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(54) Polyester photographic film support

(57) The present invention is a photographic film base comprising a polyester support having a layer thereon. The layer thereon is a polymer/hydrophilic binder blend wherein the polymer contains a photoreactive moiety having an abstractable hydrogen, and the polymer and hydrophilic binder are present in a ratio of from 1:20 to 20:1. The present invention includes a photographic element wherein a light sensitive silver halide emulsion is superposed on the layer. The present invention provides a method of making the film base described above.

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Description

FIELD OF INVENTION

This invention relates to a method for modifying the surface properties of polymer substrates. More particularly, the present invention relates to modification of the surface of polyester supports to improve adhesion of subsequently applied layers.

BACKGROUND OF INVENTION

In photographic film manufacture, a gelatin layer containing the photographic chemicals is deposited onto a polymer film which provides support and mechanical integrity to the final product. Cellulosic or polyester supports, such as poly(ethylene terephthalate) (PET) and poly(ethylene naphthalate)(PEN), are typically employed. Polyesters have many desirable properties including high mechanical strength, dimensional stability, durability, optical clarity, and resistance to attack by most chemicals. However, the chemical inertness of PET and PEN also results in difficulty in obtaining acceptable adhesion of polar materials, such as gelatin-based photographic emulsions, to PET and PEN substrates.

To obtain acceptable adhesion of the light-sensitive emulsion layer to the support, intermediate anchoring layers are applied to the polyester film support prior to the orientation and crystallization of the support. Adhesion of the anchoring, or subbing, layer is promoted by a variety of methods, including the use of chlorine-containing copolymers, the application of the adhesive layer prior to the orientation and heat setting or crystallization of the polyester, and the addition of organic solvents which attack the polyester film surface. In addition, a subsequent gelatin-containing layer is often required prior to photographic emulsion coating.

Disadvantages of the above described approaches include the requirement of organic solvents, such as chlorophenol and resorcinol, which pose an environmental problem, and the use of chlorinated materials which degrade at elevated temperature and therefore cannot be recycled in the polyester extrusion process, causing economic and environmental problems. In addition, it is often necessary to apply a subbing layer to a polyester film which is already biaxially oriented and heat set. It is more difficult to obtain adhesion to biaxially oriented polyester support as compared to unoriented polyester. Solvents used to attack the polyester surface are less effective on the oriented support. In this case, polymer surface treatments, such as corona discharge (CDT), ultraviolet (UV), and glow discharge (GDT) treatments, are used to promote adhesion through introduction of specific functional groups which interact with subsequent coating layers as described in US Patent Nos. 4,695,532, 4,689,359, 4,933,267, 5,098,818, and 5,407,791. CDT provides sufficient adhesion improvements for many subbing applications, as demonstrated in US Patent Nos. 4,695,532 and 5,102,734, and is performed at atmospheric conditions so is inexpensive relative to other surface treatment methods. GDT provides more dramatic surface modification and rearrangement which is often necessary to obtain the desired adhesion. However, GDT is a vacuum technique so is quite expensive, requiring either very large vacuum chambers (for batch treatment) or expensive interlocks for air-to-air in-line treatment. UV treatment is preferred because it provides the necessary surface modification and can be conducted at atmospheric conditions so is less expensive than GDT.

UV treatment, as an approach to polyester surface treatment, is referred to in, for example, US Patent Nos. 5,407,791, 3,892,575, 4,824,699, and 5,098,818. In US Patent No. 5,407,791, a gel sub with high chlorophenol levels was used to obtain adhesion to UV treated PEN. In US Patent 3,892,575, a polymer/gelatin blend was grafted to polyester using UV radiation. Grafting of monomers to polymer surfaces for surface modification and adhesion improvement (not for photographic applications) is described in US Patent Nos. 4,267,202, 5,209,849, 3,977,954, 4,278,703, JP Kokoku Patent Hei[1991]-6225, and EP Patent Application 521 605 A2.

