ADHESIVE POLYESTER FILM AND LAMINATED FILM PRODUCED BY USING IT

The adhesive polyester film of the present invention is a film having an adhesive coating layer on at least one surface of an aromatic polyester film having a center line average height of the surface of from 1 to 50 nm and a maximum surface protrusion height of from 10 to 500 nm, wherein said adhesive coating layer is produced by applying a coating liquid containing a composition composed of (1) 50 to 97 % by weight of at least one kind of binder selected from a copolymerized polyester resin having an average particle diameter of 150 nm or below and an acrylic copolymer having an average particle diameter of 150 nm or below and (2) 3 to 50 % by weight of an antistatic agent and heating the applied coating liquid to dryness and the thickness of said adhesive coating layer is from 0.02 to 2 \(\mu\)m in dried state. The film has excellent high adhesivity, slipperiness and antistaticity as well as high-speed noadability with a die coated. A laminated film can be produced at a high speed by applying a magnetic layer to the film by a die coater and the produced laminated film is useful e.g. for magnetic card, magnetic tape and floppy disk.
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DESCRIPTION

ADHESIVE POLYESTER FILM AND LAMINATED FILM PRODUCED BY USING IT

Detailed Description of the Invention

Technical Field

The present invention relates to an adhesive polyester film having an adhesive coating layer and a laminated film produced by using the polyester film as a substrate. More particularly, the present invention relates to an adhesive polyester film having excellent adhesivity to a magnetic layer and antistaticity and exhibiting excellent high-speed coatability in the case of applying a magnetic coating material to said adhesive coating layer with a die coater and a laminated film produced by coating said adhesive polyester film with a magnetic recording layer with a die coater.

Background Arts

Aromatic polyester films are being widely used as a base film of a magnetic recording medium, a printing material, etc., however, static charge is liable to generate on the polyester film similar to other plastic films to cause various troubles in the film-forming process, the fabrication process and the use of the product.

A magnetic layer is formed on an aromatic polyester film in the case of using the film for a magnetic recording medium such as video tape, floppy disk or magnetic card by coating the film surface with a magnetic coating material or the surface of the film is printed with an ink in the case of using the film for a printing material, however, aromatic polyester film has a disadvantage of poor adhesivity to magnetic coating materials and printing inks.

Especially a thick white polyester film being used in a magnetic card such as telephone card or prepaid card has problems of poor antistaticity in the production and fabrication process,
antistaticity of the final product and adhesivity to a UV ink or a magnetic coating material. The application of a proper primer to a polyester film has been disposed to solve the problems.

For example, a process for forming an acrylic resin layer as a primer layer on a polyester film is disclosed in the specification of JP-A 62-204941 (hereunder, JP-A means "Japanese Unexamined Patent Publication"), etc., and a process for forming a primer layer containing a polyester resin, an acrylic resin and a compound having sulfonic acid group or sulfonic acid salt group as constituent components is disclosed as a means for improving the adhesivity and antistaticity of a film in the specifications of JP-B 5-41655, JP-B 6-94205 (hereunder, JP-B means "Japanese Examined Patent Publication"), etc.

Coating methods using a roll coater such as direct gravure coating, reverse gravure coating and microgravure coating have been widely used hitherto as the methods for coating a magnetic coating material. These methods have an advantage to enable the coating even to a base film having rough surface and disadvantages of slow coating speed and low productivity.

The improvement of the surface flatness of a magnetic layer and the reduction of the thickness of the layer are being required recently according to the increase in the recording density of magnetic tapes, magnetic disks and magnetic cards. Further, the application of the magnetic coating material at a high speed is required for improving the productivity of a magnetic recording medium.

To meet the above requirements, a die-coating method enabling high-speed coating and having high productivity is used for the application of a magnetic coating material.

The surface of polyester film is roughened in general to prevent the mutual adhesion (blocking) of the films by adding fine particles to the polyester by various means and, accordingly, a polyester film having sufficiently satisfactory quality cannot be obtained yet by the uneven fitting of the polyester film surface to
the lip surface of the die or the generation of die marks on the coated face caused by coarse protrusions sporadically distributed on the film surface to lower the yield of the product.

The above problem can be solved by improving the flatness of the film surface, however, complete flattening of the film surface is difficult owing to the deficient slipperiness of the film to cause the handling difficulty and the generation of blocking of the film.

Accordingly, a polyester film having high adhesivity to a magnetic coating material, enabling high-speed application of a magnetic coating material with a die coater and exhibiting excellent handleability has been desired.

Problems to be Solved by the Invention

The object of the present invention is to provide an adhesive polyester film exhibiting excellent high adhesivity, slipperiness and antistaticity and having excellent high-speed coatability with a die coater.

Another object of the present invention is to provide a laminated film having a magnetic recording layer applied to at least a part of an adhesive coating film of said adhesive polyester film with a die coater.

Means for Solving the Problems

As a result of investigation for solving these problems, the inventors of the present invention have found that an adhesive polyester film exhibiting excellent high adhesivity, slipperiness and antistaticity and suitable for high-speed coating with a die coater can be produced by forming a specific primer layer on a polyester film having specific surface properties.

The present invention is constituted of the following two embodiments.
1. An adhesive polyester film having an adhesive coating layer on at least one surface of an aromatic polyester film having a center line average height of the surface of from 1 to 50
nm and a maximum surface protrusion height of from 10 to 500 nm, wherein said adhesive coating film is produced by applying a coating liquid containing a composition composed of (1) 50 to 97% by weight of at least one kind of binder selected from a copolymerized polyester resin having an average particle diameter of 150 nm or below and an acrylic copolymer having an average particle diameter of 150 nm or below and (2) 3 to 50% by weight of an antistatic agent and heating the applied coating liquid to dryness and the thickness of said adhesive coating layer is from 0.02 to 2 μm in dried state.

