Fig. 1

![Graph showing the rate of solvent removal on the first Gudet.](image)

Fig. 2a

Fig. 2b
Fig. 3
HIGH SPEED SPINNING METHOD OF VISCOSE RAYON FILAMENTS HAVING HIGH WET MODULUS

Yoichi Uchida and Keniti Hukuma, Mihara-shi, Japan, assignors to Teijin Limited, Osaka, Japan, a corporation of Japan

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ABSTRACT OF THE DISCLOSURE

Method of spinning at high speed of viscose rayon filaments having a high wet modulus, characterized by heating a viscose having a high degree of polymerization and a high gamma value to 35–60° C. immediately prior to the spinning of the said viscose, extruding the said viscose into a spinning bath, and rendering the rate of solvent removal of the filaments on the first godet from 40 to 60%, and thereafter stretching said filaments the spinning being effected at a spinning speed of at least 80 meters per minute using a horizontal type spinning machine.

This invention relates to a high speed spinning method of viscose rayon filaments having high wet modulus, and its object resides in spinning with a horizontal type spinning machine at a spinning speed of at least 80 meters per minute, viscose rayon filaments having high wet modulus as well as excellent touch and good luster.

Hereinafter, as a method of producing staple fibers of the polyonotic type having high wet modulus, the method, for example, of Japanese patent publication No. 574/1954 (corresponding to French Pat. No. 1,111,571, published Mar. 1, 1956) is being practiced. However, its spinning speed, as compared with that of the conventional regular rayon, is an exceedingly low speed, for example, such as 10 meters to 20 meters per minute. Hence, since an enhancement of productivity cannot be attained, it is not suitable for the production of filaments. Accordingly, as a method of spinning high wet modulus rayon filaments at high speeds, a vertical tubular method of spinning such as that, for example, of Belgian Pat. 665,439 (1964) has been proposed, but since its spinning mechanism is highly complicated, it is beset with many difficulties from the operational standpoint.

As a result of our assiduous researches with a view to elimination of the aforementioned drawbacks, we were able to achieve this invention. Accordingly, this invention is directed to a method of spinning at high speeds viscose rayon filaments having high wet modulus and is characterized in that viscose having a degree of polymerization of 400–700 and a gamma value at the time of spinning of 65–80, and in which the relationship between the average degree of polymerization, cellulose content and total alkali content satisfies the following Formulae 1 and 2:

\[
\begin{align*}
(1) \quad & 9.5 \times 10^5 - DP\text{C} < CC \times 10^3 < 1.3 \times 10^5 - 1.3DP \\
(2) \quad & 200 + DP\text{C} < \frac{TA}{CC} \times 10^3 < 450 + DP
\end{align*}
\]

where:

- \(DP\text{C}\) is the average degree of polymerization of the viscose,
- \(CC\) is the cellulose content of the viscose in percent,
- \(TA\) is the total alkali content of the viscose in percent,
- is heated at 35–60° C. immediately prior to spinning, following which the viscose is extruded into a spinning bath containing sulfuric acid of low concentration and sulfates of low concentration to render the rate of solvent removal of the filaments on the first godet from 40% to 60%, and thereafter the filaments are stretched, the spinning being carried out at a spinning speed of at least 80 meters per minute using a horizontal type spinning machine.

The invention will be more fully described below with reference to the accompanying drawings, wherein:

FIG. 1 illustrates one instance of the relationship between the rate of solvent removal of the filaments on the first godet and the fiber properties;

FIGS. 2–a and 2–b illustrate the zones of the average degree of polymerization, cellulose content and total alkali content of the viscose used in this invention; and

FIG. 3 illustrates one instance of the relationship between the heating temperature immediately prior to spinning, the rate of solvent removal on the first godet, spinability and stretchability.

In the past, the main point of the technical improvement in the method of spinning viscose rayon concerned the control of the state of coagulation and regeneration of the viscose.

Viscose consists of cellulose xanthate dissolved in an alkali solution, and the fiber-forming process is a solvent removal diffusion with reaction from cellulose xanthate to cellulose.

The investigations concerning the mechanism of spinning cellulose have been directed mainly towards the manner of coagulation and regeneration of the cellulose.