US patent application No. 08/595,613, filed on April 19, 1996, provides a means for obtaining excellent adhesion of photographic emulsion to oriented polyester support, without the need for phenolic solvents, chlorinated polymers, or expensive GDT processes. However, the subbing layer contains a reactive monomer, such as maleic anhydride, which may be disadvantageous for health and safety reasons. Additionally, monomeric materials may migrate through the emulsion layers resulting in sensimetric keeping problems.

PROBLEM TO BE SOLVED BY THE INVENTION

Thus, there is a continuing need for polyester photographic film supports to which subsequently applied layers will readily adhere.

Further, there exists a need to provide a means for obtaining excellent adhesion of photographic emulsion to oriented polyester support.

SUMMARY OF INVENTION

The present invention is a photographic film base comprising a polyester support having a layer thereon. The layer thereon is a polymer/hydrophilic binder blend wherein the polymer contains a photoreactive moiety having an abstractable hydrogen, and the polymer and hydrophilic binder are present in a ratio of from 1:20 to 20:1. The present invention includes a photographic element wherein a light sensitive silver halide emulsion is superposed on the layer. The present invention provides a method of making the film base described above.

ADVANTAGEOUS EFFECT OF THE INVENTION

The present invention provides a silver halide photographic element which exhibits excellent adhesion between an emulsion layer and an oriented polyester support.

DESCRIPTION OF PREFERRED EMBODIMENTS

The present invention utilizes radiation treatment to modify the surface of polyester support, followed by coating with a blend of polymer and hydrophilic binder wherein the polymer is photo-reactive, thus providing excellent adhesion of gelatin or emulsion layers to the support. Further, the present invention comtemplates photograpic elements having at least one light-sensitive silver halide emulsion layer on the free surface of the polymer/hydrophilic binder layer. In addition the present invention contemplates a method of making a photographic support and element wherein a polymer/hydrophilic binder layer is applied to the surface of a polyester support.

Any suitable polyester may be employed in the practice of this invention as the photographic film support, including polyethylene terephthalate, polyethylene napthalate, polyethylene isothalate, polybutalene terephthalate, polyethylene cocyclohexane dimethylterephthalate, polyethanol codimethanol cyclohexane napthalate, polycarbonates, copolymers and blends thereof and the like.

Any suitable radiation treatment for the polyester support may be employed such as, for example, corona discharge treatment, flame treatment, high energy visible light treatment, ultraviolet light, high frequency wave treatment, glow discharge treatment, active plasma treatment, laser treatment and the like. Ultraviolet light is the preferred radiation source. Ultraviolet radiation in the range of 170 nm to 400 nm is most preferred. This can be obtained by utilizing a quartz UV lamp. A preferred intensity of UV radiation is from 100 to 5000 mJ/cm^2 per pass under the lamp, and most preferably from 800 to 2400 mJ/cm^2 per pass as measured by a UVICURE high energy UV integrating radiometer produced by Electronic Instrumentation and Technology, Inc., Sterling, VA. Typically 1-10 passes at 30 ft/min are required (preferrably 3-6 passes).

Photo-reactive monomers incorporated into the polymer of the polymer/hydrophilic binder subbing layer include any monomer with an extractable hydrogen. Photo-reactive monomers of this invention include, but are not limited to, vinylidene chloride, vinyl chloride, styrene, butadiene, acrylonitrile, acrylates, hydroxy esters, urethanes, ureas, and vinyl ethers. Examples of polymers which are suitable for this photo-reactive layer include, but are not limited to, a terpolymer of vinylidene chloride, acrylonitrile, and acrylic acid (VdCl$_2$-AN-AA); a terpolymer of vinylidene chloride, methylacrylate, and itaconic acid (VdCl$_2$-MA-IA); a terpolymer of vinylidene chloride, acrylonitrile, and an ethanaminium salt of trimethyl-2-[(2-methyl-1-oxo-2-propenyl)oxy]-methyl sulfate (VdCl$_2$-AN-S); a terpolymer of butylmethacrylate, 2-methyl-2-aminoethyl ester of propenoic acid, and 2-methyl-2-hydroxyethyl ester of propenoic acid (BAmH); polyurethanes, polyurethane-ureas, polystyrene, poly(styrene-co-butadiene), polymethacrylates and polyacrylates, and poly(vinylmethyl ether-co-maleic anhydride). Preferred polymers for this application are water insoluble (i.e., latex polymers) or water dispersible polymers.

