April 29, 1958 K. RUTHARDT 2,832,710

METHOD FOR CARBURIZING SPINNING NOZZLES COMPOSED OF TANTALUM AND ALLOYS THEREOF

Filed March 1, 1956 2 Sheets-Sheet 1

Fig. 1.

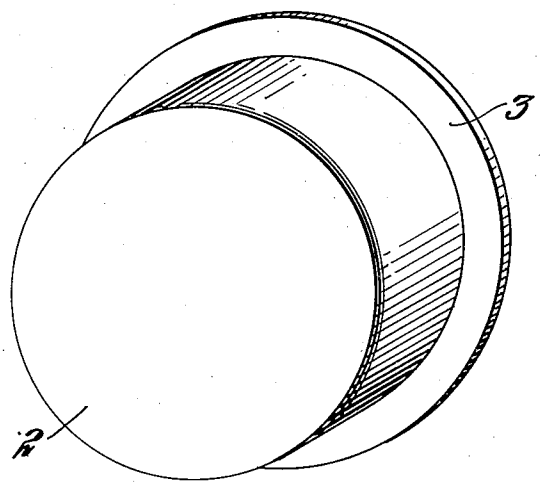
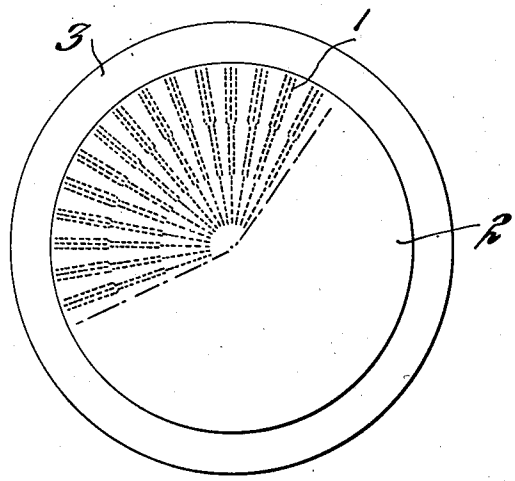


Fig. 2.



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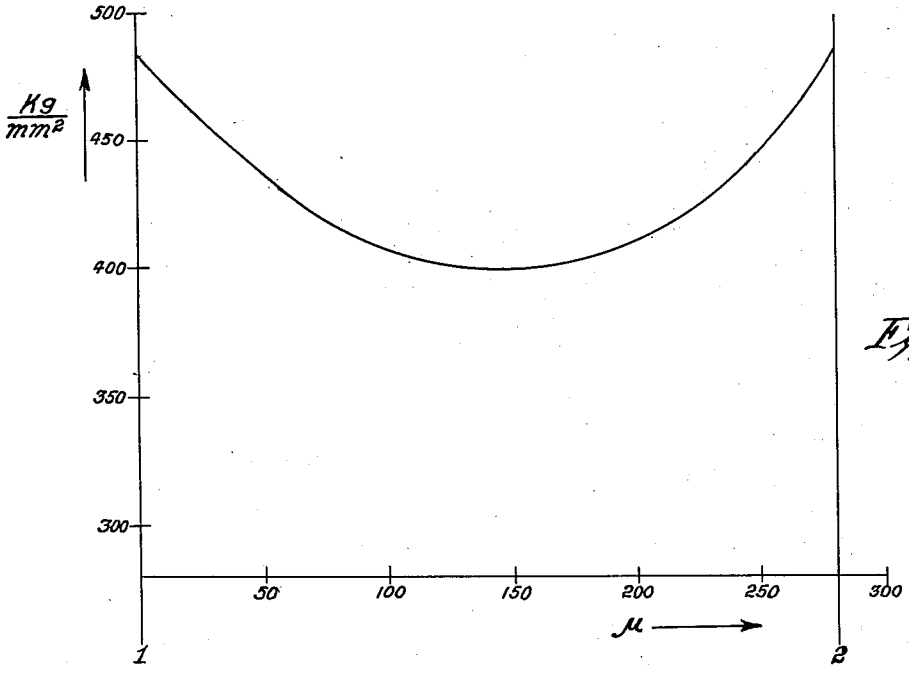


Fig. 3.

