel. Spinning bath liquid is conducted through this spinning funnel in parallel flow to the filaments. The axis of the spinning funnel is substantially vertical to the plane of the spinneret, and the flow of the spinning bath liquid is directed downwards from

-3-

above, the flow originating generally from the gravitational fall of the spinning bath liquid.

According to EP-A - 0 574 870, the drawing or stretching of the fresh extruded filaments is attained in that the filaments are accelerated substantially to their drawing rate by the spinning bath liquid flowing through the spinning funnel.

This known spinning device has the disadvantage that the funnel pipe of the spinning funnel sets an upper limit to the filament bundles to be conducted therethrough with regard to its total section, due to its relatively small diameter. Additionally, this limit is set at an unsatisfactorily low level for an industrial-scale realization of the process. Thus, according to the experience of the present applicant, when employing a diameter of 6 mm, as exemplified in EP-A - 0 574 870, it is only possible to conduct a filament bundle consisting of not more than 100 filaments through the funnel, since spinning bath liquid also has to be transported through the funnel. This means that when such a spinning funnel is used, only a spinneret comprising not more than 100 spinning holes may be employed.

On the other hand, when a wide spinneret having thousands of spinning holes is used, such as described for instance in the Austrian Patent AT-B 397.392 of the applicant, the funnel pipe has to be appropriately wider and thus a lot more spinning bath liquid is discharged and has to be circulated. This high throughput of spinning bath liquid causes turbulent flows in the spinning bath, interfering with the dry/wet-spinning process.

In GB-A - 1,017,855, a device for dry/wet-spinning of synthetic polymers is described, recommending also the use of a spinning funnel, where through spinning bath liquid is allowed to flow in parallel flow to the extruded fibres. The

-4-

spinneret is located approximately 0,5 cm above the spinning bath surface.

It is the object of the invention to provide a spinning device whereby it is possible to carry out the amine-oxide process according to the dry/wet-spinning process without complex equipment and achieving a good spinnability (high spinning safety). A good spinnability means attaining a maximum final drawing as high as possible (minimum titer) before the fibre breaks. Another indicator for spinnability is the period of time during which spinning is possible without the occurrence of spinning deficiencies which require technical assistance. Moreover, even when using spinnerets having high hole densities, the adhesion of the fresh extruded filaments in the air gap is to be prevented, and a titer as constant as possible (low titer variations) is to be attained.

The spinning device according to the invention for carrying out the amine-oxide process according to the dry/wet-spinning process comprising

- a spinneret having spinning holes for extruding filaments,
- a blowing device whereby the extruded filaments may be cooled immediately after being delivered from the spinning holes,
- a container containing spinning bath liquid,
- a bundling means provided in the spinning bath liquid for bundling the extruded filaments, and
- an air gap defined as distance of the spinneret to the surface of the spinning bath liquid,

is characterized in that

- the bundling means is located at such a distance from the spinneret that the angle ( $\alpha$ ) formed by the filaments with respect to the vertical to the surface of the spinning bath liquid does not exceed  $45^\circ$  and
- that the relation

-5-

$$0,1+0,0051 \leq 0,7 \cdot d_0 \cdot \frac{(h-l)}{h}$$

is fulfilled, wherein  $d_0$  is the distance (mm) between a spinning hole and its neighbouring spinning hole on the spinneret,  $h$  is the distance (mm) of the bundling means to the spinneret and  $l$  is the air gap (mm), and wherein

$$0,4 \text{ mm} \leq d_0 \leq 2 \text{ mm} \text{ and} \\ 0 \text{ mm} < l < 60 \text{ mm}.$$

It has been shown that the object of the invention is attained by designing the employed spinning device such that the two criteria mentioned above (angle  $\alpha$  does not exceed  $45^\circ$ ; inequations mentioned above are fulfilled) are met. When using spinnerets having a high hole density it is necessary to cool the freshly extruded filaments immediately after being delivered by the spinning holes. This cooling is known to the skilled person from the state of the art (see e.g. WO 95/04173 of the applicant).