2. A laminated film produced by forming a magnetic recording layer on at least a part of the surface of the adhesive coating layer of the above adhesive polyester film.

The aromatic polyester constituting the aromatic polyester film of the present invention is a crystalline linear saturated polyester synthesized from an aromatic dibasic acid or its ester-forming derivative and a diol or its ester-forming derivative. Preferable examples of the aromatic polyester are polyethylene terephthalate, polyethylene isophthalate, polybutylene terephthalate, poly(1,4-cyclohexylene dimethylene terephthalate) and polyethylene-2,6-naphthalenedicarboxylate, including its copolymer and its blended material with a small amount of other resins. Especially preferable polymers among the above examples are polyethylene terephthalate and polyethylene-2,6-naphthalenedicarboxylate.

The aromatic polyester is preferably incorporated with inert particles to impart the film with slipperiness.

Examples of the inert particles are (1) silicon dioxide (including hydrated silicon dioxide, silica sand, quartz, etc.), (2) alumina (including various crystal forms (α-form, β-form, etc.)), (3) a silicate having an SiO₂ content of 30% by weight or above (such as amorphous or crystalline clay mineral, aluminosilicate (including calcined or hydrated material), chrysotile, zircon and fly ash), (4) oxide of Mg, Zn, Zr or Ti, (5) sulfate of Ca or Ba, (6)
phosphate of Li, Ba or Ca (including monohydrogen salt and dihydrogen salt), (7) benzoate of Li, Na or K, (8) terephthalate of Ca, Ba, Zn or Mn, (9) titanate of Mg, Ca, Ba, Zn, Cd, Pb, Sr, Mn, Fe, Co or Ni, (10) chromate of Ba or Pb, (11) carbon (carbon black, graphite, etc.), (12) glass (glass powder, glass bead, etc.), (13) carbonate of Ca or Mg, (14) fluorite, (15) ZnS, (16) particulate heat-resistant polymer (crosslinked silicone resin particle, crosslinked acrylic resin particle, crosslinked polystyrene resin particle, crosslinked styrene-acrylic resin particle, crosslinked polyester resin particle, Teflon resin particle, polyimide resin particle, polyimide-amide resin particle, melamine resin particle, etc.), etc. These inert particles preferably have a narrow particle size distribution and the relative standard deviation is preferably 0.4 or below, more preferably 0.3 or below and especially 0.2 or below.

The kind, average particle diameter and content of the above inert particle in the aromatic polyester are selected to get an aromatic polyester film having a center line average height of 1 to 50 nm and a maximum surface protrusion height of 10 to 500 nm.

The aromatic polyester may be incorporated as necessary with resins other than aromatic polyester such as polyethylene, polypropylene, ethylene-propylene copolymer or olefin-based ionomer and various additives such as stabilizer, antioxidant, ultraviolet absorber and fluorescent brightener.

When the adhesive polyester film of the present invention is for magnetic tape use, the average particle diameter and the content of the inert particle are preferably selected within the ranges of from 0.001 to 10 μm and from 0.001 to 20% by weight, respectively.

Two or more kinds of inert particles having different kinds or different average particle diameters may be used in combination.

When the adhesive polyester film of the present invention is for magnetic card use, the aromatic polyester film is preferably a white film and the aromatic polyester preferably contains a white pigment such as titanium oxide and/or barium sulfate in an amount
of 3 to 30% by weight, preferably 5 to 20% by weight.

The above white film preferably has a light transmittance of 10% or less. The light transmittance is determined in conformity with ASTM D1003 by using a haze meter (manufactured by Murakami Color Research Lab., Type HR-100).

A desired surface glossiness can be attained by selecting the content of the white pigment. Similar to the case of the aromatic polyester film for magnetic tape use, the white film may be incorporated with inert particles in addition to the white pigment. In this case, the average particle diameter and the content of the inert particle are properly selected within the ranges of preferably from 0.0001 to 0.15 μm, especially preferably 0.0005 to 0.1 μm and preferably from 0.001 to 20% by weight, respectively.

The aromatic polyester film of the present invention has surface properties characterized by the center line average height of 1 to 50 nm and the maximum surface protrusion height of 10 to 500 nm. When the center line average height is smaller than 1 nm or the maximum surface protrusion height is smaller than 10 nm, the slipperiness of the film is deteriorated to cause poor handleability. On the contrary, when the center line average height exceeds 50 nm or the maximum surface protrusion height exceeds 500 nm, the high-speed coatability with a die coater is deteriorated.

The thickness of the aromatic polyester film of the present invention is preferably from 1 to 500 μm. It is preferably 1 to 75 μm, especially 2 to 50 μm for a polyester film for magnetic tape use or magnetic disk use and is preferably 20 to 500 μm, further preferably 50 to 500 μm and especially 75 to 300 μm for a polyester film for magnetic card use.

The aromatic polyester film of the present invention may be a single-layer film or a laminated film composed of two or more layers.

For example, a laminated film composed of two or three layers may be used as the film for magnetic tape use or magnetic disk use, wherein at least one surface is produced from an aromatic
polyester containing the aforementioned inert particles and the opposite surface is produced from an aromatic polyester free from inert particles.

The primer layer (adhesive coating layer) to be applied to at least one surface of the aromatic polyester film in the present invention is formed by coating at least one surface of the above aromatic polyester film with a coating liquid containing a composition composed of 50 to 97% by weight of one or more kinds of binders selected from a copolymerized polyester resin having an average particle diameter of 150 nm or below and an acrylic copolymer having an average particle diameter of 150 nm or below and 3 to 50% by weight of an antistatic agent, and heating the applied coating liquid to dryness.

The copolymerized polyester resin constituting the binder of the present invention is produced by the polycondensation of an acid component and a polyl component.

Examples of the acid component are terephthalic acid, isophthalic acid, phthalic acid, 2,6-naphthalenedicarboxylic acid, 1,4-cyclohexanedicarboxylic acid, adipic acid, sebacic acid, phenylindanedicarboxylic acid and dimer acid. Two or more kinds of the above acid components may be used in combination. Terephthalic acid, isophthalic acid and 2,6-naphthalenedicarboxylic acid are preferable among the above examples.

