

POLYBUTYLENE TEREPHTHALATE LAMINATE FILM AND ITS USE

FIELD OF THE INVENTION

[0001]

5 The present invention relates to a polybutylene terephthalate laminate film useful for lid sheets for food containers and wrapping materials, and its use.

BACKGROUND OF THE INVENTION

10 [0002]

Conventionally known lid sheets for sealing the openings of instant food containers are laminates comprising an easily peelable sealant film, an aluminum foil and paper in this order, for instance, in the case of containers for instant noodles.

15 [0003]

The lid sheet having an aluminum foil layer is excellent in barrier to steam, etc., dimensional stability to heat, etc., filling adaptability (no curling occurs in the lid sheet when filling the container with a content and sealing it), light screening, etc. The lid sheet having an aluminum foil layer further has
20 properties of keeping a lift-up state when it is peeled from the container substantially to its center position and lifted up (dead-folding characteristics). Because of the dead-folding characteristics, hot water can easily be poured into a container for instant Chinese noodle, etc., for instance, with the peeled lid kept in a half-open state. Thereafter, the lid is closed until the instant noodle
25 becomes eatable, and then reopened to eat.

[0004]

However, the aluminum-foil-containing lid sheets are disadvantages in that when incinerated after use, aluminum foils are fused like ingots in an incinerator, damaging the incinerator and lowering its burning efficiency.

From the aspect of environmental protection, it is desirable to use as few aluminum-foil-containing lid sheets as possible. In the case of instant foods like cupped Chinese noodles, it is necessary from the aspect of food safety to inspect whether or not foreign metallic materials are present in containers.

5 However, the inspection with a metal detector cannot be conducted after the heat-sealing of aluminum-foil-containing lids. Accordingly, the inspection is presently conducted before heat-sealing the lids. Also, the aluminum-foil-containing lids are disadvantageously heavy.

[0005]

10 As a lid sheet containing no aluminum foil, for instance, a laminate film having a paper layer, an adhesion resin layer, a polyethylene terephthalate film layer and an easy-to-peel resin layer in this order is proposed. The polyethylene terephthalate (PET) film has such properties as steam barrier, dimensional stability to heat, filling adaptability (no curling), etc. However,
15 because the PET film has large toughness, it fails to be kept in an open state, (poor in dead-folding characteristics). Accordingly, the lid is easily closed after opening, making it difficult to pour hot water into the container.

[0006]

As a light-weight lid sheet for instant food containers having excellent
20 dead-folding characteristics, which is free from a metal such as aluminum, so that it permits easy detection of metallic foreign matter by a metal detector after sealing, JP 2004-83099A proposes a laminate sheet comprising a polybutylene terephthalate (PBT) film, a paper sheet and a sealant film in this order.

25 [0007]

However, because the PBT film generally has insufficient transparency, a printed layer formed on a PBT film or a paper sheet in the lid sheet described in JP 2004-83099A would be insufficient in the clarity of printed letters and

design.

OBJECT OF THE INVENTION

[0008]

5 Accordingly, an object of the present invention is to provide a laminate comprising at least a polybutylene terephthalate film, a printed layer and a substrate, and capable of providing excellent clarity in printed letters and design.

10 DISCLOSURE OF THE INVENTION

[0009]

 As a result of intense research in view of the above object, it has been found that in a laminate comprising at least a polybutylene terephthalate film, a printed layer and a substrate, the formation of a transparent resin coating layer
15 on a surface of the polybutylene terephthalate film, or on a surface of a printed layer on the polybutylene terephthalate film can make the printed letters and design clearly visible. The present invention has been completed based on this finding.

[0010]

20 Thus, the polybutylene terephthalate laminate film of the present invention comprises at least a polybutylene terephthalate film, and a substrate constituted by a paper sheet or a resin film in this order from an upper surface, a printed layer being formed on an upper surface of the substrate, and/or on a surface of the polybutylene terephthalate film, and a transparent resin coating
25 layer being formed on an upper surface of the polybutylene terephthalate film, or on an upper surface of the printed layer on the polybutylene terephthalate film.

[0011]

The transparent resin coating layer is preferably made of polyvinyl

alcohol. The transparent resin coating layer is preferably as thick as 1-5 μm .
[0012]

The laminate of the present invention preferably comprises at least the transparent resin coating layer, the polybutylene terephthalate film, the printed
5 layer, the substrate and a heat-sealable film in this order from an upper surface. The laminate preferably has a transparent adhesive layer between the transparent resin coating layer and the polybutylene terephthalate film. The transparent resin coating layer is preferably formed by applying a coating composition comprising the transparent resin dissolved or dispersed in a
10 solvent and then dried, so that the shrinkage of the transparent resin coating layer provides the polybutylene terephthalate laminate film with curlability. The laminate having such a layer structure is useful as a lid sheet for food containers.

