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(54) **Printable multilayer film.**

(57) This invention relates to a multilayer film which can be printed with various printing systems. The multilayer film comprises a top layer consisting of a mixture of one or more polyesters and one or more polyacrylates, and one or more other layers. Preferably the polyester of the top layer is a copolyester of a diacid consisting of terephthalic acid or 1,4-cyclohexanedicarboxylic acid, and a diol consisting of cyclohexanedimethanol, polyalkylene glycol and/or alkylene glycol. A preferred range of the ratio of polyester to polyacrylate in the top layer is from 99:1 to 80:20. Examples of multilayer films include three- and five-layer films. The multilayer film may be provided with a pressure-sensitive adhesive layer. This invention also relates to the top layer composition as such.

EP 0 464 921 A1

This invention relates to films, in particular pressure-sensitive films which can be printed with various printing systems.

Pressure-sensitive films are often printed for decorative purposes. For the application of such a print various techniques can be applied such as screenprinting, letterpress, offset, flexographic printing, stipple printing, laser printing, copperplate printing, etc. One can also select various types of ink, such as one and two component inks, oxydatively or UV-drying inks, dissolved or disperged or 100% ink systems. In this manner it seems that many combinations film/printing ink/printing technique are possible. In practice, however, particular films for particular purposes are printed by means of specific printing ink/printing technique combinations. For instance, pressure-sensitive polyvinyl chloride (PVC) films to be used outside are usually printed with the combination of PVC-solvent ink/screen printing.

Beside the many advantages of PVC such as durability, chemical resistance, mechanical properties, flame resistance, etc. there are also many disadvantages of using PVC as film: PVC may contain a plasticizer which may be damaging for health; PVC contains heavy metals as heat stabiliser; PVC causes upon ageing and combustion hydrochloric acid; PVC may under certain conditions form dioxines.

Therefore there is a need for a product which does not have these disadvantages but indeed has the physical properties of PVC, particularly the flexibility and printing characteristics. In order to manufacture such a product the expert generally has two alternatives:

a) manufacturing a film on which a printable top layer is applied, and

b) manufacturing a multilayer film having a printable top layer by means of coextrusion.

As far as method a) is concerned, it is already known to make polyethylene terephthalate (PET) films suitable for printing with PVC-silk screen printing ink by providing them with a thin top layer of polyvinylidene chloride (PVDC). It is also known to provide polyolefin films with a nitrocellulose or acrylate top layer for this type of printing. Applying such top layers often takes place in the production process after the formation of the film. It would be advantageous when a film having a top layer printable by PVC silk screen printing ink can be obtained in one step, for example by coextrusion. Moreover, and this is more important, it would be advantageous when a film is provided which not only can be printed with PVC silk screen printing ink but also with other ink/printing systems.

Now it has been found that a particular mixture of polymers is extremely suitable as top layer for a multilayer film having the above-mentioned desired properties.

According to one aspect the invention provides for a multilayer film comprising a top layer A consisting of a mixture of one or more polyesters and one or more polyacrylates, and one or more other layers.

The polyacrylate per se is printable with PVC screen-printing ink but it does not have the mechanical properties which could make it suitable for flexible film. Deformation causes the relatively rapid formation of cracks and fractures. It also has not been possible to realize a sufficient adhesion between the pure polyacrylate and a tie layer which is usually used in multilayer films. The mixture of polyester and polyacrylate provides a good adhesion to tie layers which are common in polyolefin-containing coextrudates. The top layer mixture according to the invention surprisingly appears to be very well printable with a large number of different printing ink systems.

The polyesters which can be used in the top layer A may be any conventional polyester or copolyester. Preferably copolyesters are used which are composed of terephthalic acid or 1,4-cyclo-hexanedicarboxylic acid as diacid and cyclohexanedimethanol, especially 1,4-cyclohexanedimethanol, polyalkylene glycol and/or alkylene glycol as diol. Such copolyesters are for example described in US patent specifications 3,523,923, 3,943,189 and 2,901,466.