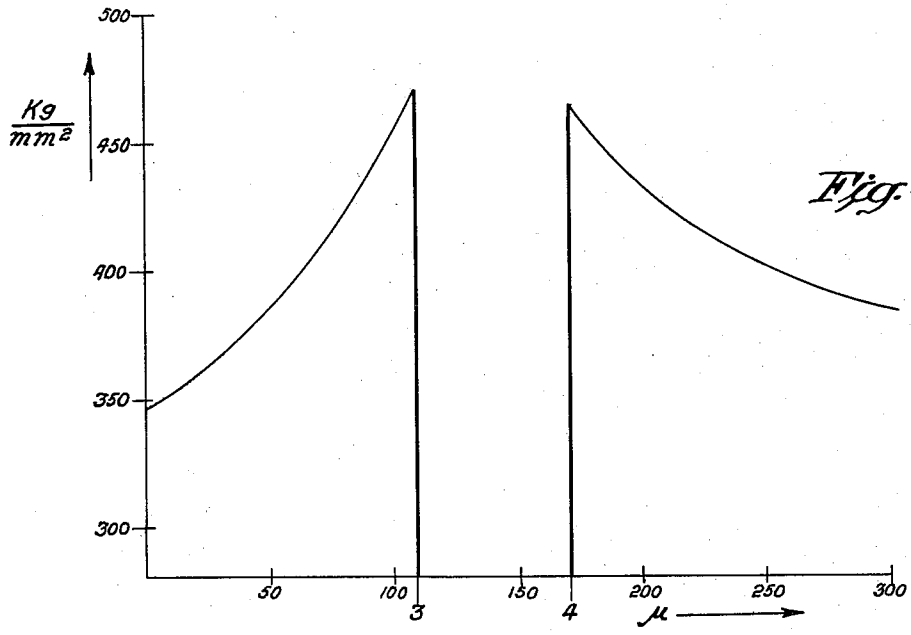


Fig. 4.

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United States Patent Office 2,832,710 Patented Apr. 29, 1958

2,832,710

METHOD FOR CARBURIZING SPINNING NOZZLES COMPOSED OF TANTALUM AND ALLOYS THEREOF

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Application March 1, 1956, Serial No. 568,831

5 Claims. (Cl. 148—13.1)

The present invention relates to spinning nozzles for the production of synthetic filaments. It is primarily concerned with spinning nozzles for the manufacture of rayon and viscose wool, while, however, spinning nozzles for the manufacture of synthetic filaments of other materials are not excluded. More specifically the invention relates to spinning nozzles made of tantalum, and has for an object to improve the qualities of the material constituting the nozzles.

This application is a continuation-in-part of application Serial No. 240,774, filed August 7, 1951, now abandoned.

Tantalum, owing to its high chemical resistance, is widely used for the manufacture of spinning nozzles. A disadvantage, however, consists in the fact that tantalum is relatively soft, and that it is difficult to harden this metal without having to contend with other considerable disadvantages. Thus for example, alloying only has led to brittle alloys, and those alloys which retain the characterizing desirable qualities of tantalum are unsuitable for being shaped by cutting tools. Out of various methods for surface hardening tantalum, only hardening by oxidation has hitherto found entrance into practice; this method, however, is unsuitable for many purposes and, to say the least, reduces the utility value of the products because the material loses its glossy metallic surface. Thus for example, in one known method the completed tantalum spinning nozzles are heated for a short time in air, oxygen, nitrogen or carbon monoxide, at temperatures between 300 and 600° C. In this manner a surface hardening is obtained at the price of the disadvantage that the tantalum becomes discoloured and its surface loses the gloss. Hereby the quality of the spinning nozzles is impaired to such an extent as to cause them to get rapidly blocked during the spinning.