These acid components may be used in combination with a small amount of unsaturated polybasic acids such as maleic acid, fumaric acid or itaconic acid or hydroxycarboxylic acids such as p-hydroxybenzoic acid or p-(β-hydroxyethoxy)benzoic acid. The amount of the unsaturated polybasic acid component and the hydroxycarboxylic acid component is not more than 10 mol%, preferably not more than 5 mol%.

Examples of the polyl component constituting the copolymerized polyester resin are ethylene glycol, 1,4-butanediol, neopentyl glycol, diethylene glycol, propylene glycol, 1,6-hexanediol, 1,4-cyclohexanediol, xyylene glycol, dimethylolpropionic
acid, glycerol, trimethylolpropane, poly(ethylene-oxy)glycol, poly(propylene
oxoy) glycol, ethylene oxide adduct of bisphenol A and propylene
oxide adduct of bisphenol A. Two or more kinds of these polyols
may be used in combination. Among the above examples, ethylene
glycol, ethylene oxide adduct of bisphenol A, propylene oxide adduct
of bisphenol A and 1,4-butanediol are preferable, and ethylene
glycol, ethylene oxide adduct of bisphenol A and propylene oxide
adduct of bisphenol A are especially preferable.

It is allowable and preferable to copolymerize the above
copolymerized polyester resin further with a compound having a
sulfonic acid salt group and/or a compound having a carboxylic acid
salt group to facilitate the uniform dispersion of the resin in an
aqueous coating liquid. The amount of the compound having a
sulfonic acid salt group and/or the compound having a carboxylic
acid salt group is 1 to 20 mol% based on the total dicarboxylic acid
group. It is especially preferably 1 to 20 mol% for the compound
having a sulfonic acid salt group and 5 to 20 mol% for the compound
having a carboxylic acid salt group.

Preferable examples of the compound having a sulfonic acid
salt group are sulfonic acid alkali metal salt compound or sulfonic
acid amine salt compound, etc., such as 5-Na sulfoisophthalic acid,
5-ammonium sulfoisophthalic acid, 4-Na sulfoisophthalic acid, 4-
methylammonium sulfoisophthalic acid, 2-Na sulfoisophthalic acid,
5-K sulfoisophthalic acid, 4-K sulfoisophthalic acid, 2-K
sulfoisophthalic acid and Na sulfosuccinic acid. Especially
preferable compounds among the above examples are 5-Na
sulfoisophthalic acid and 5-K sulfoisophthalic acid.

Examples of the compound having a carboxylic acid salt
group are trimellitic anhydride, trimellitic acid, pyromellitic
anhydride, pyromellitic acid, trimesic acid, cyclobutanetetracarboxylic acid, dimethylolpropionic acid or their
monoalkali metal salts. The free carboxyl group is converted to a
carboxylic acid salt group by reacting with an alkali metal
compound or an amine compound after copolymerization.

A modified polyester copolymer such as a block polymer or graft polymer produced by modifying the above polyester copolymer with an acrylic resin, polyurethane resin, silicone resin, epoxy resin, phenolic resin, etc., can be used as the copolymerized polyester resin.

The copolymerized polyester resin used in the present invention can be produced by known or conventional processes for the production of polyesters. For example, the resin can be produced by reacting 2,6-naphthalenedicarboxylic acid or its ester-forming derivative (especially dimethyl ester), isophthalic acid or its ester-forming derivative (especially dimethyl ester) and trimellitic anhydride with ethylene glycol and propylene oxide adduct of bisphenol A, subjecting the produced monomer or oligomer to polycondensation reaction in vacuum to obtain a copolymerized polyester having a prescribed intrinsic viscosity (preferably between 0.2 and 0.8 measured at 35°C using o-chlorophenol), and reacting the free carboxyl group with an alkali compound or an amine compound to form a salt. The reaction is carried out preferably by using a reaction-promoting catalyst such as an esterification or transesterification catalyst, a polycondensation catalyst, etc. Various additives such as stabilizer may be added to the reaction system.

Examples of the constituent component of the acrylic copolymer constituting the binder in the present invention are acrylic acid, methyl acrylate, ethyl acrylate, butyl acrylate, sodium acrylate, ammonium acrylate, 2-hydroxyethyl acrylate, methacrylic acid, methyl methacrylate, ethyl methacrylate, butyl methacrylate, sodium methacrylate, ammonium methacrylate, 2-hydroxyethyl methacrylate, glycidyl methacrylate, sodium vinylsulfonate, sodium methallylsulfonate, sodium styrenesulfonate, acrylamide, methacrylamide, N-methylol methacrylamide and N-methylol acrylamide. Acrylic acid, ethyl acrylate, butyl acrylate, methacrylic acid, methyl methacrylate, 2-hydroxyethyl methacrylate,
glycidyl methacrylate, N-methylol methacrylamide and N-methylol acrylamide are preferable among the above examples, and ethyl acrylate, butyl acrylate, methacrylic acid, methyl methacrylate, 2-hydroxyethyl methacrylate and N-methylol methacrylamide are especially preferable.

The above constituent components may be used in combination with other unsaturated monomers such as styrene, vinyl acetate, acrylonitrile, methacrylonitrile, vinyl chloride, vinylidene chloride and divinylbenzene.

A modified acrylic copolymer such as a block polymer or graft polymer produced by modifying the above acrylic copolymer with a polyester resin, polyurethane resin, silicone resin, epoxy resin, phenolic resin, etc., may be used as the acrylic copolymer.

The average particle diameter of the copolymerized polyester resin and acrylic copolymer of the binder constituting the coating liquid of the present invention is preferably 150 nm or below, especially preferably 0.1 to 100 nm each in the coating liquid. Coating liquid having an average particle diameter exceeding 150 nm is undesirable because the surface of the formed adhesive coating layer becomes rough to deteriorate the high-speed coating performance with a die coater.