In the present invention, the polyester support is exposed to radiation, preferable UV radiation in the range of 170 nm to 400 nm. This is obtained using, for example, a quartz UV lamp. A preferred intensity of UV irradiation ranges from 100 to 5000 mJ/cm^2 per pass under the lamp and more preferably from 800 mJ/cm^2 to 2400 mJ/cm^2 per pass under the lamp, as measured by a UVICURE high energy UV integrating radiometer calabrated in the UVA range, from 320 nm to 390 nm, produced by Electronic Instrumentation and Technology, Inc., Sterling, VA. One to ten passes under the lamp at 30 ft/min (preferrably 3-6) are typically used to obtain the desired adhesion.

It is desirable to include a hydrophilic binder in the polymer layer. Suitable hydrophillic binders include gelatin, gelatin derivatives, casein, agar, sodium alginate, starch, polyvinyl alcohol, polyacrylic acid copolymer, maleic anhydride copolymer, cellulose ester, such as carboxymethyl cellulose and hydroxy ethyl cellulose; latex polymers such as a vinyl chloride-containing copolymer, a vinylidene chloride-containing copolymer, an acrylic acid ester-containing copolymer, a vinyl acetate-containing copolymer, a butadiene-containing copolymer, and the like. Gelatin is preferred.

The polymer, typically a latex polymer, is blended with hydrophilic binder, preferably gelatin, in water. Polymer/gelatin ratios of 1/20 to 20/1, preferably 1/10 to 2/1, are coated on the irradiated support. Total solids concentration of 0.1% to 20% are used (preferably 1-5%) and coated to obtain dry coverages of from 0.5 to 10 mg/dm^2, preferably 1 - 5 mg/dm^2. The polymer/gelatin blend is then coated onto the irradiated support using a suitable surfactant to obtain sufficient wetting of the coating. Such surfactants include, but are not limited to, sodium lauryl sulfonate, dioctyl sodium sulfosuccinate, sodium octylphenylpolyether sulfonate, saponin and the like.

Additionally, the polymer/hydrophilic binder blend may contain an antistatic agent, a matting agent, a surface active agent, a crosslinking agent, and/or a dye. The polymer/gelatin blend is then dried at a temperature ranging from 80°C to 140°C, preferably between 100°C and 140°C for a period of 10 s to 10 min, preferably between 1 min and 5 min.

Polymer coatings described above may contain photosensitizers including, but not limited to, α-diketones as described in US patent 3,933,607 and free radical producers such as benzoin ethers and azobisisobutyronitrile, triplet state sensitizers such as benzophenone, photo-redox photosensitizers, and dye-reduction photosensitizers, as described in US Patent 4,267,202.

The polymer/hydrophilic binder layer can be coated by any suitable coating process well known in the art, for example, dip coating, air knife coating, curtain coating, roller coating, wire bar coating, gravure coating, or extrusion, utilizing a hopper as described in U.S. Patent 2,681,294. When two or more layers are coated they can be applied sequentially or simultaneously according to the processes described in U.S. Patents 2,761,791; 3,508,947; 2,941,898 and 3,526,528.

The polymer/hydrophilic binder layer may be applied to the polyester at any suitable point in the preparation of the polyester. For example, the polymer/hydrophilic binder blend may be applied after extrusion of the polyester into a sheet before any orientation of the polymer sheet is carried out, it may be applied after orientation in a first direction such as, for example, in the machine direction or it may be applied after the biaxial orientation is completed, for example, should the polyester first be subjected to a machine direction stretching and subsequently to a transverse direction stretching, the polymer/hydrophilic binder blend may be applied at any point in the procedure.

