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3,704,144

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FLAME RETARDANT RAYON COMPOSITION
AND PROCESS

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No Drawing. Continuation-in-part of application Ser. No. 737,354, June 17, 1968. This application Jan. 20, 1971, Ser. No. 108,203

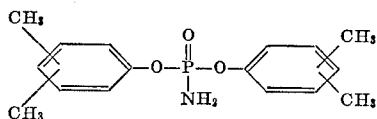
Int. Cl. C09d 5/18; C08b 21/20

U.S. Cl. 106-15 FP

4 Claims

ABSTRACT OF THE DISCLOSURE

A process for spinning synthetic fibers, such as rayon, consisting of incorporating therein a fire retardant composition of matter consisting essentially of a dixylyl phosphoramidate having the formula:



wherein the methyl radicals may be attached to any of 2, 3, 4, 5 or 6 positions.

CROSS REFERENCE OF RELATED
APPLICATION

This application is a continuation-in-part of U.S. application, Ser. No. 737,354; filed June 17, 1968, now abandoned.

BACKGROUND OF THE INVENTION

In the process of producing synthetic fibers, it is the practice to press a viscous mass out of a fine aperture and, by means of a subsequent stretch, the coagulated filament is wound in some fitting manner. The pressing of this viscous mass in continuous filament through apertures is commonly known as spinning. Thus, in order to produce a suitable filament, a spinning solution must first be formed. The solution must have a property of tenacity and viscosity so that it can be pulled into fine filaments of great length. It must also have the property of hardening quickly in this form, and it must produce a filament of proper strength and elasticity so that it can be handled and used. The solution should further have the desired characteristics of being of a stable and controllable viscosity under given conditions. It should also have a certain toughness which permits not only generation of the thread, but its further stretching in this form, without breaking, before reaching its fixation point. The viscosity of the solution is dependent on the molecular structure and concentration of the fluid, and this is influenced by the concentration and the size of the molecules. There are also limits to the viscous properties which must not be exceeded. If the viscosity is inordinately increased, it would be difficult to force the mass through the fine apertures, even with tremendous pressure. And if this were accomplished, the speed of thread formation would be too slow for the purpose of commercial production. As a general principle, it may be said that those solutions that are most desirable have a sufficient viscosity to give form and strength to the thread and at the same time possess sufficient fluidity for rapid filament formation.

With the advent of more strict specifications for the fibers, various additives have been proposed for incorporation into the spinning solution to provide specific properties to the end product. One such additive is for the purpose of providing the fiber filament with fire re-

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ardant properties. But any additive that may be added to the spinning solution must have a viscosity within fixed limits so as not to disturb the over-all viscosity of the spinning solution. One such composition is tris(bromochloropropyl)phosphate. This material provides the requisite fire retardant properties and has the proper viscosity limits to be incorporated within the spinning solution in the formulation of rayon or other synthetic fibers.

Another approach that has been proposed is to employ the low cost tris-dichloropropyl phosphate as the fire-retarding agent. This reagent can be manufactured by reacting a phosphorus oxychloride material with epichlorohydrin. However, this material has a too fluid viscosity and cannot be used. Various additives have been proposed for incorporation into the tris-(dichloropropyl) phosphate to adjust the viscosity, but each resulted in impairing the fire retardant property of the material.

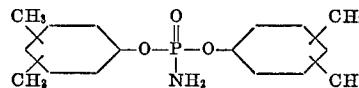
Thus, there has been a long felt need in the synthetic fiber industry for a low cost fire retardant material provided with the requisite viscosity properties for incorporating within the spinning solution.

BRIEF DESCRIPTION OF THE INVENTION

It has been discovered that dixylyl phosphoramidates can be incorporated into rayon spinning solutions so that after regeneration, the compositions will remain in the finished rayon. A sufficient amount of the phosphoroamidates are incorporated into the synthetic fibers so as to impart flame retardance thereto.