A preferred embodiment of the spinning device according to the invention is characterized in that the bundling means is designed as a diverting means whereon the filaments are not only bundled but also diverted.

It has proven advantageous to provide the diverting means such that it will not rotate when diverting the filaments. Thus, according to this embodiment, no rotatable roll or cylinder is provided. This serves to prevent torn-off filaments from wrapping around the diverting means, thus facilitating the operation of the amine-oxide process.

Another preferred embodiment of the spinning device according to the invention is characterized in that the angle  $\alpha$  does not exceed  $20^\circ$ . It has been shown that it is of vital importance for the spinning safety in the dry/wet-spinning process that the drawing angle  $\alpha$  in the air gap is as small

-6-

as possible and preferably does not exceed 20°. Thus the risk of an adhesion between individual filaments in the space between the spinneret and the surface of the spinning bath can be minimized and the spinning safety can be increased.

The invention is also concerned with a spinning device for carrying out the amine-oxide process according to the dry/wet-spinning process comprising

- a spinneret having spinning holes for extruding filaments,
- a blowing device whereby the extruded filaments may be cooled immediately after being delivered from the spinning holes,
- a container containing spinning bath liquid,
- a diverting means provided in the spinning bath liquid for bundling and diverting the extruded filaments, and
- an air gap defined as the distance of the spinneret to the spinning bath liquid,

which is characterized in that the diverting means is designed such that it will not rotate when diverting the filaments.

Another appropriate embodiment of the spinning device according to the invention consists in that the spinneret comprises:

- a substantially rotationally symmetrical nozzle body having in its center a feeding for cooling gas,
- a feeding for the cellulose solution,
- an annular spinning insert having spinning holes and
- a baffle plate for diverting the cooling gas flow towards the filaments which are extruded from the spinning holes, so that the cooling gas stream hits the filaments essentially at right angles.

That embodiment enables spinning with a still higher hole density while at the same time effectively preventing the adhesion of the fresh extruded filaments in the air gap.

-7-

Cooling of an annular filament bundle by means of blowing cooling air at it is known from WO 95/04173 of the applicant.

Another preferred embodiment of the spinning device according to the invention consists in that the container containing the spinning bath liquid is connected to a lifting device whereby the container can be moved towards and away from the spinneret in vertical direction, distance  $l$  thus being varied, and that the bundling means is arranged such that the distance  $h$  remains constant inspite of this movement.

By means of the drawing consisting of Figures 1, 2 and 3, an embodiment of the invention will be illustrated. The drawing shows a general illustration of the dry/wet-spinning process including the relevant parameters for the relation according to the invention.

In Figure 1, 1 denotes a container for the spinning bath liquid, the surface of the spinning bath liquid being indicated by 1a. During spinning operation, dope is extruded through spinneret 3, and extruded filaments 4, 5 are drawn across the air gap  $l$  into the spinning bath liquid, wherein they coagulate. At diverting means 2, which is a non-rotatable cylindrical bar, the coagulated filaments are bundled, diverted and drawn diagonally upwards. As the air gap  $l$ , the distance of the bottom of spinneret 3 to the surface 1a of the spinning bath liquid is defined. The angle defined above formed by the filaments vertically to the surface of the spinning bath liquid is indicated by  $\alpha$ .

Reference number 4 denotes a filament delivered from a spinning hole located at the outermost edge of an annular ring formed by the spinning holes in spinneret 3.  $d_1$  is the radius (mm) of the circle limiting the annular ring formed by the spinning holes towards the outside.  $d_0$  indicates the distance of this spinning hole to its neighbouring spinning hole 5, denoting in each case the distance between each of the centers of the two neighbouring spinning holes.  $h$  is the

-8-

distance of diverting means 2 to spinneret 7 and 1 is the air gap.