15 BRIEF DESCRIPTION OF THE DRAWINGS [0013]

Fig. 1 is a cross-sectional view showing one example of the layer structure of the PBT laminate film of the present invention;

Fig. 2 is a cross-sectional view showing another example of the layer
20 structure of the PBT laminate film of the present invention;

Fig. 3 is a cross-sectional view showing a further example of the layer structure of the PBT laminate film of the present invention;

Fig. 4 is a cross-sectional view showing a still further example of the layer structure of the PBT laminate film of the present invention;

25 Fig. 5 is a cross-sectional view showing a still further example of the layer structure of the PBT laminate film of the present invention;

Fig. 6 is a cross-sectional view showing a still further example of the layer structure of the PBT laminate film of the present invention;

Fig. 7 is a photograph showing a surface of the PBT laminate film of

Example 1; and

Fig. 8 is a photograph showing a surface of the PBT laminate film of Comparative Example 1.

5

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

[0014]

[1] Layer structure of polybutylene terephthalate laminate film

Figs. 1-6 exemplify the preferred layer structures of the polybutylene terephthalate (PBT) laminate film 1 of the present invention. In the example shown in Fig. 1, it comprises a transparent resin coating layer 2, a PBT film 3, a transparent adhesive layer 4, a printed layer 5, and a paper sheet 6 in this order from an uppermost surface. The example shown in Fig. 2 is the same as shown in Fig. 1, except that the printed layer 5 is formed on a rear surface of the PBT film 3, that the transparent adhesive layer 4 is formed between the transparent resin coating layer 2 and the PBT film 3, and that an adhesive layer comprising a hot-melt layer 7 and an extrusion-laminated polyethylene layer 8 is formed between the printed layer 5 and the paper sheet 6. The example shown in Fig. 3 is the same as shown in Fig. 2, except that the printed layer 5 is formed on an upper surface of the PBT film 3 without the transparent adhesive layer 4.

[0015]

When the PBT laminate film 1 is used as a lid sheet for containers for such food as instant food, the paper sheet 6 is preferably provided with an outer layer 1a and an inner layer 1b as shown in Figs. 4-6. The example shown in Fig. 4 is the same as shown in Fig. 1, except that the inner layer 1b comprises a resin film 9, a light-blocking ink layer 10, and a sealant film 11 in this order from the side of the paper sheet 6, with a polyethylene adhesive layer 8 formed between the paper sheet 6 and the resin film 9, and between the

light-blocking ink layer 10 and the sealant film 11.

[0016]

The example shown in Fig. 5 is the same as shown in Fig. 4, except that the inner layer 1b does not have a resin film 9, without the adhesive layer 8 between the paper sheet 6 and the light-blocking ink layer 10. The example shown in Fig. 6 is the same as shown in Fig. 5, except that the inner layer 1b has an aluminum foil 12 in place of the light-blocking ink layer 10 as a light-blocking layer. The details of each layer will be explained below.

[0017]

10 (1) Transparent resin coating layer

The transparent resin coating layer 2 formed on an upper surface of the PBT film 3 absorbs fine surface roughness of the PBT film 3, thereby providing the laminate 1 with a flat surface, and thus high clarity free from translucency, making printed letters and design clearly visible. Though the transparent resin 2 is not particularly restrictive as long as it has transparency, it is preferably colorless. The transparent resin 2 may be a known water-soluble or water-insoluble resin, and it is preferably a water-soluble resin because no organic solvent is needed. Examples of the water-soluble transparent resin may be vinyl alcohol polymers, cellulose polymers, polyvinyl pyrrolidone, polyethylene glycol, polyacrylamide, etc. The vinyl alcohol polymers may be polyvinyl alcohol; modified polyvinyl alcohols such as anion-modified polyvinyl alcohol, cation-modified polyvinyl alcohol, acetal-modified polyvinyl alcohol, etc.; ethylene-vinyl alcohol copolymers; partially saponified polyvinyl acetate, etc. The cellulose polymer may be hydroxyethylcellulose, etc.

[0018]

Among them, polyvinyl alcohol is preferable as the water-soluble transparent resin. Because polyvinyl alcohol is inexpensive and has excellent

heat resistance, the polyvinyl alcohol-containing laminate 1 used for lid sheets for food containers does not pose any problems in food safety. The polyvinyl alcohol preferably has a degree of polymerization of 1,000 or less, because it is not easily gelled when preparing an aqueous dispersion. The water-insoluble transparent resins may be acrylic resins, polyurethanes, epoxy resins, polystyrenes, ultraviolet-curable unsaturated acrylic resins, ultraviolet-curable unsaturated ester resins, polycarbonates, etc.