For this reason various suggestions have already been made aiming at hardening tantalum in such a manner as to retain a glossy surface. With this object in view, the annealing has, for example, been effected in air or oxygen under reduced pressure. It is true that in this manner hardening is obtained without the tantalum becoming dark; however, an increase in brittleness occurs at the same time, the tantalum thus hardened being apt to crack and being quite unsuitable as material for spinning nozzles. Endeavours to carry out the hardening with nitrogen under reduced pressure have likewise not yielded satisfactory results. Although these endeavors have succeeded in obtaining a bright, glossy, tough, hard and non-brittle tantalum, the tantalum obtained has the disadvantage that the nitride formed during the hardening reacts with the lye of the spinning solution—presumably with formation of hydrides—and owing to this in practical use the originally tough tantalum becomes so brittle as to crack after some time.

It is therefore a further object of the present invention to produce spinning nozzles of a tough and hard tantalum which is free from the disadvantages referred

to, that is to say which has as light a colour as possible, and which is resistant to chemical attack.

With this and other objects in view, the present invention, in one aspect, consists in partly converting the tantalum from which the spinning nozzle is made into tantalum carbide. In another aspect the invention consists in a spinning nozzle consisting of tantalum which contains tantalum carbide, at least in its surface layer.

The transformation of the surface layer into tantalum carbide may be so conducted that the said layer consists wholly or preferably so that said layer consists partly of tantalum carbide.

While it is generally sufficient for attaining the main object of the invention to incorporate tantalum carbide in the surface layer, the greatest improvement is obtained when the tantalum carbide extends througout the cross-section of the spinning nozzle with the content either decreasing gradually towards the interior or extending throughout the cross-section in almost equal magnitude.

The tantalum carbide contents of the whole tantalum spinning nozzle should be so high as to correspond to an overall carbon content of 0.01 to 5%, and preferably to an overall carbon content of 0.05 to 0.5%.

Generally the tantalum carbide content is paralleled by a content of tantalum oxide in quantities up to the indicated limits for the tantalum carbide content.

A spinning nozzle according to the invention may have the form of any of the numerous spinning nozzles generally used in the art and described in literature, for example that of the well-known cap or dish-like nozzles having a narrow edge at the upper end. The spinning apertures may be distributed in the bottom of the nozzle in any of a great variety of manners; they may, for example, be arranged in rows extending radially of the bottom of the spinning nozzle. In a preferred form of the method according to the invention, the conversion of the surface layers into tantalum carbide extends not only to the coherent surface of the spinning cap, but also to the internal surfaces of the fine spinning passages.

Spinning nozzles which, in accordance with this invention, have a content of tantalum carbide are distinguished by great hardness. While the hardness of tantalum is normally, for example, 100 kg./mm.^{2}, the micro-hardness of the surface layers of the novel spinning nozzles after the partial conversion of the tantalum into tantalum carbide is 200–600 kg./mm.^{2}, and preferably 300–500 kg./mm.^{2}.

When, in accordance with the preferred form of the invention the partial conversion of the tantalum into tantalum carbide is also effected into the interior of the cross-section of the spinning nozzle, an increase of the micro-hardness can there be likewise ascertained, the hardness either decreasing somewhat towards the interior or being nearly constant throughout the cross-section according to the degree of conversion into tantalum carbide.

In order that the invention may be more readily understood, reference will now be made to the accompanying drawings, in which:

Fig. 1 is an enlarged perspective view of a spinning nozzle according to one form of the invention;

Fig. 2 is an end view, the arrangement of the spinning passages being indicated on a portion only of the bottom surface;

Fig. 3 is a diagram showing the variations of the micro-hardness across the thickness of the bottom, and

Fig. 4 is a similar diagram showing the variations of the micro-hardness along a line extending inside the bottom, parallel to the bottom surface and extending radially of one of the spinning passages.

Referring now to the drawings, a tantalum spinning

nozzle of the usual form is illustrated in Fig. 1 in an enlarged perspective view. In Fig. 2 the distribution of the spinning passages on part of the bottom of the spinning nozzle is indicated by way of example, although obviously the shape of the spinning nozzle and the distribution of the spinning passages are of no importance for the nature of the present invention. The production of the raw material and the shaping of the nozzle may be effected according to the normal practice.