Having perceived that the rate of solvent removal during the early stages of spinning had essentially a greater influence on the properties of resulting fibers rather than the matter of coagulation and regeneration, and as a result of numerous experiments that we carried out varying the several factors such as the viscose composition, degree of polymerization and gamma value of the viscose, the temperature at which the viscose was heated and the composition of the spinning bath, it was found that the best results, i.e. the optimum fiber properties which were intended to be attained by high speed spinning, could be obtained when the rate of the solvent removal of the filaments on the first godet was from 40% to 60%.

The results of one instance of the foregoing experiments is graphically illustrated by means of FIG. 1. In the figure, the curve (a) drawn with the dotted line, represents the dry elongation (percent), the curve (b) drawn with a solid line, represented the dry tensile strength (d/De), and the curve (c) drawn with the dot-and-dash line represents the wet tensile strength at 5% elongation. These results were obtained by carrying out the experiments in which the total alkali content of the viscose was varied from 3.0% to 7.0% while holding the other factors constant. The graph shows the relationship between the fiber properties obtained and the rate of solvent removal of the filaments on the first godet. As is apparent from the figure, the fiber properties obtained when the rate of solvent removal was 40–60% are indicated by pronounced peaks and hence were very desirable. The experiments conducted were not limited to those regarding the total alkali content but experiments relative to the other factors were also conducted, with entirely similar tendencies being noted. The specific numerical values will be more fully described in the hereinafter given examples.

The term "rate of solvent removal," as herein used, denotes the rate at which the solvent (the aqueous solution of alkali) is driven out of the viscose and is a ratio of the amount of the solvent removed during the spinning step to the amount of the solvent of the original viscose, expressed as a weight percentage.

Letting the cellulose content of the viscose be \(C_1\)%, the cellulose content of the filament during the spinning step, \(C_2\)% and the rate of solvent removal, \(R\)% then the
3,539,678

relationship shown by Formula 3 will hold as a matter of course.

\[
(C_2 - C_1) \times 10^4 \%
\]

Now, the cellulose content of the viscose, \(C_1\), is determined in customary manner, while the cellulose content during the spinning step, \(C_2\), is determined in the following manner. About 2 grams of gel filament collected during the step in which the measurement is to be made are dipped in an ice-cooled decomposition stopping solution for about 30 minutes, then subjected to centrifuging for 10 minutes to eliminate the excess liquid and thereafter accurately weighed to obtain the gel weight (A gram).

Next, this specimen is dipped for 10 minutes in an aqueous sulfuric acid solution of a concentration 100 g/l to carry out the regeneration completely. This is followed by washing with water, drying for 3 hours at 100-105° C, and thereafter accurately weighing the product to obtain the weight (B gram). \(C_2\) % is then calculated by means of the Equation 4.

\[
C_2 = \frac{B}{A} \times 100(\%)
\]

Having obtained \(C_1\) and \(C_2\), as hereinbefore described, the solvent removal rate \(R\) can readily be computed by means of the hereinafove given Equation 3.

When the spinning speed has been merely raised in spinning rayon filaments having high wet modulus, it becomes necessary to raise the sulfuric acid concentration in the spinning bath and in concomitance therewith the rate of solvent removal at the time of stretching rises to above 60%, and hence the intended fiber properties cannot be obtained. Namely, when the solvent removal rate is above 60%, the effects on the filament of orientation by stretching is small and thus satisfactory fiber properties are unobtainable. On the other hand, satisfactory fiber properties also cannot be obtained, as a rule, when the rate of solvent removal is below 40%, since the orientation of the molecular chains is not brought about during the stretching and the strength as well as elongation is low.

For making the rate of solvent on the first godet from 40% to 60% so as to obtain the hoped-for good fiber properties by high speed spinning, the following three conditions must be controlled.

The first condition concerns the composition of the viscose. We conducted numerous experiments concerning the three factors of the average degree of polymerization (DF) of the viscose and the cellulose content (CC) and total alkali content of the viscose, with the consequence that it was found that the hoped-for end could be achieved when the average degree of polymerization ranged between 400 and 700 and the mutual relationships of these three factors were such that they would fall within the zone indicated by means of the oblique lines in FIGS. 2-a and 2-b. If this zone is represented by mathematical expressions, the previously presented Formulae 1 and 2, which correspond to FIGS. 2-a and 2-b, respectively, are obtained.