[0019]

Though not particularly restricted, the transparent resin coating layer 2 is as thick as preferably 1-5 μm , more preferably 2-4 μm . When it is thinner than 1 μm , the clarity of printed letters and design is not sufficiently improved. On the other hand, when it is thicker than 5 μm , it is too costly.

[0020]

(2) Transparent adhesive layer

To improve the adhesion of the transparent resin coating layer 2 to the PBT film 3, a transparent adhesive layer 4 may be formed between the transparent resin coating layer 2 and the PBT film 3 if necessary (see Fig. 2). The transparent adhesive 4 may be used as a layer for adhering the PBT film 3 to the paper sheet 6. Though the transparent adhesive 4 is not particularly restrictive as long as it is transparent, it may be made of polyurethane resins, epoxy resins, acrylic resins, ethylene-vinyl alcohol copolymers (EVA), polyvinyl acetals (for instance, polyvinyl formal, polyvinyl butyral (PVB resin), modified PVB, etc.), vinyl chloride resins, etc. The transparent adhesive layer 4 may be formed by applying the above transparent adhesive dissolved or dispersed in a proper solvent, and drying it. The transparent adhesive layer 4 is preferably as thick as 1 μm or less. The thickness of the transparent adhesive layer 4 is not limited as long as it provides sufficient adhesion.

[0021]

(3) Polybutylene terephthalate film

The PBT film 3 can impart dead-folding characteristics, odor retention, gas barrier and heat resistance to the laminate 1. The PBT film 3 is essentially a film of a saturation polyester composed of 1,4-butanediol and terephthalic acid. Within ranges not deteriorating properties such as a thermal shrinkage resistance, etc., other diols than 1,4-butanediol and other carboxylic acids than terephthalic acid may be included as comonomers. Such diols may be, for instance, ethylene glycol, diethylene glycol, neopentyl glycol, 1,4-cyclohexanemethanol, etc. The dicarboxylic acids may be, for instance, isophthalic acid, sebacic acid, adipic acid, azelaic acid, succinic acid, etc. A specific example of PBT resins forming the PBT film 3 may be, for instance, a homo-PBT resin commercially available from Toray Industries, Inc. under the tradename of "Toraycon."

15 [0022]

The PBT resin 3 is not restricted to PBT only, but may contain other thermoplastic resins within ranges not hindering the effects of the present invention depending on its applications. The other resins may be polyesters such as polyethylene terephthalate (PET) and polyethylene naphthalate (PEN); polyphenylene sulfide (PPS); polyamides (PA); polyimides (PI); polyamideimides (PAI); polyethersulfone (PES); polyetheretherketone (PEEK); polycarbonates; polyurethanes; fluororesins; polyolefins such as polyethylene and polypropylene; polyvinyl chlorides; thermoplastic elastomers, etc. For instance, when the PBT resin as a film material contains polyethylene, it has improved film formability, resulting in the PBT film 3 with excellent thickness uniformity. When the other resins are contained, their proportions are preferably 5 to 20% by mass, more preferably 5 to 15% by mass, particularly 5 to 10% by mass, based on the entire PBT resin (100% by mass). Accordingly,

unless otherwise particularly mentioned, it should be noted that the term "PBT resin" used herein includes both PBT only, and a composition comprising PBT and other resins.

[0023]

5 The PBT film 3 may properly contain deodorants, anti-fungus agents, anti-mold agents, flame retarders, water repellants, stain-proofing agents, perfumes, minus ion-generating agents, body pigments such as titanium oxide and calcium carbonate, plasticizers, stabilizers such as antioxidants, UV
10 pigments, lubricants for improving flowability, nucleating agents, inorganic fillers, etc., depending on the use of the PBT laminate film 1.

[0024]

 Though not particularly restrictive, the thickness of the PBT film 3 is preferably about 5 to 50 μm for practical applications. It is technically
15 difficult to make the PBT film 3 as thin as less than about 5 μm , resulting in a higher cost. On the other hand, when the PBT film 3 is as thick as more than about 50 μm , the laminate 1 has poor tearability. The preferred thickness of the PBT film 3 is 10-30 μm . The PBT film 3 as thick as about 5-50 μm has sufficient dead-folding characteristics, odor retention and gas barrier.

20 [0025]

 The thermal shrinkage ratio of the PBT film 3 is preferably 2% or less in both MD (machine direction) and TD (transverse direction). The thermal shrinkage ratio is measured on the PBT film exposed to 150°C for 10 minutes. The PBT film 3 having a thermal shrinkage ratio of 2% or less can be
25 produced by an air-cooled inflation method, which is described in WO 2004/026558.

