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[54] **CREASEPROOFING CELLULOSE-BASED FABRICS**

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[58] **Field of Search** ..... 117/139.4, 76 T, 117/145, 143 A

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[57] **ABSTRACT**

A method for rendering cellulose-based fabrics wrinkle resistant by impregnating the fibers of the fabric with a polymer builder, polymerizing the polymer builder in the impregnated fabric while the fibers are in a wet and swollen state, drying the fabric, and then depositing a film of silicone polymer on the fabric. Fabrics obtained from the process are described.

8 Claims, No Drawings

CREASEPROOFING CELLULOSE-BASED FABRICS

BACKGROUND OF THE INVENTION

1. Field of the Invention

This invention pertains to the field of wrinkle resistant textiles. More particularly, this invention concerns a method for creaseproofing of cellulose-based textile fibers and fabrics.

2. Background of the Invention

Several methods of imparting crease or wrinkle resistance to textile fibers and fabrics based on, or containing cellulose fibers are known. One such conventional method is known as the pad/dry/cure technique. In this process, a solution of the resin is impregnated into the fabric by padding. The fabric is then dried and baked at an elevated temperature during which curing of the resin takes place. This method is disadvantageous in that the treated cellulose fabrics usually exhibit a substantial deterioration in strength.

A more recent method of imparting crease resistance to cellulose-based fabrics, has been reported by N.R.S. Hollies, and N.F. Getchell, Textile Research Journal 37, pages 70–76, 1964. This method, which they have called the "wet-fixation" process consists essentially of impregnating the fabric with an aqueous, acidic solution of a polymerizable monomer, such as, for example, a melamine resin, and polymerizing the methylol resin within the interstitial spaces of the wet, swollen, fiber. After washing and drying, the fabric is then subjected to cross-linking with a suitable cellulose cross-linking reagent and a catalyst therefor. Alternately, the cross-linking agent can be introduced in the original impregnation step and then subsequently activated after drying of the fabric by treatment with an appropriate catalyst.

Cellulose fabrics treated by the "wet-fixation" process exhibit a degree of wrinkle resistance similar to that obtained with the pad/dry/cure method. Additionally, such fabrics exhibit a relatively higher retention of physical strength than fabrics treated with the pad/dry/-cure method.

Even so, the "wet-fixation" method does result in significant deterioration of the fabric strength as compared with the untreated fabric.

It is also known that the deposition of a film of silicone polymer on a cotton fabric imparts to the fabric a moderate degree of wrinkle resistance without essentially any deterioration in the fabric strength. However, the wrinkle resistance is not nearly as great as that obtained with the pad/dry/cure method or "wet-fixation" method. (See Bullock, J.B. and Welch, C.M. Textile Research Journal 35, pages 459–471, 1965; and U.S. Pat. No. 3,434,875.

SUMMARY OF THE INVENTION

I have discovered a new method for the treatment of cellulose-based fabrics which results in fabrics possessing both a degree of wrinkle resistance equivalent to that obtained by the pad/dry/cure or "wet-fixation" processes described hereinabove and a substantially greater physical strength than fabrics treated by either of those methods.

This method comprises impregnating the fibers of a cellulose-based fabric with a polymer builder, polymerizing the polymer builder in the impregnated fabric while said fibers are in a wet and swollen state, drying the fabric, and depositing a film of silicone polymer on the fabric.

I have also discovered a new class of wrinkle-resistant fabrics which comprise a fabric having a cellulose content of at least about 20 percent based on the total weight of the fabric, which fibers contain between about 2 to 20 percent by weight of a polymerized polymer within the interstitial spaces thereof. The hydroxyl groups of said cellulose fibers may be in either a cross-linked or non-cross-linked state.

Additionally, the present invention comprises garments, draperies, bedspreads, and bedsheets made from such wrinkle-resistant fabric.

DESCRIPTION OF THE PREFERRED EMBODIMENT

In accordance with the present invention, a polymer builder is impregnated into the fibers of a cellulose based fabric.

Any type of cellulose based or cellulose containing fabric may be used in the process of the present invention, such as, for example, cotton, blends of cotton with synthetics, acetate and acetate blends, etc.