As can be seen from the figures and the formulae, there is a correlation between the cellulose content and the average degree of polymerization of the viscose, and while the cellulose content can vary within the range of 2.5-7.8%, particularly desirable is a range of 4-6%. On the other hand, there is a correlation between the total alkali content of the viscose and the average degree of polymerization of the viscose, and while the value of TA/CC can vary within the range of 0.6-1.15, particularly to be preferred is a range of 0.7-0.9.

When the average degree of polymerization of the viscose and its cellulose content is higher than the hereinbefore prescribed range, not only does the viscose viscosity rise, but also the coagulation and regeneration speed becomes great to result in the solvent removal rate on the first godet deviating from the range of 40-60%, with the consequence that desirable fiber properties cannot be obtained.

On the other hand, when the average degree of polymerization of the viscose and its cellulose content is lower than the range hereinbefore prescribed, not only does the viscose viscosity fall to a viscosity lower than is suitable for the stretching of the filaments subsequent to their extrusion from the nozzle, but also adverse effects are had on the fiber properties to be obtained. Particularly, when the cellulose content is low, the coagulation and regeneration does not take place smoothly, since the feed of the viscose increases for obtaining filaments of a given denier.

Further, when the alkali content of the viscose exceeds the aforesaid prescribed range, satisfactory spinning cannot be accomplished on account of the insufficiency of the coagulation and regeneration. On the other hand, when the alkali content of the viscose is lower than the foregoing prescribed range, the viscosity of the viscose becomes great to hinder the spinnability of the viscose.

Thus, as hereinafove described, the cellulose and alkali contents of the viscose must be chosen to come within their proper ranges in accordance with the average degree of polymerization of the viscose.

Next, the second condition concerns the ripening of the viscose. It was found that the gamma value of the viscose at the time of its spinning must be in the range of 65-80%.

When the gamma value of the viscose is less than 65, the rate of solvent removal on the first godet becomes less than 40%, whereas when the gamma value is above 80, the rate of solvent removal on the first godet becomes greater than 60%. Both cases, being departures from the range of this invention, result in deterioration of the fiber properties.

On the other hand, the third condition of this invention is the requirement that the viscose be heated at 35-60° C. immediately prior to spinning of the viscose.

In the case of rayon having high wet modulus, the viscosity is relatively high owing to the high degree of polymerization of the viscose, and along with the rise in the spinning speed, the viscose viscosity at the time of the extrusion has a very great effect on the state of spinning.

One instance of an experiment conducted to show specifically the relationship between the heating temperature, spinnability, stretchability and rate of solvent removal is graphically shown in FIG. 3. In the figure, the curve (e) drawn with the dotted line, represents the spinnability (percent), the curve (d) drawn with the solid line represents the stretchability, while the curve (e) drawn with the dot-and-dash line indicates the rate of removal of solvent.

This experiment was conducted under the following conditions.

Namely, the viscose having a average degree of polymerization of 530, a cellulose content of 5.0%, a total alkali content 4.44% and a gamma value of 73 at the time of spinning, extruded from a spinneret having 75 holes, each 0.06 mm in diameter, into a 50° C. spinning bath composed of 5.7% sulfuric acid, 0.02% zinc sulfate and 3.0% sodium sulfate, whose immersion length was 30 cm.

The stretchability, as here used, is a value expressed in percentage of the relative peripheral speed of the second godet to that of the first godet at the time breakage of monofilaments occurs between the first and second godets when the peripheral speed of the first godet is held constant while that of the second godet is raised. The spinnability, on the other hand, is a value expressed in percentage of the ratio of the peripheral speed of the first godet to the linear speed of viscose being extruded from the spinneret at the time breakage of yarn occurs.
between the spinneret and the first godet when the linear speed of the viscose being extruded from the nozzle is held constant while the peripheral speed of the first godet is raised.