The subbed support is then coated with a photographic emulsion. Subsequent to the application of the polymer/hydrophilic binder layer to the polyester support, the polymer/hydrophilic bonder layer is coated with a photosensitive layer or layers that contain photographic silver halide emulsion. The present invention is applicable to both negative and reversal silver halide elements. For reversal films, the emulsion layers as taught in U.S. Patent 5,236,817, especially Examples 16 and 21 are particularly suitable. Any of the known silver halide emulsion layers, such as those described in <u>Research Disclosure</u>, Vol. 176, December 1978, Item 17643 and <u>Research Disclosure</u> Vol. 225, January 1983, Item 22534 are useful in preparing photographic elements in accordance with this invention. Generally, one or more layers comprising a dispersion of silver halide crystals in an aqueous solution of gelatin are applied to the substrate having a polymer/hydrophilic binder layer. The coating process can be carried out on a continuously operating machine wherein a single layer or a plurality of layers are applied. For multicolor elements, layers can be coated simultaneously on the composite support film as is described in U.S. Patent 2,761,791 and 3,508,947. Additional useful coating and drying procedures are described in <u>Research Disclosure</u>, Vol. 176, December 1978, Item 17643. Suitable photosensitive image forming layers include those which provide color or black and white images.

The adhesion tests used are described below.

<u>Dry adhesion test</u>:

The emulsion surface of the green sample (before development) or processed dry sample (after development) was cross-hatched with a razor at 5 mm intervals to make nine squares. An adhesive tape (3M 610 tape) was adhered thereto and rapidly stripped off at a peel angle of 180°. The adhesion was evaluated according to the estimated percent removal of the emulsion.

<u>Wet adhesion test:</u>

A 35 mm x 12.7 cm strip of the coating is soaked at 37.8°C. for 3 min. 15 sec. in Kodak Flexicolor Developer Replenisher. The strip is then scored with a pointed stylus tip across the width of the strip and placed in a small trough filled with a developer solution. A weighted (900 g) filled natural rubber pad, 3.49 cm diameter, is placed on top. The pad is moved back and forth across the strip 100 times. The amount of emulsion removal is then assessed in units of % removed. The lower the value the better the wet adhesion of the system.

<u>Post-Process Dry adhesion test</u>:

The sample is run through the standard C41 development process. Dry adhesion is then measured as described above.

EXAMPLES

Adhesion test results for the following examples are in Table 1.

Example 1 (Invention)

Irradiation of 100 mm poly(ethylene naphthalate) (PEN) manufactured by Imperial Chemicals Incorporated (ICI) was conducted using the Fusions F300 curing system with model LC-6 benchtop conveyor (Fusions UV Curing Systems, 7600 Standish Place, Rockville, MD 20855-2798). The uncoated PEN sample was passed under the lamp six times at a conveyor speed of 30 fpm. The lamp was the D bulb (emission from 200 nm to 450 nm, with major output between 350 nm and 390 nm) with an output of 120 W/cm. The energy density of one pass under the lamp at 30 fpm is approximately 1.5 J/cm^2 as measured by the UVICURE high energy UV integrating radiometer described previously. A photo-reactive polymer/blend of the following formulation was then coated on a 30°C coating block with the following formulation to obtain a total dry coverage of 4 mg/dm^2:

Component 1:	1.2 weight percent gelatin
Component 2:	0.6 weight percent polystyrene
Component 3:	0.01 weight percent saponin surfactant
balance:	deionized water

The coated sample was then dried for 3 min at 120 °C in a standard convection oven. Finally, the coated sample is overcoated with a thick test emulsion (approximately 100 mg/dm^2) of black colloidal silver sol containing 0.236 g of silver with 2.44 g of gelatin.

Example 1 (Comparison)

The procedure in example 1 was repeated, but the UV irradiation step was eliminated.

Example 2 (Comparison)

The procedure in example 1 was repeated, but the polymer (component 2) was eliminated.