This spinning nozzle may originally, for example, have a hardness of 100 kg./mm.^{2}. According to the invention the hardness is increased to preferably between 400 and 500 kg./mm.^{2} by partial conversion of the tantalum into tantalum carbide. The distribution of hardness in the interior of the spinning nozzle may, for example, be as shown in Figs. 3 and 4. In Fig. 3, the micro-hardness is plotted as a function of the distance from the inner bottom surface of the spinning nozzle throughout the cross-section of the spinning nozzle.

In the drawing the micro-hardness values at the inner surface of the nozzle bottom are indicated at 1 and those at the outer surface of the bottom are indicated at 2. It will be seen that the hardness gradually decreases from a value of 480 kg./mm.^{2} at the two surfaces to about 400 kg./mm.^{2} in the interior of the nozzle. These hardness values have been obtained with the Zeiss micro-hardness tester.

It is important that the high hardness values found at the surface should also be present in the interior surfaces of the spinning passages. Fig. 4 shows the micro-hardness values as found in the neighborhood of a passage in an interior layer of the nozzle extending parallel to the bottom of the nozzle, the micro-hardness values kg. per square millimeter being plotted as a function of the distance measured in μ. It will be seen that the hardness values are generally of the order of 350 kg./mm.^{2} and increase at the surface of a passage 3, 4 to values between 450 and 500 kg./mm.^{2}.

The high values of the micro-hardness, more particularly on the surface of the spinning passages, are of importance since they are responsible for the unusual resistance which the spinning nozzles according to the invention have been found to possess to grinding action, for example that caused in the case of matt rayon by pigments such as titanium dioxide present in the spinning substance.

Owing to their tantalum carbide content the spinning nozzles according to the invention also show considerably increased Vickers hardness, hardness values of 150 to 400 kg./mm.^{2} having been observed. This increase of the Vickers hardness in conjunction with the abovementioned increase of the micro-hardness results in a substantial improvement of the mechanical strength of the nozzle body as compared with previously known tantalum spinning nozzles.

Although possessing these high hardness values, spinning nozzles according to the invention have no tendency of becoming brittle. They also are highly resistant to chemical attack, and more particularly are not appreciably affected either by acid or alkaline mediums. The surface of the novel spinning nozzles shows a metallic gloss; when the tantalum-carbide content is low, the surface shows the light colour of pure tantalum while in the case of higher tantalum carbide contents it has a golden yellow metallic gloss.

Mainly due to their great hardness, the novel spinning nozzles have a long life and give excellent spinning results; the spinning passages do not become clogged even after long use.

The spinning passages are produced before the partial conversion of the tantalum into tantalum carbide, thus obtaining the advantage that the spinning passages may be readily produced true to measure while the metal is soft, whereafter the hardness of the completed spinning nozzle is increased to a value which in practice would only with the greatest difficulty permit a drilling of spinning passages.

Consequently, a spinnerette blank is first formed in the form of a disc, plate or cup, etc., from tantalum in a substantially pure state, or from an alloy of tantalum wherein the tantalum constitutes the major portion of the alloy. Thereafter spinnerette holes are drilled or otherwise formed through the said metal blank and subjected to treatment with carbonaceous gases as hereinafter more particularly described, whereby a hard glossy surface is imparted to the spinning passages or holes, said passage walls being very smooth and free of such deposits of carbon particles or partially converted tantalum irregularities which would tend to interfere with the passage of a spinning mass therethrough.

A number of methods are available for the partial conversion of the tantalum spinning nozzles produced by customary methods into tantalum carbide.

Treatment of tantalum in carbon monoxide between 300 and 600° C. leads in the first place to the formation of tantalum oxide. On the other hand one readily succeeds in producing tantalum carbide if the treatment is effected at higher temperatures, preferably between 700 and 1500° C., and at reduced pressures, preferably between 0.001 and 100 mm. Hg, and more particularly between 0.001 and 1 mm. Hg with carbonaceous gases which should not contain any elementary oxygen. Suitable gases are, for example, methane or carbon monoxide.