According to this invention, the viscose is extruded into a spinning bath containing sulfuric acid of low concentration and sulfates of low concentration while controlling the foregoing conditions. As to the composition of the spinning bath, it is preferred that the concentration of the sulfuric acid is, for example, 4–6%, that of zinc sulfate is, for example, 0.001–0.1% and that of sodium sulfate is, for example, 3–6%. On the other hand, the temperature of the spinning bath is preferably within a range of 30–60°C. A ratio of stretching of, say, 1.2–1.8× is desirable in this invention.

According to this invention, viscose rayon filaments of excellent touch and good luster and having high wet modulus, the production of which, even though possible heretofore, could only be practiced at very low speeds, can be spun at a high speed of such as at least 80 meters per minute, employing a horizontal type spinning machine, for example, the Cephalon type spinning machine. The spinning can moreover be raised readily to as high as, say, about 180 meters per minute, and fibers having fully satisfactory properties can be obtained at such a speed.

This invention is applicable to monofilaments of, say, 0.7–5.0 deniers and a wide range of total deniers of such, for example, 30–1000 deniers. Thus, it is possible to obtain according to this invention even products whose total denier is of a fineness of such as 30 Denier and having moreover serviceable strength.

**Example 1**

Alkaline cellulose obtained by steeping pulp in an alkaline and rending was aged for 2 hours at 25°C, following which 45% of carbon disulfide was added based on the cellulose followed by reacting for 3 hours at 25°C, and thereafter the cellulose xanthate was dissolved in a dilute alkali. Holding the total alkali content at 3.75%, three classes of viscose were then prepared whose cellulose contents were respectively 4.1%, 5.0% (present invention) and 6.0%. A fourth class was also prepared in which the cellulose content was 5.0%, while the total alkali content was 3.24%. The average degree of polymerization of the viscose was 520 in all four instances.

These viscose were filtered and dried, after which the viscose having a gamma value of 75 at the time of spinning were heated at 45°C immediately prior to their spinning and extruded from a spinneret having 75 holes, each 0.06 mm. in diameter, into a 45°C spinning bath composed of 0.025% sulfuric acid, 0.0025% zinc sulfate and 4% sodium sulfate, whose immersion length was 30 cm. to be spun at the spinning speed of 125 meters per minute at a ratio of stretching between the first and second godets of 1.5× to obtain a rayon yarn having a total denier of 75 Denier. The results of the experiments and the properties of the yarn obtained are shown in Table I. The number of breaks as shown in the table is the number of breaks per 24 hours of the spinning process, while the number of fluffs is the number of fluffs in the yarn per 10,000 meters thereof.

**Table I**

<table>
<thead>
<tr>
<th>Experiment No.</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cellulose content of viscose, CC percent</td>
<td>4.1</td>
<td>5.0</td>
<td>5.0</td>
<td>6.0</td>
</tr>
<tr>
<td>Total alkali content of viscose, TA percent</td>
<td>3.75</td>
<td>3.75</td>
<td>3.75</td>
<td>3.75</td>
</tr>
<tr>
<td>T/A, CC</td>
<td>0.91</td>
<td>0.91</td>
<td>0.91</td>
<td>0.91</td>
</tr>
<tr>
<td>Average degree of polymerization of viscose</td>
<td>520</td>
<td>520</td>
<td>520</td>
<td>520</td>
</tr>
<tr>
<td>Rate of breaking of filaments on the first godet, percent</td>
<td>34</td>
<td>51</td>
<td>67</td>
<td>69</td>
</tr>
<tr>
<td>Number of breaks time/24 hrs</td>
<td>0.67</td>
<td>0.64</td>
<td>0.41</td>
<td>0.25</td>
</tr>
<tr>
<td>Number of fluffs of 80,000 m</td>
<td>0.23</td>
<td>0.11</td>
<td>0.13</td>
<td>0.05</td>
</tr>
<tr>
<td>Dry strength, g/dl</td>
<td>2.45</td>
<td>2.05</td>
<td>2.87</td>
<td>2.71</td>
</tr>
<tr>
<td>Wet strength, g/dl</td>
<td>1.50</td>
<td>1.95</td>
<td>1.84</td>
<td>1.59</td>
</tr>
<tr>
<td>Dry elongation, percent</td>
<td>9.0</td>
<td>9.5</td>
<td>9.0</td>
<td>8.0</td>
</tr>
<tr>
<td>Wet elongation, percent</td>
<td>9.4</td>
<td>10.3</td>
<td>9.0</td>
<td>8.0</td>
</tr>
<tr>
<td>Value of swelling, percent</td>
<td>116</td>
<td>121</td>
<td>125</td>
<td>132</td>
</tr>
<tr>
<td>Wet strength at 5% elongation, g/dl</td>
<td>0.48</td>
<td>1.01</td>
<td>0.78</td>
<td>0.70</td>
</tr>
</tbody>
</table>