Examples are copolyesters which are commerically available under the name Ecdel® of Eastman Kodak, Arnitel® of Akzo, Hytrel® of Du Pont de Nemours and Kodar® of Eastman Kodak Company. Suitable copolyesters are for example polymers consisting of several of the following coponents: 1,4-cyclohexanedicarboxylic acid; 1,4-cyclohexanedimethanol; polytetramethylene glycol; terephthalic acid; ethylene glycol; polyalkylene glycol, e.g.resulting in copolyesters known as PET,PETG,PCTG,PBT and PCCE. Optionally, beside the mentioned diacids and diols also other diacids and diols may be present. Mixtures of the mentioned polyesters may also be applied.

The polyacrylates which can be used in the top layer A may be a homopolymer, copolymer or terpolymer of acrylates, methacrylates, acrylic acid and methacrylic acid. Preferably polyacrylates are applied which are based on (C_1-C_4 alkyl)methacrylates, such as methylmethacrylate (MMA), ethylmethacrylate (EMA), butylmethacrylate (BMA) and isobutylmethacrylate (IBMA), which for instance give the following polymers: PMMA, PEMA, PBMA, PIBMA, P(MMA/IBMA), P(MMA/BMA), P(BMA/MMA), P(EA/MMA), P(EA/EMA/MMA) and copolymers with acrylic acid. Examples are polyacrylates which are commercially available under the name of Paraloid® of Rohm and Haas, and Neocryl® of ICI.

The ratio of the polyester to the polyacrylate in the mixture which forms the top layer A, is not very

critical to obtain the desired printing properties. The ratio may vary from 1:99 to 99:1. To obtain a good adhesion to the underlaying layer it is, however, desirable that the polyester is present in a major amount. The ratio of polyester to polyacrylate preferably varies from 99:1 to 50:50 and more preferably from 99:1 to 80:20.

When PTCG is used as polyester, the proportion of polyacrylate is preferably very small, e.g. a ratio of 98:2 to 99:1 is preferred.

The top layer A may optionally contain, beside the polyester and the polyacrylate, other polymers, such as poly-carbonates and poly-amides or small amounts of rubbers , in order to obtain special properties.

According to one embodiment the multilayer film comprises three layers, the top layer A, a polymeric tie layer B and a core polymeric layer C, successively, the core layer C being the thickest layer.

The tie layer B serves to adhere the layers A and C together. The use of tie layers in multilayer films is known per se. The tie layers are usually modified polyolefins composed of polyolefin main chains, such as polyethylene, polypropylene, ethylene vinylacetate (EVA), etc. on which functional groups like ethyl-methacrylate, etc. are grafted. Instead of a grafting a block or terpolymerisate can also be made. Examples of suitable tie layers in the present multi-layer films are the commerical products Admer® of Mitsui Petrochemical Industries, Ltd. and Lotader® of Orkem and Orevac® of Atochem.

The core layer C may consist of all kinds of materials, plastics as well as non-plastics. Usually for economical reasons a material such as a polyolefin will be selected. Usual polyolefins are low-density polyethylene (LDPE), high-density polyethylene (HDPE), polypropylene (PP), propylene-ethylene copolymer, ethylene vinylacetate and mixtures of such materials or mixtures which contain such materials. For example, polyamids, polyesters, polystyrene etc.may also be used as core layer C.

As mentioned above the core C is the thickest layer. This layer is at least 5µm and may further have any desirable thickness. Usually the layer C will have a thickness of approximately 30 to 90µm. The tie layer usually has a thickness of 0.5 to 30µm, often 5 to 15µm, and the top layer has a thickness of 0.5 to 30µm, preferably 5 to 15 µm.

According to another embodiment of the invention the multi-layer film comprises two layers, the top layer A and the tie layer B. The composition and further properties of these two layers are discussed above.

According to yet another embodiment the multilayer film is a symmetric five-layer film A/B/C/B/A. Such a film having on both sides a top layer A as defined above is printable with many printing/ink systems on both sides.

According to a second aspect the invention provides for a pressure-sensitive multilayer film comprising a multi-layer film as described above, the back layer of which (in case of a three-layer film: layer C, and in case of a symmetrical five-layer film: one of the top layers A) is provided with a pressure-sensitive adhesive layer. The adhesive may be any conventional pressure-sensitive adhesive.