Understandably, a certain amount of cellulosic component is needed in the fiber inasmuch as the improved crease-resistant properties result primarily from the modification of the cellulosic portion of the fiber. However, the amount varies depending on the polymer builder used, the other components of the fiber, etc. Preferably, the fabric possesses a cellulosic content of at least about 20 percent based on the weight of the total fabric.

Polymer builders suitable for use in the present invention include urea-formaldehyde resins, melamine-formaldehyde resins, phenol-formaldehyde resins, hydroxyethyl methacrylate and the like.

The polymer builder may be applied to the fabric in any one of the number of conventional methods such as for example, padding, dipping, spraying, etc.

Typically, for example, when the polymer builder is a methylolated melamine resin, the fabric is immersed in an aqueous bath containing from about 5 to 25 percent, preferably about 10 to 15 percent, and most preferably about 12 percent based on the total weight of the mixture, of the methylolated melamine solids. The pH of the bath should be below about 5, preferably below about 4 and most preferably in the range from about 1.5 to 3. Generally, the pH of the bath is easily adjusted by the addition of a mineral acid thereto.

After immersion, the cellulosic fabric is padded to a wet pick-up corresponding to the final amount of melamine solids desired on the fabric. Generally, the solids content of the fabric is desirably in the range from about 2 to 20 percent, preferably from about 4 to 12 percent and most preferably in the range from about 8 to 10 percent, based on the weight of the fabric. Usually, the fabric after immersion is padded to approximately an 80 percent wet pick-up based on the weight of the fabric.

The polymerization of the melamine is dependent on both the pH of the bath and the temperature. The lower the pH, the more rapid the polymerization reaction. Alternatively, the higher the temperature, the more rapid the polymerization reaction. Generally, the polymerization step can be carried out at a temperature ranging from about room temperature to about 212°F and for a time period from about 15 minutes to 24 hours. I have

found it desirable to adjust the pH and the temperature so that the polymerization can be accomplished in a period from about 8 to 24 hours at room temperature.

The polymerization step is carried out in such a manner that the fabric remains in a wet, swollen state during the entire period. Thus, for example, in order to prevent evaporation of water from the fabric, the fabric may be wrapped in plastic or otherwise encapsulated so that no moisture is allowed to escape during the polymerization period. As a result, the fibers are maintained in a wet, swollen condition during the polymerization such that the resin polymerizes to a great extent within the interstitial spacing of the fibers.

After the polymerization period, the fabric is washed, desirably with an alkaline solution to neutralize any residual acid thereon. For example, in a mill operation such neutralization with washing would be achieved during the alkaline soaping in the back washing procedure.

After washing, the fabric is dried in the conventional manner, usually at a temperature in the range from about 100° to 350°F.

As noted hereinabove, other types of polymer builders may be used and the amount of such polymer builder used expressed as its content by weight on the final fabric as well as the specific polymerization conditions depend on the specific polymer builder used. Generally, it is desirable that the fabric obtained after processing contain between about 2 to 20 percent by weight of the polymer therein, based on the weight of the fabric.

After the fabric is dried, a film of a silicone polymer is applied to the fabric. The silicone polymer or elastomer may be applied by a number of methods well known to the art. Preferably, it is applied from an organic solvent solution. After application, the fabric with the silicone polymer thereon is dried and cured.

As used herein, the term "silicone elastomer" means any type silicone polymer composition which can be cast on a glass plate, dried, and if necessary, cured to give a continuous film having elastomeric properties and moderate strength such that the film does not powder or disintegrate when rubbed lightly by hand.

Typically useful silicone elastomers are described in U.S. Pat. No. 3,076,726, and are generally within the class known as methyl polysiloxanes. Additionally, suitable elastomers are described in U.S. Pat. No. 3,434,875, incorporated herein by reference.

Preferred silicone elastomers for use herein are characterized by a very high molecular weight although low molecular weight silicones may also be used so long as the composition is appropriately modified or the conditions selected so as to give a continuous film having the characteristics indicated when the silicone is cast on glass, dried, and if necessary, cured.