According to the embodiment described in Figure 1, container 1 is located on a lifting device (not shown) whereby container 1 can be moved vertically, thus varying in a simple way the size of air gap 1.

It has proven particularly advantageous not to fix diverting means 2 to container 1, but provide that container 1 can be moved while at the same time distance h remains constant. In this simple way, air gap 1 can be varied while keeping distance h constant. This represents an essential simplification when adjusting the spinning device according to the invention. Figures 2 and 3 show such embodiments of the spinning device according to the invention.

Figure 2 substantially shows the spinning device of Figure 1, the same means being denoted by the same reference numbers. The non-rotatable diverting means 2 is connected to a fixed means 7 by means of a rigid arm 6 not connected to container 1, so that when lifting or lowering container 1 means 7 will not be moved simultaneously. Means 7 may be for instance a wall. In Figure 2, two positions of container 1 are indicated, the lower position being indicated by a dotted line. The device for lifting and lowering container 1 is not shown. From Figure 2 it is evident that by means of lifting and lowering container 1, the air gap can be shortened or lengthened, while distance h remains constant.

Figure 3 illustrates another embodiment of the spinning device according to the invention. In this embodiment, diverting means 2 is anchored to the ground by means of a rigid arm 9. Arm 9 rises to an appropriate outlet 11 provided in container 1. To prevent liquid from being lost from said container 1, a jacket 10 for sealing is provided, which simply is folded when container 1 is lowered by means of a device not shown.

By means of the following Examples 1, 2, 3 and 4, the invention will be described in still more detail, Examples 1 and 2 revealing the effect of angle  $\alpha$  on the spinnability of cellulose solutions. Example 4 demonstrates the advantageous effect of a non-rotatable diverting means on spinnability.

#### Example 1

A spinning device corresponding substantially to Figure 1 was used, although as the bundling means a spinning funnel according to EP-A - 0 574 879 was used. As the spinneret, the one described in WO 95/04173, which is incorporated herein by reference, was used.

This known spinneret (number of holes: 3960; hole diameter: 100  $\mu\text{m}$ ; external diameter of the nozzle (outermost row of holes)  $d_1$ : 145 mm) exhibits a substantially rotationally symmetrical nozzle body having in its center a feeding for cooling gas, a feeding for cellulose solutions (13,5% of cellulose; temp.: 120°C), an annular, deep-drawn spinning insert of precious metal having spinning holes, said spinning insert having a tub-shaped section, and a baffle plate to direct a cooling gas stream to cellulose filaments which are extruded from the spinning holes (output: 0,025 g/min), so that the cooling gas stream (24 m<sup>3</sup>/h) strikes the extruded cellulose filaments substantially essentially at right angles. The spinning holes in the spinning insert are spaced substantially at identical distances from each other (hole/hole-distance  $d_0$ : 1000  $\mu\text{m}$ ).

The air gap l had a length of 15 mm. The air in the air gap had a temperature of 24,5°C and a water content of 4,5 g of water/kg of air.

Several spinning tests were carried out, varying distance h of the bundling point of the funnel (boundary between the cylindrical pipe to the funnel itself) to the spinneret surfache in such a way that the relation

$$0,1+0,0051 \leq 0,7 \cdot d_0 \cdot \frac{(h-1)}{h}$$

( $l = 15$  and  $d_0 = 1000$ ) was fulfilled, while air gap  $l$  remained unchanged. In each test, the highest achievable final drawing, i.e. the maximum drawing rate of the filaments at breaking of the fibre, was measured. The results are shown in Table 1:

| TABLE 1 |                |                          |
|---------|----------------|--------------------------|
| h (mm)  | Angle $\alpha$ | final drawing<br>(m/min) |
| 240     | 16,8°          | 43                       |
| 190     | 20,9°          | 42                       |
| 140     | 27,4°          | 42                       |
| 90      | 38,8°          | 41                       |
| 70      | 46,0°          | 29                       |
| 40      | 61,1°          | 0                        |

From Table 1 it can be seen that up to an angle of about 40° no reduction of the final drawing rate und thus no deterioration of spinnability can be observed. From an angle of 45° onwards however, the maximum final drawing rate is significantly reduced. At an angle of about 61°, the solution is no more spinnable.