The copolymerized polyester resin and/or the acrylic copolymer are preferably present in the coating liquid in a state dispersed in the form of particles, however, these resins may be dissolved in the coating liquid.

The presence of the copolymerized polyester resin and/or the acrylic copolymer in the coating liquid in a state dispersed in the form of particles is effective for forming an adhesive coating layer having small waviness by the application of the particles to the aromatic polyester film and the heating and drying of the coating liquid to bond the particles to the film. The adhesive coating layer having small waviness improves the adhesivity of the aromatic polyester film to the magnetic recording layer applied by a die coater in the subsequent process and improves the high-speed
coatability with the die coater.

The average particle diameter was determined by using a 200 ppm aqueous liquid of the particle as a specimen, measuring the equivalent sphere diameters of the whole particles by light-scattering method using a fine particle analyzer (Pacific Scientific; Nicomp Model 270) and using the diameter corresponding to the 50% by weight of the whole particles as the average particle diameter.

The binder of the present invention is composed of at least one kind of resins selected from copolymerized polyester resin and acrylic copolymer and is preferably composed of both of a copolymerized polyester resin and an acrylic copolymer. The ratio of both components (copolymerized polyester resin/acrylic copolymer) is preferably 90/10 to 30/70, more preferably 80/20 to 40/60.

The antistatic agent used as a component of the coating liquid of the present invention may be known low-molecular antistatic agent or polymeric antistatic agent. Examples of the low-molecular antistatic agent are alkylsulfonic acid salt, alkylbenzenesulfonic acid salt, alkyl phosphoric acid ester salt, alkyl ethosulfate and betaine ethosulfate, and the examples of the polymeric antistatic agent are acrylic copolymer containing sulfonic acid salt group, vinyl copolymer containing carboxylic acid salt, acrylic copolymer containing quaternary ammonium sulfate, acrylic copolymer containing quaternary ammonium sulfate, polyethylenesulfonic acid salt, ionene polymer, polypyrrolidium derivative, polypyrrole derivative and polyaniline derivative.

A surfactant may be used as the other component constituting the coating liquid of the present invention. Examples of the surfactant are polyoxyethylene alkyl ether, polyoxyethylene alkylphenyl ether, polyoxypropylene alkyl ether, polyoxypropylene alkylphenyl ether, polyoxyethylene alkyl ester, polyoxyethyelene alkylphenyl ester, polyoxypropylene alkyl ester, polyoxypropylene alkylphenyl ester, polyoxyethylene phenyl ether, polyoxypropylene
phenyl ether, poly(oxyethylene-oxypropylene) alkyl ether, poly(oxyethylene-oxypropylene) alkylphenyl ether, polyethylene oxide-polypropylene oxide block copolymer, polyoxyethylene-fatty acid ester, sorbitan fatty acid ester, and glycerol fatty acid ester. Preferable surfactants among the above examples are poly(oxyethylene-oxypropylene) alkyl ether, poly(oxyethylene-oxypropylene) alkylphenyl ether and polyethylene oxide-polypropylene oxide block copolymer, especially poly(oxyethylene-oxypropylene) alkyl ether and poly(oxyethylene-oxypropylene) alkylphenyl ether.

A surfactant other than the above surfactants may be added to the coating liquid, especially an aqueous coating liquid of the present invention, within a range not to spoil the object of the present invention for further improving the wettability to an aromatic polyester film.

The amount of such surfactant is especially preferably selected to give a coating liquid having a surface tension of 50 dyne/cm or below, preferably 40 dyne/cm or below to improve the wettability to an aromatic polyester film.

The composition of the present invention composed of at least one kind of binder selected from copolymerized polyester resin and acrylic copolymer and an antistatic agent contains 50 to 97% by weight, preferably 60 to 90% by weight of the binder and 3 to 50% by weight, preferably 10 to 40% by weight of the antistatic agent. The adhesive force of the primer layer to the aromatic polyester film is lowered to an undesirable level when the ratio of the binder is less than 50% by weight and that of the antistatic agent exceeds 50% by weight. On the contrary, when the amount of the binder exceeds 97% by weight and that of the antistatic agent is less than 3% by weight, the antistaticity of the adhesive polyester film is deteriorated to increase the troubles caused by the static charge generated in handling.

The coating liquid of the present invention preferably contains water as a main component of the solvent, i.e. the coating
liquid is an aqueous coating liquid for keeping the environment and hygiene of the film-manufacturing process. However, the use of an organic solvent as a solvent of the coating liquid is also possible. Examples of the organic solvent are methyl ethyl ketone, acetone, ethyl acetate, tetrahydrofuran, dioxane, cyclohexanone, n-hexane, toluene, xylene, methanol, ethanol, n-propanol and isopropanol. These solvents may be used singly or in the form of a combination of plural solvents.

The coating liquid may be incorporated, in a range not to spoil the object of the present invention, with an ultraviolet absorber, a pigment, a lubricant, an antiblocking agent, a crosslinking agent such as melamine, epoxy compound or aziridine and other additives such as other antistatic agent.

The solid concentration of the coating liquid of the present invention is preferably from 0.5 to 30% by weight.

In the present invention, the coating liquid containing the aforementioned components is applied to at least one surface of an aromatic polyester film, and the film is preferably an aromatic polyester film of the state before completing the crystal orientation.

The aromatic polyester film of the state before completing the crystal orientation is, for example, an undrawn film produced by forming a molten aromatic polyester in the form of a film as it is, a uniaxially drawn film produced by orienting the undrawn film in longitudinal direction or lateral direction, or a film drawn and oriented at low draw ratios in two directions, i.e. longitudinal and lateral directions (a biaxially drawn film before completing the orientation crystallization by the final redrawing in longitudinal or lateral direction).