**Example 2**

Alkaline cellulose obtained by steeping pulp in an alkaline and rendering was ripened for 2 hours, after which 46% of carbon disulfide based on the cellulose was added thereto followed by reacting for 3 hours at 25°C. Three classes of viscose were prepared by dissolving the cellulose xanthate in a dilute alkali, adjusting the cellulose content to a constant value of 4.5% while varying the total alkali contents respectively to 3.15%, 3.42% and 4.3%.

The degree of polymerization of the viscose was 520 in all three instances.

After filtering and desaturing these viscoses having a gamma value of 72 at the time of spinning were heated at 50°C immediately prior to spinning and extruded from a spinneret having 75 holes, each 0.06 mm. in diameter, into a 50°C spinning bath composed of 5.3% sulfuric acid, 0.025% zinc sulfate and 3.0% sodium sulfate, whose immersion length was 30 cm. to be spun at a spinning speed of 125 meters per minute at a ratio of stretching between the first and second godets of 1.35 to obtain a rayon yarn having a total denier of 75 Denier. The results are shown in Table II.

**Table II**

<table>
<thead>
<tr>
<th>Experiment No.</th>
<th>1</th>
<th>2</th>
<th>3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cellulose content of viscose, CC percent</td>
<td>4.4</td>
<td>4.5</td>
<td>4.5</td>
</tr>
<tr>
<td>Total alkali content of viscose, TA percent</td>
<td>3.15</td>
<td>3.42</td>
<td>4.10</td>
</tr>
<tr>
<td>T/A, CC</td>
<td>0.79</td>
<td>0.76</td>
<td>1.00</td>
</tr>
<tr>
<td>Degree of polymerization of viscose</td>
<td>520</td>
<td>520</td>
<td>520</td>
</tr>
<tr>
<td>Ratio of solvent removal on filaments on the first godet</td>
<td>72</td>
<td>98</td>
<td>98</td>
</tr>
<tr>
<td>Number of breaks time/24 hrs</td>
<td>1.79</td>
<td>0.96</td>
<td>0.52</td>
</tr>
<tr>
<td>Number of fluffs of 80,000 m</td>
<td>25.3</td>
<td>20.2</td>
<td>14.5</td>
</tr>
<tr>
<td>Dry strength, g/dl</td>
<td>2.35</td>
<td>2.54</td>
<td>2.36</td>
</tr>
<tr>
<td>Wet strength, g/dl</td>
<td>1.25</td>
<td>1.25</td>
<td>1.25</td>
</tr>
<tr>
<td>Dry elongation, percent</td>
<td>8.0</td>
<td>9.1</td>
<td>8.2</td>
</tr>
<tr>
<td>Wet elongation, percent</td>
<td>0.4</td>
<td>0.5</td>
<td>0.7</td>
</tr>
<tr>
<td>Value of swelling</td>
<td>119</td>
<td>106</td>
<td>128</td>
</tr>
<tr>
<td>Wet strength at 5% elongation</td>
<td>0.88</td>
<td>1.00</td>
<td>0.85</td>
</tr>
</tbody>
</table>

**Example 3**

Viscose having a cellulose content of 5.0%, a total alkali content of 3.7% and a degree of polymerization of 520, as obtained in Example 1, was varied with respect to its degree of ripening (gamma value) and spun under identical conditions as in the aforesaid Example 1 with the results shown in Table III.