#### Example 2

A spinning device corresponding to Figure 2 was used, and again as the spinneret the one described schematically in WO 95/04173 (number of holes: 28 392; hole diameter: 100  $\mu\text{m}$ ; external diameter of the nozzle (outermost row of holes)  $d_1$ : 155 mm; hole/hole-distance  $d_0$ : 500  $\mu\text{m}$ ) was used.

The cellulose solution used contained 13,5% of cellulose and had a temperature of 120°C. The output was 0,025 g/min. The air gap  $l$  had a length of 20 mm. The air in the air gap had a

-11-

temperature of 12°C and a water content of 5 g of water/kg of air.

The filaments were diverted at a cylindrical, non-rotatable bar 2 and drawn diagonally upwards from the spinning bath.

While air gap 1 remained unchanged, distance h again was varied, and the maximum final drawing rate and angle  $\alpha$  were determined. The results are given in Table 2.

| TABLE 2 |                |                          |
|---------|----------------|--------------------------|
| h (mm)  | Angle $\alpha$ | final drawing<br>(m/min) |
| 345     | 13°            | 18                       |
| 165     | 25°            | 18                       |
| 115     | 34°            | 18                       |
| 75      | 46°            | 4                        |

As can be seen from Table 2, when changing the angle  $\alpha$  in a range of from 13° to 34°, the maximum final drawing rate will not be reduced. However, when increasing the angle  $\alpha$  to 46°, the final drawing rate, i.e. the spinnability, is drastically reduced. When distance h is further reduced (and thus the angle  $\alpha$  increased), the solution is no more spinnable.

### Example 3

The same spinning device as described in Example 2 was used, the air gap 1 however was held constant at 30 mm. Distance h was again varied. The spinning safety of the solution under the given conditions was determined according to the occurrence of spinning deficiencies (filament break, extreme adhesion of the filaments among each other).

The spinning safety is high when in a period of time of more than 15 minutes practically no spinning deficiencies will occur. When spinning deficiencies abound within a period of 15 minutes or even before, spinning at an industrial scale is

-12-

only possible when continuous technical assistance is provided.

Subsequently, the spinning safety is defined by a time indication. In the following Table 3, the indication "> 15 min" means that spinnability was good (practically no spinning deficiencies within 15 minutes). An indication of e.g. "< 10 min" means that already in less than 10 minutes after spinning start, massive spinning deficiencies which require an interruption of spinning will occur.

| TABLE 3 |                |                 |
|---------|----------------|-----------------|
| h (mm)  | Angle $\alpha$ | Spinning safety |
| 345     | 13°            | > 15 min        |
| 165     | 25°            | > 15 min        |
| 115     | 34°            | > 15 min        |
| 100     | 38°            | 10-15 min       |
| 85      | 42°            | < 10 min        |

From Table 3 it can be seen that up to a distance h of 115 mm, the spinnability is good. However, when h is chosen still smaller, the relation defined according to the invention is no more fulfilled and spinnability will deteriorate drastically. This is the case for the last two tests. This deterioration of the spinning behaviour occurs in the present Example already at an angle  $\alpha$  clearly below 45°.

#### Example 4

In a pilot plant for the production of cellulose fibres according to the amine-oxide process, the way the filaments are diverted in the spinning bath in the spinning device according to the invention was examined in numerous individual tests.