[0026]

 A lot of substantially parallel linear scratches and/or a lot of fine pores

may be formed to make the PBT film 3 easily tearable linearly as described below.

[0027]

(4) Printed layer

5 The PBT laminate film 1 has a printed layer 5 bearing letters, design, etc. The printed layer 5 is formed on an upper surface of the substrate 6, and/or on a surface of the PBT film 3. As shown in Fig. 1, the printed layer 5 is preferably on an upper surface of the substrate 6. The printed layer 5 may be formed by gravure printing, inkjet printing, etc. Printing ink may have a
10 formulation comprising a base thermoplastic resin such as acrylic resins, polyester resins and polyurethanes, pigments and/or dyes, and various additives. A two-pack polyurethane printing ink has excellent water resistance.

[0028]

15 (5) Substrate

 The laminate 1 comprises a paper sheet or a resin film as the substrate 6. When the laminate 1 is used as a lid sheet for containers of instant food such as cupped Chinese noodle, instant Yakisoba noodle, etc., the paper sheet is preferably used. When the laminate 1 is used as a lid sheet for containers
20 of semi-solid food such as jelly and pudding, the resin film is preferably used.

[0029]

(a) Paper sheet

 The paper sheet 6 can provide the laminate 1 with dead-folding characteristics. The types of papers are not restricted but may include
25 synthetic papers. The basis weight of the paper sheet 6 is preferably about 60 to 120 g/m², more preferably about 75 to 110 g/m². When the basis weight of the paper sheet 6 is less than about 60 g/m², the paper sheet is easily foldable, failing to provide sufficient dead-folding characteristics. On the other hand,

even when the basis weight of the paper sheet 6 is more than about 120 g/m², there is no further improvement in dead-folding characteristics, resulting in only cost increase.

[0030]

5 (b) Resin film

The laminate 1 may have a resin film substrate 6 depending on its applications. Though not particularly restricted, materials for the resin film 6 may be polyesters, polypropylene, nylons, etc. A polyester film particularly provides the laminate 1 with excellent odor retention and gas barrier. A
10 polypropylene film provides the laminate 1 with excellent moisture retention.

[0031]

The polyester film may be made of polybutylene terephthalate (PBT), polyethylene terephthalate (PET), polyethylene naphthalate (PEN), polybutylene naphthalate (PBN), etc. The PET films are commercially
15 available at low costs. The PET film is particularly preferable to impart high rigidity to the laminate 1.

[0032]

The PET film is essentially a film of a saturated polyester made of ethylene glycol and terephthalic acid. Within ranges not deteriorating
20 properties such as a thermal shrinkage resistance, etc., other diols than ethylene glycol and other carboxylic acids than terephthalic acid may be included as comonomers. Such diols may be, for instance, diethylene glycol, 1,4-butanediol, neopentyl glycol, 1,4-cyclohexanemethanol, etc. The dicarboxylic acids may be, for instance, isophthalic acid, sebacic acid, adipic
25 acid, azelaic acid, succinic acid, etc. The PET films are commercially available as biaxially oriented films, such as "Lumirror" of Toray Industries, Inc., "TOYOBO ESTER FILM" of Toyobo, Co., Ltd., and "Emblet PC" of Unitika Ltd., etc.

[0033]

Like the PBT film 3, the PET film is not restricted to a film made of PET only, but may contain other thermoplastic resins within ranges not hindering their properties. Accordingly, unless otherwise particularly mentioned, it should be noted that the "PET film" includes not only PET alone, but also compositions of PET and other thermoplastic resins.

[0034]

Like the PBT film 3, the resin film 6 may contain additives depending on the applications of the PBT laminate film 1. The resin film 6 is preferably as thick as about 5-50 μm as in the PBT film 3.

[0035]

For easy tearing of the laminate 1, it is preferable to use an easy-to-tear resin film 6. Polyester films monoaxially or biaxially orientated to different degrees are preferable as films easily tearable in one direction. The PET film biaxially orientated to different degrees may be, for instance, "Emblet PC" commercially available from Unitika Ltd. As mentioned below, a lot of substantially parallel linear scratches and/or a lot of fine pores may be formed on the resin film 6 for straight tearing.

[0036]

(6) Other layer

When the PBT laminate film 1 is used as a lid sheet for instant food containers, it preferably comprises other layers described later, in addition to the PBT film 3, the printed layer 5 and the paper sheet 6.

[0037]

(a) Resin film

The PBT laminate film 1 comprising a paper sheet as the substrate 6 preferably comprises a resin film 9 for improved rigidity and gas barrier (see Fig. 4). Because the resin film 9 may be the same as usable as the substrate 6,

its explanation will be omitted.