Example 2 (Invention)

The procedure of Invention Example 1 was repeated except that the uncoated PEN sample was passed under the lamp three times at a conveyor speed of 30 fpm and the 0.6 weight percent of BAmH was used for component 2. The coated sample was dried for 3 min at 130°C in a standard convection oven.

Example 3 (Invention)

The procedure of Invention Example 1 was repeated using 0.3 weight percent BAmH for component 2. The coated sample was dried for 3 min at 130°C in a standard convection oven.

Example 4 (Invention)

The procedure of Invention Example 1 was repeated using 0.6 weight percent of an aqueous dispersible polyurethane (55% hard segment) for component 2. The coated sample was dried for 3 min at 130°C in a standard convection oven.

Example 5 (Invention)

The procedure of Invention Example 1 was repeated using 0.6 weight percent of a polymethylmethacrylate latex polymer for component 2. The coated sample was dried for 3 min at 130°C in a standard convection oven.

Example 6 (Invention)

The procedure of Invention Example 1 was repeated using 0.6 weight percent of a polystyrene-butadiene latex copolymer (GenFlow 8045) for component 2. The coated sample was dried for 3 min at 130°C in a standard convection oven.

Example 7 (Invention)

The procedure of Invention Example 1 was repeated using 0.6 weight percent gelatin for component 1, 1.2 weight percent of a polystyrene-butadiene latex copolymer (GenFlow 8045) for component 2, and the PEN support was passed under the lamp three times at a conveyor speed of 30 fpm, prior to coating. The coated sample was dried for 3 min at 130°C in a standard convection oven.

Example 8 (Invention)

The procedure of Invention Example 1 was repeated using 0.6 weight percent gelatin for component 1 and 1.2 weight percent of a polyurethane-urea (Witcobond 236) for component 2. The uncoated PEN sample was passed under the lamp three times at a conveyor speed of 30 fpm. The coated sample was dried for 3 min at 120°C in a standard convection oven.

Example 9 (Invention)

The procedure of Invention Example 1 was repeated using 0.6 weight percent gelatin for component 1 and 1.2 weight percent of a self-crosslinking polyurethane urea (Witcobond 240) for component 2. The coated sample was dried for 3 min at 120°C in a standard convection oven.

Example 10 (Invention)

The procedure of Invention Example 1 was repeated using 0.6 weight percent of an acrylic adhesive (Rohm and Haas PR71) for component 2. The uncoated PEN sample was passed under the lamp three times at a conveyor speed of 30 fpm. The coated sample was dried for 3 min at 120°C in a standard convection oven.

Example 11 (Invention)

The procedure of Invention Example 1 was repeated using 0.75% gelatin for component 1 and 1.5 weight percent of $VdCl_2$-AN-S for component 2. The uncoated PEN sample was passed under the lamp three times at a conveyor speed of 30 fpm. The coated sample was dried for 3 min at 120°C in a standard convection oven.

The results of Examples 1-11 are set forth in Table I.

Table 1

Wet and Dry Adhesion Results for Invention and Comparison Examples			
Sample	Dry Adhesion (N/m)	Post Process Dry Adhesion	Wet Adhesion
Invention Ex. 1	A	A	A
Comparison Ex. 1	D	D	D
Comparison Ex. 2	A		D
Invention Ex. 2	A	A	A
Invention Ex.3	A	A	A
Invention Ex. 4	A	A	A
Invention Ex.5	A	A	A
Invention Ex. 6	A	A	A
Invention Ex. 7	A	A	A
Invention Ex.8	A	A	A
Invention Ex.9	A	A	A

Table 1 (continued)

Wet and Dry Adhesion Results for Invention and Comparison Examples			
Sample	Dry Adhesion (N/m)	Post Process Dry Adhesion	Wet Adhesion
Invention Ex. 10	A	A	A
Invention Ex. 11	A	A	A
Definition of codes: Cross hatch dry tape adhesion and wet adhesion - A: 0-5% removed, B: 5-20% removed, C: 20-50% removed, D: 50-100% removed.			