The coating liquid can be applied to the aromatic polyester film by arbitrary conventional coating methods. For example, it can be performed by roll coating, gravure coating, microgravure coating, reverse roll coating, roll brushing, spray coating, air knife coating, impregnation, curtain coating, etc., or their combination. In the case of using an aqueous coating liquid, the liquid may be
incorporated with a small amount of an organic solvent to improve the stability or the coatability of the coating liquid. The amount of the liquid to be applied to the running film is preferably 3 to 50 g, especially 5 to 40 g per 1 m² of the film. The thickness of the final dried coating layer is 0.02 to 2 μm, preferably 0.05 to 1 μm. The antistaticity is insufficient when the thickness of the coating layer is thinner than 0.02 μm and the slipperiness is deteriorated when the thickness exceeds 2 μm.

The coating can be applied to one surface or both surfaces according to the use of the film. A uniform coating layer is formed by drying the applied coating liquid.

In the present invention, the aromatic polyester film is coated with a coating liquid, heated to dry the liquid and preferably subjected to drawing treatment, and the heating and drying process is carried out preferably at 90 to 130°C for 2 to 20 seconds. The heating and drying process may be used also as the preheating treatment or heating treatment in the drawing process.

The solvent is removed from the coating liquid applied to an aromatic polyester film by the heating and drying treatment, and mainly the binder component in the composition of the coating liquid is bonded to the aromatic polyester film to form an adhesive coating layer. The binder component is effective for improving the adhesivity of the aromatic polyester film to a magnetic recording layer applied by a die coater in the latter process and improving the high-speed coatability with the die coater.

Although there is no particular restriction on the process for producing the aromatic polyester film of the present invention, the preferable production process is a consecutive biaxial drawing process described below.

An aromatic polyester is melted and kneaded at a temperature between Tm and (Tm+70)°C (Tm is the melting point of the aromatic polyester), extruded through a die (T-die, I-die, etc.) and cooled on a cooled rotary drum to form an undrawn film. Subsequently, the undrawn film is drawn in one direction
(longitudinal or lateral direction) at a temperature between (Tg-10) and (Tg+70) °C (Tg is the glass transition temperature of the aromatic polyester) at a draw ratio of 2.5 to 7 and then drawn in a direction perpendicular to the former drawing direction (for example in lateral direction when the first drawing direction is longitudinal) at a temperature between Tg and (Tg+70)°C at a draw ratio of 2.5 to 7. The areal draw ratio is 8 or more, preferably 9 to 32. The biaxially drawn film is heat-set at a temperature between (Tg+70) and Tm °C for 1 to 60 seconds.

Concretely, the drawing is preferably carried out at 70 to 140°C in longitudinal direction at a draw ratio of 2.5 to 7 and then in lateral direction at a draw ratio of 2.5 to 7 and the areal draw ratio of 8 or more, especially 9 to 28 when the aromatic polyester is a polyethylene terephthalate. The redrawing of the biaxially drawn film is performed if necessary at a draw ratio of preferably 1.05 to 3 (provided that the areal draw ratio is maintained in the above range). The heat-setting treatment of the drawn film is preferably carried out at 170 to 240°C for 1 to 30 seconds.

The magnetic recording layer of the present invention can be formed by coating and drying a coating liquid containing magnetic powder and a resin for dispersing the powder. Such magnetic powder is, for example, magnetic metal oxide powder such as γ-Fe₂O₃, CrO₂, Co-γ-Fe₂O₃, Fe₃O₄ and BaO·6FeO₃ and magnetic metal powder such as iron, and the examples of the resin for dispersing the magnetic powder are vinyl resins such as polyvinyl acetate, polyvinylidene chloride or PVC, acrylic resins, rubbery resins such as acrylonitrile/butadiene copolymer, cellulosic resins such as nitrocellulose, epoxy resins, phenolic resins and polyurethane resins. The coating liquid may be incorporated as necessary with a dispersing agent, a lubricant, a stabilizer, an antistatic agent such as carbon, a plasticizer, etc.

A concrete example of a coating liquid containing magnetic powder and a resin for dispersing the magnetic powder is shown below.
Fe (acicular fine particle, aspect ratio: 10/1, coercive force: 20,000 e) 100 parts
Polyurethane resin 15 parts
Vinyl chloride-vinyl acetate copolymer 5 parts
Nitrocellulose resin 5 parts
Aluminum oxide powder (average diameter: 0.3 μm) 3 parts
Carbon black 1 part
Lecithin 2 parts
Methyl ethyl ketone 100 parts
Methyl isobutyl ketone 100 parts
Toluene 100 parts
Stearic acid 2 parts

A laminated film (A) produced by applying a coating liquid containing the above magnetic powder and resin for dispersing the magnetic powder to a part or the whole faces of the adhesive coating layer formed on one or both surfaces of an aromatic polyester film and drying the applied coating liquid is used for magnetic tape and magnetic disk. The thickness of the dried magnetic layer is preferably 1 μm or less, more preferably 0.1 to 1 μm.

The use of a laminated film (A-1) produced by forming adhesive coating layers on both faces of an aromatic polyester film and applying a magnetic layer to each of the adhesive coating layers, i.e. a film having magnetic layers on both faces of an aromatic polyester film is especially preferable for the magnetic disk use.

For the magnetic tape use, the use of a laminated film (A-2) produced by forming adhesive coating layer on one or both faces of a film and applying a magnetic layer on the coating layer is preferable. Especially preferable film is a laminated film (A-3) having a magnetic layer on one surface and a back-coating layer on the surface opposite to the surface having the magnetic layer.

The back-coating layer can be formed e.g. by dispersing inorganic powder such as carbon black in a resin such as a vinyl chloride-vinyl acetate copolymer or a polyurethane, applying the
dispersion to a base film and drying the applied dispersion. A concrete example of the back-coating layer is described in the specification of JP-A 58-200426.