The pressure-sensitive multilayer film according to the invention is suitable for many applications, e.g. for stickers and for use outside, for example advertising on shop-windows, busses, etc.

The multilayer film according to the invention is transparent, but in some cases it may be desirable to colour the film with e.g. titanedioxide (white) or soot (black) or another desired pigment combination.

To improve the weather-resistance of the film the top layer A, the tie layer B and/or the core layer C may be stabilized with suitable UV-absorbers/stabilisers, like benzotriazole, benzophenon, hindered amines or combinations thereof, in concentrations of up to 3% by weight.

According to a third aspect the invention provides for a process for the manufacture of the above described multilayer films. In principle, any usual process for manufacturing multilayer films may be used, such as coextrusion, extrusion-coating and lamination. Coextrusion is preferred, because then the product can be obtained in one step.

According to a fourth aspect the invention provides for a composition consisting of a polyester an a polyacrylate as defined above. When applying it as top layer A of a multi-layer film this mixture provides special properties, such as a very broad printability and good adhesion possibilities to conventional tie layers. This composition may be present in any form desired and also in the form of a film. However, from an economical point of view it is not obvious to use such a film per se.

Some examples will follow hereinafter to further elucidate the invention.

Example I

a) Three-layer film having as top layer a mixture of Ecdel 9966 (copolyester of 1,4-cyclohexane-dicarboxylic acid, 1,4-cyclohexane dimethanol and polytetramethylene glycol) and Paraloid B 48 N (copolymer of BMA and MMA).

Ecdel 9966 granulate and Paraloid B 48 N granulate are charged into a vessel in a weight ratio of 60:40.

This vessel is put into a tumbler and tumbled during a half hour.

The macroscopic homogeneous tumbled material is dried in a hot air dryer (mol. sieves) during 4 hours at 70 °C. The dried mixture is then fed into an extruder at temperatures of from 50 to 60 °C.

The single screw extruder has a length of 27 L/D (>20) and a screw diameter of 45 mm. The screw preferably should be equipped with a mix element (so-called Maddock section and dispersion head); however, this is not necessary since an ordinary screw already has a mixing activity. The mixture is extruded at a melt temperature of about 250 °C. The screw causes by its mixing activity a homogenous melt, which flows into the feedblock (according to Dow patent) after leaving the extruder. Also the extrudates of the other two extruders flow into this feedblock. The main extruder (in this case 60 mm and 33 L/D) processes a polypropylene ethylene random copolymer (Appryl 3080 FG3) and the other extruder of 45 mm processes the tie layer material (Admer S 3000 of Mitsui Petrochemical Industries, Ltd). In the feedblock these polymer flows are formed into an A/B/C sandwich.

The cooling roller which follows the flat film head has a temperature of between 10 and 60 °C, so that the liquid film becomes solid. Depending on the surface structure of the cooling roller a high-gloss or matt film can be obtained. The polyester/acrylate blend preferably has contact with this cooling roller. After cooling the side edges are cut off and the film is wound up. In this manner a transparent, flexible film is obtained. The thicknesses of the layers A, B and C are 10, 10 and 60μm, respectively.

The anchorage of the layers to each other is checked. It is tested whether and if so how easy the layers can be separated. This is done by means of a rating. For most examples given the layers are not to be separated at all ("good").

The thus obtained rolls may then be unwound on a coater and provided with a layer of pressure-sensitive adhesive on the layer C. For this purpose this layer of the film should first be corona treated (increase of surface tension by partial surface oxidation), so that a good adhesive adhesion is obtained. The adhesive can be a melt adhesive or based on solvent or water (emulsion). These rolls of pressure-sensitive film may be processed to sheets or stickers which then can be printed.