**Table III**

<table>
<thead>
<tr>
<th>Experiment No.</th>
<th>1</th>
<th>2</th>
<th>3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gamma value of viscose</td>
<td>86</td>
<td>79</td>
<td>65</td>
</tr>
<tr>
<td>Rate of solvent removal, percent</td>
<td>70</td>
<td>54</td>
<td>40</td>
</tr>
<tr>
<td>Number of breaks time/24 hrs</td>
<td>0.62</td>
<td>0.04</td>
<td>0.45</td>
</tr>
<tr>
<td>Number of fluffs of 80,000 m</td>
<td>3.00</td>
<td>1.20</td>
<td>1.00</td>
</tr>
<tr>
<td>Dry strength, g/dl</td>
<td>2.78</td>
<td>2.00</td>
<td>1.90</td>
</tr>
<tr>
<td>Wet strength, g/dl</td>
<td>1.55</td>
<td>1.55</td>
<td>1.55</td>
</tr>
<tr>
<td>Dry elongation, percent</td>
<td>8.3</td>
<td>8.2</td>
<td>8.0</td>
</tr>
<tr>
<td>Wet elongation, percent</td>
<td>9.9</td>
<td>10.1</td>
<td>9.1</td>
</tr>
<tr>
<td>Value of swelling, percent</td>
<td>97</td>
<td>98</td>
<td>96</td>
</tr>
<tr>
<td>Wet strength at 5% elongation, g/dl</td>
<td>0.91</td>
<td>1.06</td>
<td>1.02</td>
</tr>
</tbody>
</table>

Thus, it is seen that the state of spinning according to this invention and the properties of the yarn obtained were excellent.

**Example 4**

Viscose having a cellulose content of 4.5%, a total alkali content of 3.4% and a degree of polymerization of 520, as obtained in Example 2, was varied with respect to its degree of ripening (gamma value) and spun under identical conditions as in said Example 2 with the results shown in Table IV.
Thus, it is apparent that the state of spinning according to this invention and the properties of the resulting yarn are very excellent.

EXAMPLE 5

Viscose having a cellulose content of 5.0%, a total alkali content of 3.75% and a degree of polymerization of 520, as obtained in Example 1, immediately prior to its being spun with a horizontal Topham type spinning machine, was heated at 45°C. (present invention), 35°C. (present invention), or not heated, following which the spinning was carried out at the speeds of 125, 150 and 170 meters per minute. The properties of the yarn so obtained are shown in Table V. In this case, excepting for the sulfuric acid concentration of the spinning bath, the spinning conditions were otherwise identical to those of Example 1. The sulfuric acid concentrations were as indicated in Table VI.

Thus, it is seen that satisfactory fiber properties can be obtained even at such high speeds as 170 meters per minute by heating the specified viscose immediately prior to its spinning.

EXAMPLE 6

Viscose having a cellulose content of 4.5%, a total alkali content of 3.42% and a degree of polymerization of 520 as obtained in Example 2, immediately prior to its being spun with a horizontal Topham type spinning machine, was heated at 50°C, 35°C., or not heated, following which the spinning was carried out at the spinning speeds indicated in Table VI. The properties of the so obtained yarn are shown in said Table VI. In this case, excepting for the sulfuric acid concentration, the spinning conditions were otherwise identical to those of Example 2. The sulfuric acid concentrations were as indicated in Table VI.

Thus, it is clear that satisfactory fiber properties can be obtained even at such high spinning speeds as 175 meters per minute by heating the invention specified viscose immediately prior to its spinning.

We claim:

1. A method of spinning viscose rayon filaments having a high wet modulus at high speeds which comprises heating of viscose at 35–60°C. The viscose having a degree of polymerization ranging between 400 and 700 and a gamma value at the time of spinning of 65–80, and in which the relationship between the degree of polymerization, cellulose content and total alkali content of said viscose satisfies the following Formulae 1 and 2

\[ 9.5 \times 10^{-2} \times 7 \times 10^{1/4 - 3} \times 10^{-1} / 1.3 \times 10^{-7} \times 12DP \]

\[ 200 + DP \times T - A \times 10^{-1} < 450 + DP \]

wherein DP is the average degree of polymerization, C is the cellulose content of the viscose in percent and TA is the total alkali content of the viscose in percent.
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ning being effected at a spinning speed of at least 80
meters per minute using a horizontal type spinning ma-
chine.

2. The method according to claim 1 wherein the
stretching effected using a ratio of stretching of 1.2–1.8×.

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JULIUS FROME, Primary Examiner
J. H. WOO, Assistant Examiner

U.S. Cl. X.R.