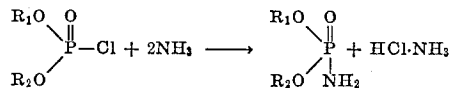
DETAILED DESCRIPTION OF THE
INVENTION

The fire retardant compounds of the present invention are the various isomers of dixylyl phosphoramidates and may be represented by the following formula:

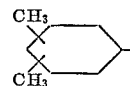


wherein the methyl radicals may be attached to any of 2, 3, 4, 5, or 6 positions on the benzene radicals.

These compounds may be made by reacting dixylyl phosphorochloridate with ammonia and removing the hydrogen chloride which is thus formed according to the following formula:



wherein R₁ and R₂ are



radicals. Either an aqueous ammonia solution may be used for the reaction or gaseous ammonia may be bubbled through the phosphorochloridate until the reaction is complete.

The dixylyl phosphorochloridate may be made by reacting commercially available mixed xylenols with phosphorus oxychloride with or without the addition of a catalyst such as magnesium chloride, aluminum chloride, etc., as is well known in the art.

Since six isomers of xyleneol are known (For example: 2,3-; 3,4-; 2,6-; 2,4-; 3,5-; and 2,5-isomers), and all boil within the range of about 211-255° C., it is difficult, if not impossible, to separate the isomers. The products are necessarily mixtures of these isomers.

For a more complete and detailed description for the manner of manufacturing the dixylyl phosphoramidates, reference is made to U.S. Pat. No. 2,878,255.

The fire retardant composition of this invention can be incorporated into a viscose solution as fully described in U.S. Pat. 3,455,713, issued July 15, 1969. The viscose solution which consists essentially of cellulose, sodium hydroxide, and carbon disulfide, can be passed through the fine apertures of spinnerettes and spun in the shape of one or more filaments into a coagulating and regenerating medium. The formed filaments can then be treated using techniques well known in the rayon field to provide continuous filaments, fibers, and yarn as well as staple fibers. These may then be used to prepare any known textile article in which the flame retardant property is desirable.

The flame retardant, the dixylyl phosphoramidate, of this invention is a liquid of pumpable consistency, which is prepared for example, by the method described hereinabove.

According to the present process, a controlled amount of the flame retardant phosphoramidate can be injected into the viscose just prior to its extrusion through the spinnerettes. The amount of phosphoramidate flame-retardant that should be dispersed in the viscose can vary from about 1 to about 35%, by weight of the viscose solution.

The viscose after being extruded through the spinnerettes can then be passed into an acid coagulating bath, and processed in a conventional manner. The bath may contain, among other materials, sulfuric acid, zinc sulfate, and sodium sulfate and is maintained at a suitable temperature between about 40° and 75° C.

The following examples illustrate the merits of the present invention.

EXAMPLE 1

The flame retardance of dixylyl phosphoramidate was tested in rayon fibers by obtaining four different 1 gram samples of rayon staple. Then, the dixylyl phosphoramidate was dissolved in an acetone carrier. The rayon fibers were soaked in the acetone dixylyl phosphoramidate solution. All the excess acetone was removed and the samples were allowed to stand overnight to evaporate all the acetone. In a conventional flame test stocks, A, B, and C were self-extinguishing while stock D burned readily. The amount of acetone and dixylyl phosphoramidate for each sample are indicated below:

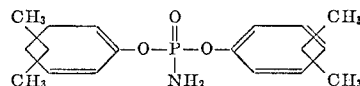
TABLE I

	A	B	C	D
Weight of rayon sample, gr.....	1.0	1.0	1.0	1.0
Acetone, cc.....	50	20	10	0
Dixylyl phosphoramidate, gr.....	5.0	5.0	5.0	0

What is claimed is:

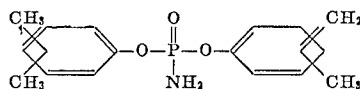
1. In a process for spinning synthetic fibers comprising the steps of: formulating a rayon spinning solution; pressing the solution through fine apertures to form continuous filaments; the improvement comprising incorporating into the spinning solution prior to passing the

solution through the fine apertures an amount ranging between 1 and about 35%, by weight, of a fire retardant composition of matter consisting essentially of a dixylyl phosphoramidate having the following formula:



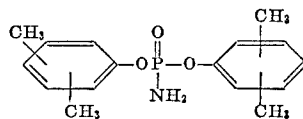
wherein the methyl radicals may be attached to any of 2, 3, 4, 5, or 6 positions.