Claims

1. A photographic film base comprising:

    a polyester support having a layer thereon comprising:

        a polymer/hydrophilic binder blend wherein the polymer contains a photoreactive moiety having an extractable hydrogen, and the polymer and hydrophilic binder are present in a ratio of from 1:20 to 20:1.

2. The film base of claim 1, wherein the polymer containing a photoreactive moiety having an extractable hydrogen is selected from the group consisting of terpolymers of vinylidene chloride, acrylonitrile, and acrylic acid; terpolymers of vinylidene chloride, methylacrylate, and itaconic acid; terpolymers of vinylidene chloride, acrylonitrile, and an ethanaminium salt of trimethyl-2-[(2-methyl-1-oxo-2-propenyl)oxy]-methyl sulfate; terpolymers of butylmethacrylate, 2-methyl-2-aminoethyl ester of propenoic acid, and 2-methyl-2-hydroxyethyl ester of propenoic acid; polyurethanes; polyurethane-ureas; polystyrenes; poly(styrene-co-butadiene); polymethacrylates; polyacrylates; and poly(vinylmethyl ether-co-maleic anhydride).

3. The film base according to claim 1 wherein the polyester support is selected from the group consisting of polyethylene terephthalate, polyethylene napthalate, polyethylene isothalate, polybutalene terephthalate, polyethylene cocyclohexane dimethylterephthalate, polyethanol codimethanol cyclohexane napthalate, polycarbonates, and copolymers thereof.

4. The film base according to claim 1 wherein the hydrophilic binder is selected from the group consisting of gelatin, gelatin derivatives, casein, agar, sodium alginate, starch, polyvinyl alcohol, polyacrylic acid copolymer, maleic anhydride copolymer, cellulose esters, vinyl chloride-containing copolymers, vinylidene chloride-containing copolymers, acrylic acid ester-containing copolymers, vinyl acetate-containing copolymers, and butadiene-containing copolymers.

5. The film base according to claim 1 wherein the hydrophilic binder comprises gelatin.

6. The film base according to claim 1 wherein the polymer/hydrophilic binder blend further comprises an antistatic agent.

7. The film base according to claim 1 wherein the polymer/hydrophilic binder blend further comprises a matting agent.

8. The film base according to claim 1 wherein the polymer/hydrophilic binder blend further comprises a surface active agent.

9. The film base according to claim 1 wherein the polymer/hydrophilic binder blend further comprises a surfactant.

10. The film base according to claim 1 wherein the polymer/hydrophilic binder blend further comprises a cross-linking agent.

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EUROPEAN SEARCH REPORT

Application Number
EP 97 20 3832

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.6)
X	EP 0 516 275 A (MINNESOTA MINING & MFG) * page 2, line 50 - line 58; claims 1-14 *	1-10	G03C1/93 G03C1/91 C08J7/04
X	EP 0 729 063 A (EASTMAN KODAK CO) * page 4, line 50 - line 59; claims 1-10 *	1-5,8-10	
X	US 3 501 301 A (NADEAU GALE F ET AL) * claims 1-4; examples XXVII-XXV *	1-5,8-10	
X	GB 1 039 935 A (EASTMAN KODAK COMPANY) * claims 1-21; examples I-III *	1-5,8-10	
X	US 3 271 178 A (GALE F NADEAU ET AL) * claims 1,7; examples I-VII *	1-5,8-10	
X	US 4 001 023 A (VAN PAESSCHEN AUGUST JEAN ET AL) * claim 1; examples 1-11 *	1-6	
X	US 3 403 116 A (REAM MEYER ET AL) * claims 1-6; examples XIII,,XV *	1-5	**TECHNICAL FIELDS SEARCHED (Int.Cl.6)**
X	US 3 892 575 A (WATTS RONALD E ET AL) * claims 1-6; example 12 *	1-5	G03C C08J

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
THE HAGUE	3 April 1998	Bolger, W

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
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D : document cited in the application
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