[0038]

(b) Light-blocking layer

To prevent sunlight and ultraviolet rays from causing the deterioration
5 of instant food, the laminate 1 comprises a light-blocking ink layer 10 or an
aluminum foil 12. The light-blocking ink is not particularly restrictive, as
long as it is ink containing a black or dark-color pigment or dye such as carbon
black. The use of the light-blocking ink layer 10 makes it possible to avoid
adverse effect on environment in incineration. It also makes it possible to
10 detect foreign metallic materials by a metal detector after sealing. This
further improves the safety of instant food, and extremely decreases an
examination cost because the metal detector can be utilized after sealing. The
aluminum foil 12 can provide excellent light screening or blocking, gas barrier,
odor retention, etc.

15 [0039]

Though depending on the concentration of black pigment or dye in the
ink, the light-blocking ink layer 10 may generally have such thickness as
sufficiently cutting ultraviolet rays and visible rays. The thickness of the
aluminum foil 12 is preferably 3 to 15 μm , more preferably 7 to 12 μm .

20 [0040]

(c) Heat-sealing layer

A layer heat-sealed to an upper flange of a container body for instant
food is a sealant film 11 or a hot-melt layer. The sealant film 11 may be
formed by a polyethylene film, an unstretched polypropylene film, an ionomer
25 film, a polystyrene film or the like. In order that the lid sheet is easily peeled
from the container body, the sealant film 11 is preferably easy to peel. For
this purpose, the sealant film 11 preferably has relatively weak heat
adhesiveness. Known hot melts may be used.

[0041]

Usable as the sealant film 11 is, for instance, a laminate film comprising a polyethylene base film and a low-molecular-weight polyethylene film in this order from the side of the paper sheet 6. The thickness of this polyethylene base film is preferably about 10 to 40 μm , more preferably about 20 to 30 μm . The thickness of the low-molecular-weight polyethylene film is preferably about 5 to 20 μm , more preferably about 7 to 15 μm . Such laminated polyethylene film is, for instance, 760FD commercially available from Toray Plastic Films Co., Ltd. Also usable as the sealant film 11 is a film made of a mixture comprising an ethylene-vinyl acetate copolymer (EVA) and polyethylene. In the film made of this mixture, the polyethylene is preferably linear low-density polyethylene (LLDPE). The thickness of the film made of this mixture is also preferably about 10 to 40 μm , more preferably about 20 to 30 μm . The thickness of the hot-melt layer is preferably 10 to 50 μm , more preferably 20 to 40 μm .

[0042]

The sealant film 11 is made of a resin composition comprising a linear ethylene- α -olefin copolymer of ethylene and an α -olefin having 3 to 18 carbon atoms, which has a density of 0.870 to 0.910 g/cm^3 (JIS K6922), and MFR of 1 to 100 $\text{g}/10$ minutes (JIS K6921, at 190°C under 2.16kg), and polystyrene (see JP 2004-26190 A). The lid comprising this sealant film 11 can have both sealability and easy-to-open characteristics when heat-sealed to the container body 1.

[0043]

To provide the laminate 1 with excellent straight-tearing characteristics, the sealant film 11 may also be provided with a lot of substantially parallel linear scratches and/or fine pores as described below.

[0044]

(7) Pre-working of each film

(a) Formation of linear scratches

To enhance the easy-to-tear characteristics of the laminate 1, at least one of the PBT film 3, the resin film 6, 9 and the sealant film 11 may be
5 provided with a lot of substantially parallel linear scratches formed by the method described below. To have both high film strength and good easy-to-straight-tear characteristics, the depth of the linear scratches is preferable 1 to 40% of the thickness of each film, particularly 0.1 to 10 μm . The linear scratches are preferably as wide as 0.1-10 μm with intervals of 10 to
10 200 μm .

[0045]

Usable for forming the linear scratches on film 3, 6, 9 and 11 is, for instance, a method disclosed in WO 03/091003. In this method, a continuously moving film is slidably pressed onto a roll or plate comprising a
15 lot of fine particles having Mohs' hardness of 5 or more on a surface, so that a lot of substantially parallel linear scratches are formed on the film surface. The fine particles are preferably fine diamond particles. A means for pressing the roll or plate onto the film is preferably an air-blowing means, or a brush slidable with the film. Because linear scratches formed by this method
20 do not penetrate the film, the use of a polyester film provided with linear scratches as the resin film 6, 9 secures sufficient odor retention and gas barrier.
[0046]

The linear scratches are preferably formed on at least one entire surface of each film 3, 6, 9 and 11. This provides each film 3, 6, 9 and 11 with
25 easy-to-straight-tear characteristics in one direction regardless of the orientability of the film per se, so that the film can be torn straight along the linear scratches from an arbitrary point.