The elastomer may be applied to the fabric by conventional methods such as a kiss roll, spraying, knife coating, padding, etc. Usually the amount of elastomer solids added to the fabric depends on a number of conditions such as the fabric construction, the type of elastomer used, etc. Generally, the solids add-on is in the range from about 0.1 to 30 percent and preferably from about 0.6 to 16 percent, based on the weight of the original fabric.

Solvent systems suitable for use in the application of the silicone elastomer include any type of inert solvent which will dissolve the elastomer, such as, for example, hydrocarbon or chlorinated hydrocarbon solvents, e.g. mineral spirits, perchloroethylene, and the like. Particularly suitable is a solvent falling within the classification known as Stoddard solvent. Such a solvent is defined by Commerical Standard C.S. 3–41 and A.S.T.M. D 484–52.

After applying the elastomer in the solution or by whatever method is used, the fabric can be dried in a conventional manner such as air drying or heating. Temperature and time of drying may be selected as desired and can be widely varied so long as all the solvent is removed before curing.

Methods for curing the silicone elastomers and polymers suitable for use in the present invention are well known and depend on the particular elastomer as well as the equipment used. Thus for example, curing times in the range from about 30 minutes at 250°F or 1–2 minutes at 425°F may be used. Alternatively, if a different type of curing oven is used, such as, a roller type oven, heating at a temperature in the range from about 250° to 375°F for about 1 to 10 minutes may be sufficient. Additionally, the silicone elastomers may, if desired, be used in combination with other high polymer elastomers, such as, for example, polyurethanes, polysulfides, or acrylics.

In addition, to the characteristics noted hereinabove for suitable silicone elastomers, it is desirable that the material not be tacky but possess some lubrication or tack-free properties and that it be resilient and possess good recovery from elongation or deformation. Preferably the recovery when tested as a film is in the range from about 70 to 100 percent.

If desired, after drying of the fabric subsequent to polymerization of the polymer builder, but prior to coating with the silicone polymer, the fabric may be subjected to a cross-linking step in accordance with the "wet-fixation" process. Such cross-linking treatment may be carried out in the manner described in the article by N.R.S. Hollies and N.F. Getchell noted hereinabove. The cross-linking treatment can be carried out either by dry or wet methods with various types of cross-linking agents such as dihydroxydimethylolethylene urea and methylolated carbamates. Additionally, of course, a suitable catalyst is included in the treatment.

Alternately, as noted hereinabove in the description of the "wet-fixation" process, it is possible to include the cross-linking agent in the original impregnation step with the polymer builder. In this case, the fabric may be treated with the catalyst activator subsequent to washing and drying but prior to coating with the silicone elastomer. However, the cross-linking agent may be included in the impregnation step without the catalyst treatment.

Thus, cross-linking of the hydroxyls of the cellulosic component is not critical to the achievement of superior wrinkle resistant properties in the fabric when treated in accordance with the process of the present invention.

Additionally, various types of adjuvants well known to the art may be added to the impregnation solution. Thus, for example, softeners may be added to improve the hand of the fabric. Softeners suitable for use in the present process are described in detail in Self-Smoothing Fabrics by J.T. Marsh, Chapman and Hall, Ltd., London, 1962, Chapter 10.

Additionally, the present invention comprises a wrinkle resistant fabric consisting of cellulose-containing fibers, having between about 2 to 20 percent by weight based on the total weight of the fabric of a polymer within the interstitial spaces of the fibers, said polymer preferably being urea-formaldehyde resin, melamine-formaldehyde resin, phenol-formaldehyde resin, or hydroxyethyl methacrylate and wherein the fibers of the fabric are coated with a high molecular weight silicone polymer as described hereinabove, and wherein the cellulosic hydroxyl groups of the fabric may be either in a cross-linked or non-cross-linked state.

Preferably, the fabric has a cellulose content of at least about 20 percent based on the total weight of the fabric.

The fabric of the present invention may be used in any type of end use wherein wrinkle resistance is desired. Thus, for example the fabrics of the present invention may be in the form of garments, draperies, bedspreads, bedsheets, etc.

The following examples serve to illustrate the present invention.