The sheets are printed with screen techniques and various screening inks. Visprox HSV (suitable for PVC; manufacturer Visprox B.V., Haarlem) and Seriecoll Polydyne YD (suitable for plastics with a low surface tension; manufacturer Sericol Group Ltd., Kent, England) have been used as representatives for these ink categories. Some films are additionally printed with other types of inks, like flexo and UV-letterpress. The adhesion of the ink onto the surface is tested by means of the so-called "tape test" and the scratch resistance of the print is judged by means of the so-called "scratch test". In order to judge the adherence of the ink on the surface a certain tape is applied on the printing in the so-called tape test. Then the tape is drawn loose and the quantity of ink which is adhered to this tape is a measure for the ink adhesion. The scratch test is a subjective test wherein a sharp object is run through the printing; the damage thus made is a measure for the scratch resistance of the ink layer on the surface. The sharp object is often not standardized in the printing practice (finger nail). The results of these two "printability" tests as well as the above-mentioned "layer anchorage" test are given in the attached table. The ratings used are for all three tests:

very bad
bad
acceptable
sufficient
good.

The results of the various test methods are shown in the table incorporated after the examples.

For decorative purposes, letters and logos can be cut out of these films by means of apparatus suitable therefor (Fasson-Graphix), which then e.g. can be sticked on shop-windows etc. for advertising purposes.

The printing methods letterpress (UV-curing inks) and laser printing give good results for stickers. The films may be punched and stripped in the form of a matrix, so that a backing with printed stickers remains.

b) The same as a) but now the Ecdel/Paraloid is tumbled together with a masterbatch, so that the final composition of the top layer is Ecdel/Paraloid (60/40) containing 1%-2% of benzotriazole and 0.5-1% of HALS. To the PP a masterbatch is added also, so that the concentration of HALS is 0.3%. In this manner a weather-resistant film is made which is suitable for longer use outside than is the case with a).

c) The same as a) but now with 15% of TiO_2 and 6% of microtalcum in the PP-layer, so that a white opaque film is obtained.

d) The same as b) but now with 15% of Tio_2 and 6% of microtalcum in the PP-layer, so that a white opaque film for long use outside is obtained.

Example II

a) Three-layer film comprising as top layer a mixture of Ecdel 9966 and Paraloid B44 (copolomer of EA and MMA, but with less acid functionality as B48 N) The process of example I is followed, with the provison that now Paraloid B44 is used instead of Paraloid B 48 N. The weight ratio is 90:10.

b) The same as a), but now the weight ratio is 80:20. The results of the test methods are shown in the table.

Example III

Three-layer film comprising as top layer a mixture of Ecdel 9966 and Paraloid K 120 N (polymethylmethacrylate).

The process of example I is followed, with the provison that now Paraloid K 120 N is used instead of Paraloid B 48 N. The weight ratio of copolyester to polyacrylate is 90:10. The mixture of Ecdel/Paraloid K 120 N is first premixed in a double screw extruder, due to the fact that Paraloid K 120 N is a powder and Ecdel a granulate, so that mixing in the solid phase is impossible. The mixing ratio in the double screw extruder is 50/50 and is realized by gravimetric dosed feeding systems. The extrudate is granulated. The thus obtained 50/50 granulate may according to example I be diluted with Ecdel to form a 90/10 composition in a tumbler, followed by drying and extrusion. The thus obtained film is translucent, because this Paraloid (PMMA) is less compatible with the Ecdel than B48N. In this case another tie layer is used, viz. Admer AT 469 of Mitsui Petrochemicals Industries Ltd., giving the same result as Admer 3000. The thicknesses of the layers are equal to that of example I. The results of the test methods are shown in the table.

Example IV

Five-layer film comprising as outer (top) layers a mixture of Ecdel 9966 and Neocryl B 802 (polymer of IBMA).

The process of Example I is followed, with the proviso that now Neocryl B 802 is used instead of Paraloid B 48 N. The weight ratio of copolyester to polyacrylate is 80:20. The core layer C consists of Eltex P KS 413, another propylene ethylene random copolymer.

The Neocryl B 802 pearlgranulate (fine granulate) is blended with Ecdel (50/50) in the double screw extruder. This granulate is diluted by the method of example I to form a 80/20 mixture. Coextrusion with Eltex P KS 413 takes place under similar conditions as in example I.