[0047]

(b) Forming of fine pores

To enhance the easy-to-tear characteristics of the laminate 1, at least one of the PBT film 3, the resin film 6, 9 and the sealant film 11 may entirely be provided with a lot of fine pores formed by a method described below.

5 [0048]

The fine pores preferably have an average opening diameter of 0.5 to 100 μm , and a distribution density of about 500 or more per 1 cm^2 . When the distribution density of the fine pores is less than about 500 per 1 cm^2 , tearability is insufficient. The distribution density of the fine pores is more
10 preferably 1×10^3 to 1×10^4 per 1 cm^2 , particularly 1×10^3 to 5×10^3 per 1 cm^2 from the viewpoint of film strength.

[0049]

Fine pores may be formed in each film 3, 6, 9 and 11, for instance, by the methods disclosed in Japanese patent No. 2063411 and JP 2002-59487 A.
15 In the method of Japanese patent No. 2063411, for instance, a film passes under uniform pressure between a first roll having a lot of fine particles with acute corners having a Mohs' hardness of 5 or more attached to a surface (the same as the above roll for forming linear scratches), and a second roll having a smooth surface, so that the film is formed with lots of fine pores. The second
20 roll may be an iron roll having Ni or Cr plating, a stainless steel roll, a specialty steel roll, etc.

[0050]

When fine pores are formed in the sealant film 11, the fine pores may be either of penetrating holes or non-penetrating holes. When fine pores are
25 formed in the PBT film 3 or the resin film 6, 9, the fine pores are preferably non-penetrating holes from the aspect of container sealability. With pressure controlled between the first and second rolls, one or both of the penetrating holes and the non-penetrating holes can be formed.

[0051]

[2] Production method of polybutylene terephthalate laminate film

For instance, when the laminate 1 shown in Fig. 4 is formed, a long paper sheet 6 having a printed layer 5 on its entire surface is laminated with a PBT film 3 via a transparent adhesive layer 4 on a printed surface, and with a laminate film comprising a resin film 9, a light-blocking ink layer 10, a polyethylene layer 8 and a sealant film 11 via a polyethylene layer 8 on the other surface. The PBT film 3 may be provided with a transparent resin coating layer 2 in advance or after lamination. The transparent resin coating layer 2 may be formed by applying a coating composition comprising a transparent resin dissolved or dispersed in a proper solvent to the PBT film 3, and drying it. Though not particularly restricted, the coating method may be a roll-coating method, a bar-coating method, an air knife-coating method, a lip-coating method, blade-coating method, rod-coating method, gravure-coating method, etc. Each film 3, 6, 9 or 11 may be provided with a lot of substantially parallel linear scratches and/or a lot of fine pores in advance, if necessary. Each film 3, 6, 9 and 11 has an easy-to-tear direction aligned with the longitudinal direction of the long paper sheet 6.

[0052]

The laminate 1 comprising the transparent resin coating layer 2 formed by applying a transparent resin coating composition to the PBT film 3 and drying has curlability, presumably because the evaporation of a solvent by drying shrinks the transparent resin coating layer. When the PBT laminate film 1 with curlability is used for a lid heat-sealed to a container of instant food such as instant Chinese noodle, etc., the heat-sealed lid peeled from a container body is easily kept in a curled (standing) state, thereby making it easy to pour hot water into the container.

[0053]

Because the laminate 1 having a transparent resin coating layer 2 formed by applying and drying the above coating composition is curable toward the side of the PBT film 3, the laminate 1 is wound with the PBT film 3 outside, namely in an opposite direction to the curable direction, such that the laminate 1 is not substantially curled when unwound. Of course, the laminate 1 is gradually curled with time. Because the laminate 1 remains flat for some time, there is no trouble in punching and heat-sealing operations for forming the lid. When the heat-sealed lid is peeled from the container body, the lid is curled.

10 [0054]

When the PBT film 3 is laminated with the paper sheet 6, the resin PBT film 3 is preferably adhered to the paper sheet 6 while being stretched in an MD direction within an elastic deformation range (1 to 3% elongation), to provide the laminate 1 with curlability.

15 [0055]

To conduct the stretching of 1 to 3%, the PBT film 3 need only be subjected to a tension of 10 to 20 kgf/m (width). When the PBT film 3 is adhered to the paper sheet 6 without stretching, a tension applied to the PBT film 3 is 5 kgf/m (width) or less. Of course, a laminate 1 produced by adhering the PBT film 3 to the paper sheet 6 while applying tension to the PBT film 3 is wound with the PBT film 3 outside as described above.

[0056]

When the PBT film is used, its shape memory characteristics can be utilized to cause the laminate 1 to memorize a curled shape. To produce the PBT laminate film 1 memorizing a curled shape, for instance, two methods described in WO 2004/026577 may be used.