2. Rayon fibers having dispersed therein a flame retardant amount of a composition of matter consisting essentially of a dixylyl phosphoramidate having the formula:



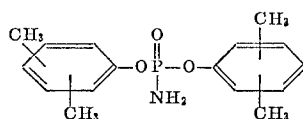
wherein the methyl radicals may be attached to any of 2, 3, 4, 5, or 6 positions.

3. A viscose solution comprising cellulose, sodium hydroxide and carbon disulfide the improvement which comprises the presence therein of from about 1 to about 35%, by weight, of the viscose solution, of a composition of matter consisting essentially of a dixylyl phosphoramidate having the formula:



wherein the methyl radicals may be attached to any of 2, 3, 4, 5 or 6 positions.

4. A method for providing flame-retardant properties to rayon fibers which comprises applying to said rayon fibers a flame-retardant amount of a composition of matter consisting essentially of a dixylyl phosphoramidate having the formula:



wherein the methyl radicals may be attached to any of 2, 3, 4, 5 or 6 positions.

References Cited

UNITED STATES PATENTS

2,878,255	3/1959	Toy et al.	260-959
3,266,918	8/1966	Schappel et al.	106-165
3,455,713	7/1969	Godfrey	106-165
3,531,550	9/1970	Herber et al.	260-959

LORENZO B. HAYES, Primary Examiner

U.S. Cl. X.R.

106-165, 168, 177; 264-194

UNITED STATES PATENT OFFICE
CERTIFICATE OF CORRECTION

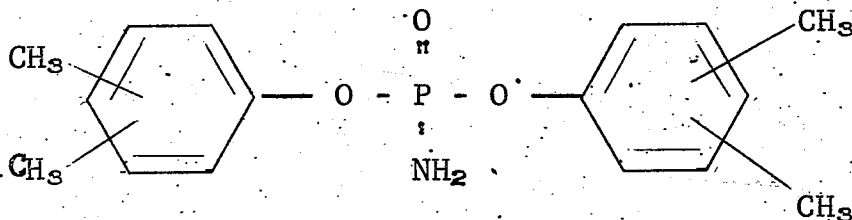
Patent No. 3,704,144 Dated Nov. 28, 1972

Inventor(s) Arthur D. F. Toy and Edward N. Walsh

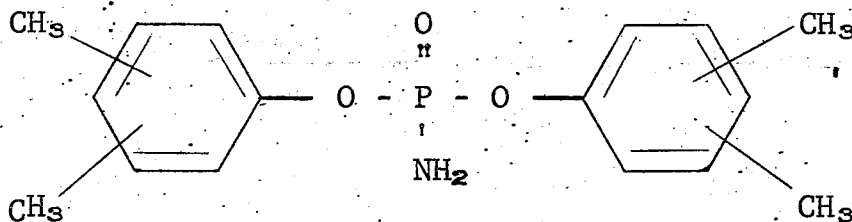
It is certified that error appears in the above-identified patent and that said Letters Patent are hereby corrected as shown below:

Column 2, line 11, the word "Ths" should read -- This --;
line 70, the number "255°" should read -- 225° --.

Column 4, line 5, Claim 1, the formula should read



Column 4, line 15, Claim 2, the formula should read



Signed and sealed this 22nd day of May 1973.

(SEAL)
Attest:

EDWARD M. FLETCHER, JR.
Attesting Officer

ROBERT GOTTSCHALK
Commissioner of Patents