The laminated film (A) of the present invention includes the aforementioned laminated film (A-1), laminated film (A-2) and laminated film (A-3) as preferable embodiments, however, it is not restricted to these preferable embodiments.

A laminated film (B) produced by applying a coating liquid containing the above magnetic powder and a resin for dispersing the magnetic powder to a part or the whole face of an adhesive coating layer formed on one or both surfaces of a film and drying the applied coating liquid is used for magnetic card use. The thickness of the dried magnetic layer is preferably 1 \( \mu \) m or less, more preferably 0.1 to 1 \( \mu \) m. A printed ink layer may be formed on the adhesive coating layer of the laminated film (B).

Although there is no particular restriction on the kind of the ink for the printed ink layer, known printing inks such as an ultraviolet ink, electron beam curable ink and heat-sensitive recording ink can be used for the purpose.

The laminated film (A) and the laminated film (B) of the present invention are produced by coating one or both surfaces of an adhesive polyester film with a magnetic layer by a die coater and drying the applied layer.

The die coater is an apparatus for continuously applying a magnetic coating material extruded through a die slit to the adhesive coating layer of a moving film.

Gravure coater, kiss roll coater, reverse roll coater, etc., are known as coaters other than die coater. These roll coaters are liable, owing to their structure, to generate a streak mark on the coated part of the film by the change of the properties of the coating liquid caused by the contact of the liquid with air and the disturbance of the retained coating liquid in the transfer of the coating liquid film to a roll or a film, and the tendency is especially remarkable in a high-speed coating process.
On the contrary, the above problem is solved by a die coater even in a high-speed coating at 200 m/min or more because the application rate of the coating liquid is controlled with a small gap between the edge at the tip end of the die lip and the running film, however, another problem of the generation of coating unevenness arises in the case of using a film having poor flatness.

The problem of the high-speed coatability with a die coater can be solved by the use of the adhesive polyester film of the present invention to enable the production of the aforementioned laminated film (A) and laminated film (B) in high productivity.

Examples

The present invention is described in more details by the following examples. Various properties cited in the examples were measured and evaluated by the following methods.

(1) Melting point (Tm) and glass transition temperature (Tg)

Melting peak and glass transition point were determined by a DSC (Du Pont Instruments, type 910) at a heating rate of 20°C /min. The amount of the specimen was about 20 mg.

(2) Intrinsic viscosity

A piece of specimen film was dissolved in o-chlorophenol, fillers such as titanium oxide were removed by a centrifugal separator and the intrinsic viscosity was measured at 35°C.

(3) Average diameter of particles

The average particle diameter was determined by using a 200 ppm aqueous suspension of the particle as a specimen, measuring the equivalent sphere diameters of the whole particles by light-scattering method using a fine particle analyzer (Pacific Scientific; Nicomp Model 270) and using the diameter corresponding to the 50% by weight of the whole particles as the average particle diameter.

(4) Center-line average height

The center-line average height was measured by a tracer-type surface roughness tester (Kosaka Kenkyusho Ltd.,
SURFCORDER SE-30C) using a tracer having a tip radius of 2 μm under a measuring pressure of 0.03 g at a cut-off level of 0.25 mm. (5) Maximum surface protrusion height

The measurement of protrusion height was performed on a surface free from the adhesive coating layer using a non-contact three-dimensional roughness tester (WYKO Co., TOPO-3D) at a magnification ratio of 40, a measuring area of 242 μm × 239 μm (0.058 mm²) and n=10 or more. The maximum value of the measured protrusion height was defined as the maximum surface protrusion height.

(6) Adhesive force of magnetic coating material

A magnetic coating material for evaluation prepared by the following method was applied to a specimen film with a Meyer bar in an amount to give a dried coating layer having a thickness of about 4 μm and the applied coating material was dried at 100°C for 3 minutes. The coated film was aged at 60°C for 24 hours, a piece of Scotch Tape No.600 (3M Corp.) of 12.7 mm wide and 15 cm long was pasted on the surface of the magnetic layer taking care of not to trap air bubbles between the magnetic layer and the Scotch Tape, and the laminate was squeezed with a hand-pressing roll described in JIS C2701 (1975) and slit to the tape width. The pasted Scotch Tape was peeled off at an angle of 180° and the peeling force was used as the adhesive force.

<Preparation of magnetic coating material for evaluation>

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<th>Urethane resin</th>
<th>Nippolan 2304 (Nihon Polyurethane)</th>
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<td>Vinyl chloride-vinyl</td>
<td>Eslec A (Sekisui Chemical) acetate resin</td>
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<td>Dispersing agent</td>
<td>Lecion P (Riken Vitamin)</td>
<td>1 pt.wt.</td>
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<tr>
<td>Magnetic powder</td>
<td>CTX-860 (Toda Kogyo Corp.)</td>
<td>500 pts.wt.</td>
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The above solid composition was dissolved in a mixture of methyl ethyl ketone/toluene/cyclohexanone in an amount of 40% by weight and dispersed with a sand grinder for 2 hours. The
dispersion was added with 25 parts by weight (in terms of solid) of a crosslinking agent (Coronate L) and thoroughly stirred to obtain a magnetic coating material.

(7) Adhesive force of UV ink

A specimen film was printed with a UV-curable printing ink (Toyo Ink Mfg.; Flash Dry FD Calton P rouge) by an RI tester (Akari Seisakusho) and cured with a medium pressure mercury lamp UV-curing apparatus (80 W/cm single-lamp apparatus; Japan Storage Battery Co., Ltd.) to form a UV ink layer having a thickness of 4.5 μm. A piece of a cellophane adhesive tape (18 mm wide; Nichiban Corp.) was pasted to the UV ink layer over a length of 15 cm and a definite load was applied to the tape with a 2-kg hand press roll. The peeling force was evaluated by fixing the substrate film and peeling an end of the cellophane adhesive tape in the direction of 90°. The adhesivity was evaluated by the following 5-stage criterion.