25 [0057]

The first method comprises (a) subjecting the PBT laminate film 1 to a

shaping treatment (cold-working) at a temperature T_1 equal to or lower than the glass transition temperature T_g of PBT while keeping a curled shape, (b) deforming the shaped PBT laminate film 1 to a flat shape at a temperature T_2 higher than T_g , and (c) cooling it to a temperature T_3 equal to or lower than T_g to fix it to the flat shape. The second method comprises (a) (i) subjecting the PBT laminate film 1 to a shaping treatment at a temperature T_4 higher than T_g and lower than the melting point of PBT while keeping a curled shape, or (ii) shaping the PBT film 3 at a temperature T_4 while keeping a curled shape, and then laminating it with a laminate film comprising a paper sheet 6 (a laminate comprising a layer constituting an inner layer 1b when forming the laminate 1 and a paper sheet 6) to produce the laminate 1 having a curled shape, (b) cooling the shaped laminate 1 to a temperature T_5 equal to or lower than T_g to fix it to a curled shape, (c) deforming the shaped laminate 1 to a flat shape at a temperature T_6 higher than T_g and lower than T_4 , and (d) cooling it to a temperature T_7 equal to or lower than T_g to fix it to a flat shape. Though the shape-memory PBT laminate film 1 produced by these methods has a flat shape, a lid formed from it by punching recovers curlability when heat-sealed, so that it is curled when peeled from a container body.

[0058]

After lamination as above, calendering may be conducted if necessary, to provide the laminate 1 with higher surface flatness and clarity.

Calendering may be carried out by a method of pressing a thermally softened transparent resin coating layer 2 by calendering rolls, such that a mirror-finished surface of the calendering roll is transferred to a surface of the transparent resin coating layer 2.

[0059]

After laminated as described above, embossing may be carried out depending on the applications of the laminate 1. Embossing may be carried

out, for instance, by a method of heat-pressing an embossing plate to the thermally softened transparent resin coating layer 2 using a reciprocal or rotary embossing machine, to transfer the surface roughness of the embossing plate to the transparent resin coating layer 2. The PBT film 3 may be embossed if
5 necessary. Because a polybutylene terephthalate (PBT) film has excellent dead-folding characteristics, which maintain a formed shape, it is much better in embossability than the other polyester films. Thus, the PBT film 3 can be provided with deep and fine embossing.

[0060]

10 Because the laminate 1 comprises a PBT film 3 having excellent embossability, the embossing of the laminate 1 can be as deep as about 1000 μm by selecting the thickness of the PBT film 3. Usually, the embossing depth of the laminate 1 is more preferably 100-500 μm , particularly 150-400 μm . The embossing temperature is preferably 100-160°C. When the
15 embossing temperature is higher than 160°C, the paper sheet 6 may be discolored. On the other hand, when the embossing temperature is lower than 100°C, embossing is insufficient. The embossing temperature is more preferably 110-140°C. The embossing pressure is preferably 196-2,942 kPa (2-30 kgf/cm^2), more preferably 490-2,452 kPa (5-25 kgf/cm^2). Roughness
20 patterns provided by the embossing may be wood grains, stone grains, textile textures, satin, sand, hairlines or their combinations.

[0061]

When the laminate 1 is used as a wall paper, the transparent resin coating layer 2 and the PBT film 3 are provided with the above fine
25 penetrating or non-penetrating holes after lamination, to impart gas permeability, moisture permeability, moisture-controlling function, etc. to the wall paper. In addition, when the PBT film 3 and/or the substrate 6 contain additives such as deodorants, anti-fungus agents, anti-mold agents, perfumes,

minus ion-generating agents, etc., the functions of these additives can be effectively exhibited. The penetrating or non-penetrating holes may have the above average distribution density.

[0062]

5 [3] Production method of lid

Pluralities of lids can be produced at one time by placing the laminate film 1 with the transparent resin coating layer 2 upward and conducting a punching and half-cutting operation with pluralities of punches. The punching and half-cutting operation per se can be carried out by known
10 methods.

[0063]

[4] Heat-sealing to container body

Explanation will be made on the heat-sealing of a lid constituted by the PBT laminate film 1 to a container body. After charging contents such as
15 dried noodle, powdered soup and ingredients into the container body, the lid is attached and heat-sealed to an upper flange of the container body with the sealant film layer 11 downward. To provide easy peelability, the heat-sealing temperature is preferably about 120 to 160°C.

[0064]

20 The punching operation and the heat-sealing operation to the container body may of course be conducted simultaneously. Particularly, because the shape-memory PBT laminate film 1 recovers a curled shape by heat-sealing, the punching operation and the heat-sealing operation to the container body are preferably conducted simultaneously.