Rotationally symmetrical rotatable diverting means of many designs (rolls including glass sticks having a smooth or ribbed surface) were tested. In these tests, it was

-13-

repeatedly found out that, as soon as the diverting means rotates around its own axis, filaments will wrap around the diverting roll within a short period of time. The reason for this wrapping around evidently is that in the spinning bath, fibres sometimes will break, are collected by the rotating diverting roll, dragged along by the diverting roll, and will lead to an increased wrapping around by dragging along other filaments. In this process, the spun filaments are deteriorated, since the filaments wrapped around the diverting means have to be removed by means of a mechanical intervention that implies a deterioration of the final product.

It has been shown that when using a rotatable diverting roll, the spinning process has to be interrupted in a period of time of less than 30 minutes to remove the fibres wrapped around the diverting means.

When rotation of the diverting means is impeded by e.g. designing it as a rigid means while maintaining the other parameters, practically no wrapping around will occur. It has been shown that in this way, a continuous spinning process can be maintained for several hours. Thus the use of rotatable diverting means has to be avoided. To guarantee a satisfactory operation, it is necessary to design, if possible, all diverting means as non-rotatable means.

## CLAIMS:



1. A spinning device for carrying out the amine-oxide process according to the dry/wet-spinning process comprising

- a spinneret having spinning holes for extruding filaments,
- a blowing device whereby said extruded filaments may be cooled immediately after being delivered from the spinning holes,
- a container containing spinning bath liquid,
- a bundling means provided in said spinning bath liquid for bundling said extruded filaments, and
- an air gap defined as distance of said spinneret to the surface of said spinning bath liquid,

characterized in that

- said bundling means is located at such a distance from said spinneret that the angle ( $\alpha$ ) formed by the filaments with respect to the vertical to the surface of said spinning bath liquid does not exceed  $45^\circ$  and
- that the relation

$$0,1+0,0051 \leq 0,7 \cdot d_0 \cdot \frac{(h-1)}{h}$$

is fulfilled, wherein  $d_0$  is the distance (mm) between a spinning hole and its neighbouring spinning hole on said spinneret,  $h$  is the distance (mm) of said bundling means to said spinneret and  $1$  is said air gap (mm), and wherein

$$0,4 \text{ mm} \leq d_0 \leq 2 \text{ mm} \text{ and} \\ 0 \text{ mm} < 1 < 60 \text{ mm}.$$



2. A spinning device according to Claim 1, characterized in that said bundling means (2) is designed as a diverting means (2) whereon the filaments (4; 5) are not only bundled but also diverted.
3. A spinning device according to Claim 2, characterized in that said diverting means (2) is designed such that it does not rotate when diverting the filaments (4; 5).
4. A spinning device according to one of the Claims 1 to 3, characterized in that said angle ( $\alpha$ ) does not exceed 20°.
5. A spinning device for carrying out the amine-oxide process according to the dry/wet-spinning process comprising
  - a spinneret having spinning holes for extruding filaments,
  - a blowing device whereby the extruded filaments may be cooled immediately after being delivered from the spinning holes,
  - a container containing spinning bath liquid,
  - a diverting means provided in the spinning bath liquid for bundling and diverting the extruded filaments, and
  - an air gap defined as distance of the spinneret to the spinning bath liquid,characterized in that said diverting means (2) is designed such that it will not rotate when diverting the filaments (4; 5).
6. A spinning device according to one of the Claims 1 to 5, characterized in that said spinneret comprises:
  - a substantially rotationally symmetrical nozzle body having in its center a feeding for cooling gas,
  - a feeding for the cellulose solution,
  - an annular spinning insert having spinning holes and



-16-

- a baffle plate for diverting the cooling gas flow towards the filaments which are extruded from the spinning holes, so that the cooling gas stream strikes the filaments essentially at right angles.
7. A spinning device according to one of the Claims 1 to 6, characterized in that said container containing the spinning bath liquid is connected to a lifting device whereby said container can be moved towards and away from the spinneret in vertical direction, distance  $l$  thus being varied, and that said bundling means is arranged such that the distance  $h$  remains constant inspite of this movement.