In the feedblock the flows of polymer from both extruders of 45 mm are divided into two flows per material and the outcome is joined to a A/B/C/B/A sandwich, which is formed in a flat film head with adjustable gap width to a film of 80μm in thickness, with layer thicknesses of 10/5/50/5/10 μm, respectively.

Example V

Three-layer film comprising as top layer a mixture of Arnitel UX 4723 (polybutylene terephthalate copolymer) and Paraloid B 44 (EA/MMA-copolymer).

The process of example I is followed, except that now a thermoplastic elastomer based on polybutylene terephathalate is used instead of Ecdel 9966. The weight ratio of polyester to polyacrylate is 70:30. The layer thicknesses of the film A/B/C are 20/10/50, respectively. An opaque film is formed the printing results of which are mentioned in the table.

Example VI

Three-layer film comprising as top layer a mixture of Kodar PETG 6763 (copolyester of terephthalic acid, cyclohexane dimethanol and 1,2-ethanediol) and Paraloid B 48 N.

The process of example I is followed, with the exception that now a PETG is used as (co) polyester. The mixability of polyester and polyacrylate is worse than in example I, but a film is formed having a higher hardness than that of example I. The results of the test methods are mentioned in the table. The layer thicknesses of the A/B/C film were 10/10/60 μm.

Example VII

Three-layer film comprising as top layer a mixture of Kodar PCTG 5445 (copolyester of terephtalic acid, 1,4-cyclohexane dimethanol and 1,2-ethanediol) and Paraloid K-120.

The process of example III is followed, with the exception that now a PCTG is used as (co)polyester. The mixability of polyester and polyacrylate is again worse than in example I, and a film is formed having an even higher hardness than that of example III. The polyester/acrylic ratio in this case is 99:1. For process and anchorage reasons another tielayer material is used (Admer SF-710 of Mitsui Petrochemical Industries, Ltd), as well as another type of polypropylene (Valtec HS 010 of Himont). The results of the test methods are mentioned in the table. The layer thicknesses of the A/B/C film were 10/10/60 μm.

Comparative example

Three-layer film comprising as top layer a mixture of Nylon 6 (Ultramid S 3 van BASF) and Paraloid B 48 N.

The process of example I is followed, with the exception that now Nylon 6 is used instead of the polyester. The mixability of the top layer components is worse than in example I. The film obtained has indeed a higher hardness than in example I. The printing characteristics, however, are worse as appears from the results mentioned in the table.

In the following table the results of the various test methods are mentioned. For comparison the results obtained with a PVC-film are also mentioned.

TABLE

Sample/ink	Tape test	Scratch test	Layer anch.
Example I (a)			acceptable
screenprinting			
Sericol Polydyne YD	good	good	
Visprox HSV	good	acceptable	
UV-letterpress printing			
Uvonyl	good	acceptable	
Example I (b)			sufficient
screenprinting			
Sericol Polydyne YD	good	good	
Visprox HSV	good	acceptable	
Example II (a)			good
screenprinting			
Sericol Polydyne YD	good	good	
Visprox HSV	good	sufficient	
flexographic printing			
Flexoplastol HGP	bad	bad	
Flexoplastol HGD	good	acceptable	
UV-letterpress printing			
Uvonyl	good	acceptable	

EP 0 464 921 A1

Sample/ink	Tape test	Scratch test	Layer anch.
Example II (b)			acceptable
screenprinting			
Sericol Polydyne YD	good	good	
Visprox HSV	good	sufficient	
UV-letterpress printing			
Uvonyl	good	acceptable	
Example III			good
screenprinting			
Sericol Polydyne YD	good	good	
Visprox HSV	acceptable	sufficient	
Example IV			good
screenprinting			
Sericol Polydyne YD	good	good	
Visprox HSV	good	good	
Example V			acceptable
screenprinting			
Sericol Polydyne YD	acceptable	sufficient	
Visprox HSV	acceptable	acceptable	
Example VI			sufficient
screenprinting			
Sericol Polydyne YD	good	good	
Visprox HSV	good	sufficient	