25 [0065]

The container body may be formed by a paper, a synthetic resin such as styrene foam. The paper container body is advantageous in easy incineration, and free from gases adversely affecting environment when incinerated. The

styrene foam container body is advantageous in excellent temperature retention.

[0066]

The present invention will be explained in more detail referring to

5 Examples below without intention of restricting the present invention thereto.

[0067]

Example 1

The following materials were used.

PBT film: 40- μ m-thick, air-cooled inflation film of PBT resin ("Toraycon
10 1200S" available from Toray Industries, Inc., melting point: 220°C, glass transition temperature: 22°C).

Paper sheet: Having a basis weight of 100 g/m².

Polyvinyl alcohol: "GOHSENOL" available from The Nippon Synthetic Chemical Industry Co., Ltd.

15 Coating liquid: Containing polyvinyl alcohol and isopropyl alcohol at a ratio of 30/70 by mass.

[0068]

After dry-laminating the above PBT film via a transparent polyurethane adhesive to a surface of the above paper sheet, on which letters and design
20 were printed, a transparent adhesive ("ROCKBOND RU-40" available from Rock Paint Co., Ltd.) was coated on a surface of the PBT film. After drying the transparent adhesive layer, the above coating liquid was applied to the adhesive layer by a roll-coating method, and dried to a dry thickness of 2 μ m, to produce a polybutylene terephthalate laminate film. The photograph of the
25 resultant laminate is shown in Fig. 7.

[0069]

Comparative Example 1

A polybutylene terephthalate laminate film was produced in the same

manner as in Example 1 except for forming no transparent resin coating layer of polyvinyl alcohol. The photograph of the resultant laminate is shown in Fig. 8.

[0070]

5 It is clear from Figs. 7 and 8 that Example 1 (Fig. 7) was superior to Comparative Example 1 (Fig. 8) in the clarity of printed letters and design. This is presumably due to the fact that the formation of a transparent coating layer of polyvinyl alcohol on a PBT film surface eliminates the translucency of the PBT film from the laminate film.

10 EFFECT OF THE INVENTION

[0071]

A laminate comprising at least a polybutylene terephthalate film, a printed layer and a substrate, with a transparent resin coating layer formed on the polybutylene terephthalate film or the printed layer, has excellent clarity in printed letters and design. The polybutylene terephthalate laminate film of the present invention with such features can be suitably used for such applications as lid sheets of food containers, wrapping materials, wallpapers, paper container materials, etc. Food containers having lids formed by the polybutylene terephthalate laminate films of the present invention may contain various foods such as instant Chinese noodle, instant Yakisoba noodle, powdery stew, powdery or fluid soybean soup and Shiruko, which is a Japan-made, sweet soup of azuki-bean, other powdery or fluid soup, drinks such as juice, semi-solid foods such as jelly, pudding, etc.

25 [0072]

Though the present invention has been explained above referring to the drawings, the present invention is not restricted thereto, but can be variously modified unless deviating from the scope of the present invention.

WHAT IS CLAIMED IS:

1. A polybutylene terephthalate laminate film comprising at least a polybutylene terephthalate film and a substrate of a paper sheet or a resin film in this order from an upper surface, with a printed layer on an upper surface of
5 the substrate and/or a surface of said polybutylene terephthalate film, said laminate film further comprising a transparent resin coating layer on an upper surface of said polybutylene terephthalate film, or on an upper surface of the printed layer on said polybutylene terephthalate film.
2. The polybutylene terephthalate laminate film according to claim 1,
10 wherein said transparent resin is polyvinyl alcohol.
3. The polybutylene terephthalate laminate film according to claim 1 or 2, wherein said transparent resin coating layer is as thick as 1-5 μm .
4. The polybutylene terephthalate laminate film according to any one of claims 1-3, wherein it comprises at least said transparent resin coating layer,
15 said polybutylene terephthalate film, said printed layer, said substrate and a heat-sealable film in this order from an upper surface.
5. The polybutylene terephthalate laminate film according to any one of claims 1-4, wherein it comprises a transparent adhesive layer between said transparent resin coating layer and said polybutylene terephthalate film.
- 20 6. The polybutylene terephthalate laminate film according to any one of claims 1-5, wherein said transparent resin coating layer is formed by applying a coating composition comprising said transparent resin dissolved or dispersed in a solvent, and drying, and wherein said polybutylene terephthalate laminate film is provided with curlability by the shrinkage of said transparent resin
25 coating layer.
7. A lid sheet formed by the polybutylene terephthalate laminate film recited in any one of claims 4-6.
8. A food container comprising a lid formed by the lid sheet recited in claim 7.