The fabrics produced in the examples were characterized using the following tests:

1. Monsanto Dry Crease Recovery - AATCC test method 66-1968
2. Wyzenbeek Abrasion - ASTM D1175-64T (oscillatory cylinder-400-J metalite cloth as abraidant)
3. Tensile Strength - ASTM D1682-64 (ravelled strip)
4. Tear Strength - ASTM D1424-63
5. Stoll Flex Abrasion - ASTM D1175-64T (flexing and abrasion - two pound tension, 1/2 pound head weight)
6. Water Repellency - AATCC test method 22-1967

Example 1

Three samples of desized, bleached and mercerized 136×64 cotton broadcloth, designated hereinafter Samples A, B, and C, were treated as follows:

Sample A was immersed in an aqueous solution containing 20 percent of a methylated methylol melamine-formaldehyde condensate (Resloom M-75 - Monsanto Company), 0.1 percent of an ethylene oxide condensate of monylphenol (Valdet 561-Valchem Company) and 1 percent concentrated HCl, all based on the weight of the bath. After immersion, the sample was squeezed through pad rolls to about an 80 percent wet pick-up. The fabric was then sealed in a plastic bag to prevent evaporation of moisture and was heated in an oven at 82°C. for 15 minutes to polymerize the melamine condensate. The fabric was then washed in a sodium carbonate bath to neutralize any residual acid, washed with water, and dried.

Sample B was immersed in a perchloroethylene bath comprising 9 percent of a high molecular weight silicone polymer having a viscosity of 10,000 to 25,000 centistokes at 77°F (Dow Corning FC-227) in xylene and 0.2 percent of an organo tin salt (Dow Corning Catalyst 27) xylene, all percentages being based on the total weight of the bath. After immersion, the sample was squeezed to a 100 percent wet pick-up through pad rolls. The fabric was then dried for 10 minutes at 100°–105°C. and cured for 10 minutes at about 165°C.

Sample C, which is a fabric prepared in accordance with the present invention, was prepared by first treating the fabric exactly in accordance with the procedure set forth under Sample A. After the final drying, Sample C was then coated with a silicone polymer in accordance with the procedure set forth for Sample B.

A fourth sample, designated "Control" was not subjected to any treatment and was held for comparison purposes.

The foregoing described Samples A, B, C, and "Control" were then subjected to the tests as set forth hereinabove. The results of the foregoing tests are set forth in Table I.

TABLE I

Sample	Monsanto dry crease recovery ave. W+F	Tensile abrasion, 0 cycles	After Wyzenbeek fill, lbs./in. 100 cycles	Elmendorf tear strength fill, grams	Stoll flex abrasion fill, cycles
A	181	56.4	46.4	712	726
B	235	44.8	16.9	1,440	5,396
C	284	45.6	28.9	904	3,958
Control	182	44.0	8.1	1,288	1,190

As shown in Table I, Sample C, the fabric of the present invention, possesses higher crease resistance than Sample A wherein the silicone coating was not used, Sample B wherein only the silicone coating was used without first impregnating, and the "Control." Moreover, a comparison of Sample A with the "Control" shows that polymerization of impregnated melamine alone has essentially no effect on the crease resistance. Thus, the improvement in crease resistance obtained with Sample C is substantially greater than that obtained with Sample B.

Example 2

A sample of desized, bleached and mercerized 136×64 cotton broadcloth was impregnated with an aqueous solution containing 20 percent Resloom M75, 15 percent of a methylolated hydroxyethyl triazone (Valrez 99N-Valchem Corp.), 5 percent of a nonionic polyethylene emulsion containing 35 percent polyethylene (Valsof PE-45-Valchem Corp.), 3 percent of a lanolin emulsion, containing 25 percent solids 0.1 percent of an ethylene oxide condensate of nonylphenol (Valdet 561-Valchem Corp.) and 1 percent concentrated hydrochloric acid, all percentages being based on the weight of the total solution. Thereafter, the fabric was squeezed through pad rolls to an 80 percent wet pick-up. The fabric was then sealed in a plastic bag to prevent moisture evaporation and was heated in an oven at 82°C for 15 minutes to polymerize the methylolated melamine. The fabric was then washed in a sodium carbonate bath to neutralize any residual acid, washed with water, and dried.