<Judgement>

5: Absolutely no peeling of the ink layer was observable.
4: The peeling ratio of the ink layer was less than 3%.
3: The peeling ratio of the ink layer was 3% or more and less than 10%.
2: The peeling ratio of the ink layer was 10% or more and less than 30%.
1: The peeling ratio of the ink layer was 30% or more.

(8) Antistaticity

The antistaticity was evaluated by the surface resistivity of the specimen film. The surface resistivity (Ω/□) was measured by using a resistivity tester of Takeda Riken Corp. at 23°C and 50% relative humidity after the application of 500V potential for 1 minute.

The surface resistivity is preferably $1 \times 10^{11} \Omega/□$ for getting satisfactory antistaticity.

(9) High-speed coating suitability (die coater suitability)

The aforementioned magnetic coating for evaluation was
applied to a specimen film with a slit-die type die coater at a line speed of 250 m/min, the surface defect of the coated film was observed and the coating suitability was evaluated by the following criterion.

5  <Judgement>

○: The coating defect was absolutely unobservable and there was no influence of the defect on the productivity.
△: Little coating defect was observable and there was no remarkable influence of the defect on the productivity.
×: Sporadic coating defects were observable to an extent to cause remarkable deterioration of the productivity.

Example 1

Fine particles of a copolymerized polyester (intrinsic viscosity; 0.57, Tg; 84 °C, average particle diameter; 42 nm) produced from terephthalic acid, isophthalic acid, 5-Na sulfoisophthalic acid, ethylene glycol and propylene oxide adduct of bisphenol A were dispersed in an aqueous solvent to prepare an aqueous coating liquid A consisting of an aqueous dispersion having a solid concentration of 10%.

Separately, a composition composed of 89.5% by weight of a polyethylene terephthalate (intrinsic viscosity; 0.63), 10% by weight of titanium oxide (average particle diameter; 50 nm) and 0.5% by weight of silicon oxide (average particle diameter; 0.2 μm) was melted and extruded on a rotary cooling drum maintained at 20°C, the produced undrawn film was drawn 3.5 times in the machine direction, and a primer coating liquid composed of 70% by weight of the above aqueous coating liquid A, 15% by weight of an aqueous solution of sodium polystyrenesulfonate (concentration; 10% by weight) as an antistatic agent and 15% by weight of an aqueous solution of a polyoxyethylene nonyl ether (concentration; 10% by weight) as a surfactant was applied to both surfaces of the film by kiss coating method at a coating rate of 8 g/m² (wet). The coated film was subsequently drawn 3.6 times in lateral direction to obtain
an adhesive polyester film having a thickness of 188 \mu m. The center-line average height of the film was 30 nm and the maximum surface protrusion height was 300 nm. The adhesivity of a magnetic coating material, the adhesivity of a UV ink, the antistaticity and the high-speed coating suitability of the film were collectively shown in the Table 1.

Example 2

An acrylic polymer composed of methyl methacrylate, butyl acrylate, 2-hydroxyethyl methacrylate and N-methylol methacrylamide (number-average molecular weight 270,000, Tg; 65°C, average particle diameter; 95 nm) was dispersed in water, and the produced aqueous dispersion having a solid concentration of 10% by weight was used as an aqueous coating liquid B.

An adhesive polyester film was produced by the method of the Example 1 except for the use of the aqueous coating liquid B in place of the aqueous coating liquid A used in the Example 1. The adhesivity of a magnetic coating material, the adhesivity of a UV ink, the antistaticity and the high-speed coating suitability of the film were collectively shown in the Table 1.

Example 3

An adhesive polyester film was produced by the method of the Example 1 except for the use of a primer coating liquid composed of 35% by weight of the aqueous coating liquid A, 35% by weight of the aqueous coating liquid B, 15% by weight of an aqueous solution (concentration; 10% by weight) of a polystyrenesulfonic acid sodium salt as an antistatic agent and 15% by weight of an aqueous solution (concentration; 10% by weight) of poly(oxyethylene-oxypropylene) nonyl ether as a surfactant in place of the primer coating liquid used in the Example 1. The adhesivity of a magnetic coating material, the adhesivity of a UV ink, the antistaticity and the high-speed coating suitability of the treated surface of the film were collectively shown in the Table 1.
Example 4

An adhesive polyester film was produced by the method of the Example 3 except for the use of an acrylic copolymer derived from methyl methacrylate, methyl acrylate and sodium methallylsulfonate in place of the antistatic agent used in the Example 3. The adhesivity of a magnetic coating material, the adhesivity of a UV ink, the antistaticity and the high-speed coating suitability of the treated surface of the film were collectively shown in the Table 1.

Comparative Example 1

An adhesive polyester film having a thickness of 188 µm was produced by the method of the Example 1 except for the change of the average particle diameter of the titanium oxide used in the Example 1 to 9 nm and that of the silicon oxide to 7 nm. The center-line average height was 0.5 nm and the maximum surface protrusion height was 8 nm. The adhesivity of a magnetic coating material, the adhesivity of a UV ink, the antistaticity and the high-speed coating suitability of the treated surface of the film were collectively shown in the Table 1.

Comparative Example 2

An adhesive polyester film having a thickness of 188 µm was produced by the method of the Example 1 except for the change of the average particle diameter of the titanium oxide used in the Example 1 to 500 nm and that of the silicon oxide to 2.5 µm. The center-line average height was 300 nm and the maximum surface protrusion height was 2,700 nm. The adhesivity of a magnetic coating material, the adhesivity of a UV ink, the antistaticity and the high-speed coating suitability of the treated surface of the film were collectively shown in the Table 1.

Comparative Example 3
The adhesivity of a magnetic coating material, the adhesivity of a UV ink, the antistaticity and the high-speed coating suitability of a biaxially oriented polyester film of the Example 1 free from the primer coating treatment were collectively shown in the Table 1.