Sample/ink	Tape test	Scratch test	Layer anch.
Example VII			good
screenprinting			
Sericol Polydyne YD	good	good	
Visprox HSV	good	good	
flexographic printing			
Flexoplastol HGP	bad	bad	
Flexoplastol HGD	good	acceptable	
Comparative Example			bad
screenprinting			
Sericol Polydyne YD	bad	bad	
Visprox HSV	bad	bad	
PVC film			n.a.
screenprinting			
Sericol Polydyne YD	good	good	
Visprox HSV	good	good	
flexographic printing			
Flexoplastol HGP	good	acceptable	
Flexoplastol HGD	bad	bad	
UV-letterpress printing			
Uvonyl	good	sufficient	

Claims

1. A multilayer film comprising a top layer A consisting of a mixture of one or more polyesters and one or more polyacrylates, and one or more other layers.

2. A multilayer film according to claim 1, **characterized in that** the polyester of the top layer A is a copolyester of a diacid consisting of terephthalic acid or 1,4-cyclohexanedicarboxylic acid, and a diol consisting of cyclohexanedimethanol, polyalkylene glycol and/or alkylene glycol.

3. A multilayer film according to claim 1 or 2, **characterized in that** the polyacrylate of the top layer A is a copolymer of (C_1-C_4 alkyl)methacrylates.

4. A multilayer film according to claims 1 to 3, **characterized in that** the ratio of the polyester to the polyacrylate in the top layer A ranges from 99:1 to 50:50.

5. A multilayer film according to claim 4, **characterized in that** the ratio of the polyester to the polyacrylate in the top layer A varies from 99:1 to 80:20.

6. A multilayer film according to claims 1 to 5, comprising a top layer A and a polymeric tie layer B.

7. A multilayer film according to claims 1 to 5, comprising a top layer A, a polymeric tie layer B and a polymeric core layer C successively, the core layer C being the thickest layer.

8. A multilayer film according to claim 7, **characterized in that** the top layer A has a thickness of 0.5 to 30µm, the tie layer B has a thickness of 0.5 to 30µm, and the core layer C has a thickness of at least 5µm.

9. A multilayer film according to claims 1 to 5, comprising successively a top layer A, a polymeric tie layer B, a polymeric core layer C, a polymeric tie layer B and a top layer A.

10. A multilayer film according to claims 6 to 9, **characterized in that** the tie layer B consists of a modified polyolefin.

11. A multilayer film according to claims 6-10, **characterized in that** the core layer c consists of a polyolefin.

12. A multilayer film according to claim 11, **characterized in that** the core layer C consists of polyethylene, polypropylene or ethylene propylene copolymer.

13. A pressure-sensitive multilayer film comprising a multilayer film according to claims 1 to 12, the back layer of which being provided with a pressure-sensitive adhesive layer.

14. A process for the manufacture of a multilayer film according to claims 1 to 12 comprising coextrusion of the two or more different polymers.

15. A composition consisting of one or more polyesters and one or more polyacrylates.

16. A composition according to claim 15, **characterized in that** the polyester is a copolyester of a diacid consisting of terephthalic acid or 1,4-cyclohexanedicarboxylic acid and a diol consisting of cyclohexanedimethanol, polyalkylene glycol and/or alkylene glycol.

17. A composition according to claim 15 or 16, **characterized in that** the polyacrylate is a copolymer of (C_1-C_4 alkyl)methacrylates.

18. A composition according to claims 15 to 17, **characterized in that** the ratio of the polyester to the polyacrylate varies from 99:1 to 50:50.

19. A composition according to claim 18, **characterized in that** the ratio of the polyester to the acrylate varies from 99:1 to 80:20.

European Patent Office

EUROPEAN SEARCH REPORT

Application Number

EP 91 20 1624

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.5)
A	US-A-4 868 581 (HIDEMASA MOURI ET AL) * column 2, lines 48 - 64; claims 1-8 *	1	B 41 M 5/00

TECHNICAL FIELDS SEARCHED (Int. Cl.5)

B 41 M

The present search report has been drawn up for all claims

Place of search	Date of completion of search	Examiner
The Hague	19 September 91	FOUQUIER J.P.

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
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A : technological background
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P : intermediate document
T : theory or principle underlying the invention

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