The dried sample was then immersed in an aqueous solution containing 1 percent of a catalyst comprising 25 percent zinc nitrate hexahydrate in water (Catalyst X-4-Sun Chemical Corp.). After the immersion, the sample was padded to an 80 percent pick-up, dried for

5 minutes at about 100°–105°C and finally cured for 5 minutes at about 150°C.

After the curing step, the fabric was coated with silicone using the identical procedure and materials as set forth under Sample B in Example 1 hereinabove.

Properties of the fabric thus treated in contrast with those of the "Control" are set forth in Table II.

TABLE II

Sample	Monsanto dry crease recovery ave. W + F	Tensile abrasion, 0 cycles	After Wyzenbeek fill, lbs./in. 100 cycles	Elmendorf tear strength fill, grams	Stoll flex abrasion fill, cycles
Treated sample...	304	38.5	26.0	768	1,788
Control.....	182	44.0	8.1	1,288	1,190

AATCC 22 Spray Rating of 80 for treated sample.

Example 3

A sample of desized, bleached and mercerized 78×78 cotton print cloth, was immersed in an aqueous solution of 20 percent Resloom-75, 15 percent Valrez 99 N, 5 percent Valsof PE 45, 0.1 percent Valdet 561 and 1 percent hydrochloric acid based on the total weight of the solution. The fabric after immersion was squeezed to 80 percent wet pick-up and then sealed in a plastic bag to prevent moisture evaporation. It was then heated in an oven at about 82° for 15 minutes to polymerize the methylolated melamine. Thereafter, the fabric was washed in a sodium carbonate bath to neutralize any residual acid, washed with water, and dried.

In this case, the fabric was not treated with a cross-linking catalyst even though a cross-linking agent was present in the immersion bath.

The dried fabric was then coated with a silicone polymer in accordance with the procedure set forth for Sample B in Example 1.

The property exhibited by the fabric in comparison with a "Control" are set forth in Table III.

TABLE III

Sample	Monsanto dry crease recovery ave. W + F	Tensile abrasion, 0 cycles	After Wyzenbeek fill, lbs./in. 100 cycles	Elmendorf tear strength fill, grams	Stoll flex abrasion fill, cycles
Treated sample...	297	30.8	8.7	592	3,215
Control.....	200	34.7	13.8	1,040	1,678

Variations and modifications may, of course, be made, without departing from the spirit and scope of the present invention.

I claim:

1. A process for preparing a wrinkle resistant cellulose based fabric comprising:

a. impregnating the fibers of a cellulose-based fabric with an aqueous solution of a polymer builder selected from the group consisting of urea-formaldehyde resins, melamine-formaldehyde resins, phenol-formaldehyde resins, and hydroxyethyl methacrylate resins;

b. polymerizing the polymer builder in the impregnated fabric while said fibers are in a wet and swollen state;

c. drying the fabric; and then

d. depositing a film of silicone polymer on the fabric; wherein said process is carried out in the absence of any cross-linking agent for cellulose.

2. The process of claim **1** wherein the silicone polymer is a high molecular weight methyl polysiloxane elastomer polymer, said polymer being in a substantially non-cross-linked state with respect to said fabric.

3. The process of claim **1** wherein the fabric is impregnated with sufficient polymer builder to produce a polymer content in the final fabric in the range from about 2 to 20 percent based on the total weight of the fabric.

4. The process of claim **1** wherein the polymer builder is melamine formaldehyde resin.

5. The process of claim **4** wherein impregnation step (a) is carried out by immersing the fabric in an aqueous bath containing from about 5 to 25 percent based on the total weight of the bath of methylolated melamine solids, said bath having a pH below about 5, adjusting the wet pick-up of the fabric to about 80 percent based on the weight of the fabric and wherein subsequent to the polymerization step (b), and prior to drying step (c), the fabric is washed with an alkaline solution to neutralize residual acidity.

6. The process of claim **5** wherein the polymerization step is carried out at a temperature in the range from about room temperature to 212°F and for a time period from about 15 minutes to 24 hours.

7. The process of claim **4** wherein the fabric has a cellulose content of at least about 20 percent by weight based on the total weight of the fabric.

8. A wrinkle resistant fabric obtained by the process of claim **1**.

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