With treatment of a short duration in the above-described manner only the surfaces of the nozzles are partly converted into tantalum carbide and thus reach the indicated high hardness values.

If the treatment is extended for a longer period, the interior of the cross-section of the nozzle is, mainly due to diffusion, likewise partly converted into a tantalum carbide. In order to obtain this invaluable condition, a longer duration of the annealing treatment at temperatures between 700 and 1500° C. is recommended. By extending the annealing treatment over a sufficiently long period, the tantalum carbide content may even be caused to be more or less uniformly distributed throughout the cross-section. In general, however, a distribution of hardness approximately corresponding to that shown in Figs. 3 and 4 will be found sufficient.

If the tantalum subjected to the above-described partial conversion into tantalum carbide, in the known manner originally contains tantalum oxide in addition to metallic tantalum, the ultimate product will contain tantalum oxide in addition to tantalum carbide without interfering with the nature of the invention. The object of the invention is also retained when during the conversion of tantalum into tantalum carbide, tantalum oxide is also formed in a certain proportion, or when the tantalum obtains from its surface a certain oxide content by contact with the air.

The following are examples for the above-described partial conversion of the tantalum into tantalum carbide:

I. A tantalum spinning nozzle is treated for sixty minutes in a carbon monoxide atmosphere of 0.01 Torr. mm. Hg at 1000° C. The spinning nozzle obtains a very high surface hardness while retaining its metallic gloss; the Vickers hardness in this treatment already amounts to 250 kg./mm.^{2}. As mentioned above, it is of advantage to provide the spinning passages already prior to the treatment with carbon monoxide because the nozzle is then still in a soft condition, and also because if the invention is carried out in this manner, the surface of the spinning passages will also obtain a hard, chemically resistant carbide coating.

II. Similar results are obtained if the spinning nozzle is annealed for approximately thirty minutes at a temperature of 1200° C. in carbon monoxide as free as possible from oxygen at a pressure of 0.005 mm. Hg.

The spinning nozzles obtained are resistant to chemical

attack both by the spinning solution and by the precipitating liquid.

What I claim is:

1. A method of making spinning nozzles comprising forming orifices through a spinning blank composed of metal taken from the group consisting of substantially pure tantalum and alloys thereof wherein the tantalum constitutes a major portion of the alloy, and thereafter treating the spinning nozzle with carbonaceous gases at temperatures between 700° C. and 1500° C. and pressures of 0.001 to 100 mm. Hg.

2. A method of making spinning nozzles comprising forming orifices through a spinning blank composed of metal taken from the group consisting of substantially pure tantalum and alloys thereof wherein the tantalum constitutes a major portion of the alloy, and thereafter treating the spinning nozzle with carbonaceous gases at temperatures of 700° C.–1500° C. and pressures of 0.01 to 1 mm. Hg.

3. A method of making spinning nozzles comprising forming orifices through a spinning blank composed of metal taken from the group consisting of substantially pure tantalum and alloys thereof wherein the tantalum constitutes a major portion of the alloy, and thereafter treating the spinning nozzle with methane at temperatures between 700° C. and 1500° C. and pressures of 0.01 to 1 mm. Hg.

4. A method of making spinning nozzles comprising forming orifices through a spinning blank composed of metal taken from the group consisting of substantially pure tantalum and alloys thereof wherein the tantalum constitutes a major portion of the alloy, and thereafter treating the spinning nozzle with carbon monoxide at temperatures between 700° C. and 1500° C. and pressures of 0.01 to 1 mm. Hg.

5. A method of making spinning nozzles comprising forming orifices through a spinning blank composed of metal taken from the group consisting of substantially pure tantalum and alloys thereof wherein the tantalum constitutes a major portion of the alloy, and thereafter treating the spinning nozzle with carbonaceous gases at temperatures between 700° C. and 1500° C. and pressures of 0.01 to 1 mm. Hg. to provide the nozzles with a surface layer containing tantalum carbide, and subjecting the nozzles to annealing treatment until the tantalum carbide content is distributed throughout the cross section of the nozzle.

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