Effect of the Invention

The adhesive polyester film of the present invention has excellent adhesive property, slipperiness and antistaticity and excellent high-speed coatability with a die coater. A laminated film can be produced from the polyester film at a high speed by applying a magnetic layer to the surface of the polyester film with a die coater, and the obtained laminated film is useful e.g. for magnetic card, magnetic tape and floppy disk.
<table>
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<th>Adhesive force of magnetic coating material (g/1/2&quot; width)</th>
<th>Adhesive force of UV ink</th>
<th>Surface resistivity (Ω/□)</th>
<th>High-speed coating suitability</th>
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<td>1</td>
<td>&gt;10^{15}</td>
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CLAIMS

1. An adhesive polyester film having an adhesive coating layer on at least one surface of an aromatic polyester film having surface properties comprising a center-line average height of the surface of from 1 to 50 nm and a maximum surface protrusion height of from 10 to 500 nm, wherein said adhesive coating layer is produced by applying a coating liquid containing a composition composed of (1) 50 to 97% by weight of at least one kind of binder selected from a copolymerized polyester resin having an average particle diameter of 150 nm or below and an acrylic copolymer having an average particle diameter of 150 nm or below and (2) 3 to 50% by weight of an antistatic agent and heating the applied coating liquid to dryness and said adhesive coating layer has a thickness of from 0.02 to 2 μm in dried state.

2. An adhesive polyester film described in the Claim 1 characterized in that the binder is composed of a copolymerized polyester resin having an average particle diameter of 150 nm or below and an acrylic copolymer having an average particle diameter of 150 nm or below and that the weight ratio of said copolymerized polyester resin to said acrylic copolymer (copolymerized polyester/acrylic copolymer) is from 90/10 to 30/70.

3. An adhesive polyester film described in the Claim 1 characterized in that the average particle diameter of the copolymerized polyester resin is from 0.1 to 100 nm and the average particle diameter of the acrylic copolymer is from 0.1 to 100 nm.

4. An adhesive polyester film described in the Claim 1 characterized in that the copolymerized polyester resin is produced by the polycondensation of an acid component and a polyol component, said acid component comprises one or more compounds selected from terephthalic acid, isophthalic acid and 2,6-naphthalenedicarboxylic acid, and said polyol component comprises one or more compounds selected from ethylene glycol, ethylene oxide adduct of bisphenol A and propylene oxide adduct of bisphenol
A.

5. An adhesive polyester film described in the Claim 4 characterized in that the copolymerized polyester resin is produced by the polycondensation of an acid component and a polyol component, said acid component contains a compound having sulfonic acid salt group and/or a compound having carboxylic acid salt group in addition to one or more compounds selected from terephthalic acid, isophthalic acid and 2,6-naphthalenedicarboxylic acid, said polyol component comprises one or more compounds selected from ethylene glycol, ethylene oxide adduct of bisphenol A and propylene oxide adduct of bisphenol A, and the total amount of the compound having sulfonic acid salt group and/or the compound having carboxylic acid salt group is from 1 to 20 mol% based on the total acid component.

6. An adhesive polyester film described in the Claim 5 characterized in that the compound having sulfonic acid salt group and/or the compound having carboxylic acid salt group are 5-Na sulfoisophthalic acid and/or 5-K sulfoisophthalic acid.

7. An adhesive polyester film described in the Claim 1 characterized in that the acrylic copolymer is produced by the copolymerization of two or more kinds of compounds selected from ethyl acrylate, butyl acrylate, methacrylic acid, methyl methacrylate, 2-hydroxyethyl methacrylate and N-methylol methacrylamide.

8. An adhesive polyester film described in the Claim 1 characterized in that the composition constituting the coating liquid contains a surfactant and that the surface tension of the coating liquid is 50 dyne/cm or below.

9. An adhesive polyester film described in the Claim 1 characterized in that the solvent of the coating liquid contains water as a main component.

10. An adhesive polyester film described in the Claim 1 characterized in that the thickness of the aromatic polyester film is from 1 to 500 μm.
11. An adhesive polyester film described in the Claim 10 characterized in that the thickness of the aromatic polyester film is from 1 to 75 µm.

12. An adhesive polyester film described in the Claim 10 characterized in that the thickness of the aromatic polyester film is from 20 to 500 µm.

13. An adhesive polyester film described in the Claim 12 characterized in that the aromatic polyester film is a white film containing 3 to 30% by weight of a white pigment.

14. A laminated film (A) produced by applying a magnetic coating layer by a die coater to at least a part of an adhesive coating layer of the adhesive polyester film described in the Claim 11.

15. A laminated film (B) produced by applying a magnetic coating layer by a die coater to at least a part of an adhesive coating layer of the adhesive polyester film described in the Claim 13.
**INTERNATIONAL SEARCH REPORT**

**A. CLASSIFICATION OF SUBJECT MATTER**

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According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)

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Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

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<td>US 4 590 119 A (KANAKAMI KEN-ICHI ET AL) 20 May 1986 see column 2, line 56 - column 3, line 35 see column 4, line 25-36</td>
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<td>A</td>
<td>EP 0 562 486 A (DIAFOIL HOECHST CO LTD) 29 September 1993 see claims</td>
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<td>A</td>
<td>EP 0 663 286 A (TEIJIN LTD) 19 July 1995 see claims</td>
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| Special categories of cited documents: |
| "A" document defining the general state of the art which is not considered to be of particular relevance |
| "E" earlier document but published on or after the international filing date |
| "L" later document published prior to the international filing date but later than the priority date claimed |

| "T" document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention |
| "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone |
| "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art |

| Patent family members are listed in annex. |

**Date of the actual completion of the international search**

27 October 1998

**Date of mailing of the international search report**

17/11/1998

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk
Tel. (+31-70) 340-2040, Tx 31 651 epo nl, Fax (+31-70) 340-3016

Authorized officer

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