#### (19) World Intellectual Property **Organization**

International Bureau





(43) International Publication Date 29 September 2005 (29.09.2005)

## (10) International Publication Number WO 2005/091213 A1

- (51) International Patent Classification<sup>7</sup>: G06K 19/077, H05K 3/02, B32B 31/00, G08B 13/24, H01Q 1/22
- (21) International Application Number:

PCT/GB2005/000999

- (22) International Filing Date: 16 March 2005 (16.03.2005)
- (25) Filing Language: English
- (26) Publication Language: English
- (30) Priority Data:

0405883.0 16 March 2004 (16.03.2004)

- (71) Applicant (for all designated States except US): DUPONT TEIJIN FILMS U.S. LIMITED PARTNERSHIP [US/US]; Barley Mill Plaza, Bldg. 22, Lancaster Pike & Route 141, P.O. Box 80022, Wilmington, DE 19880-0022 (US).
- (72) Inventors; and
- (75) Inventors/Applicants (for US only): EVANS, Ken [GB/GB]; 2 Ryedale Close, Yarm, Stockton-on-Tees TS15 9UN (GB). SANKEY, Stephen, William [GB/GB]; 5 Hambleton Court, Great Smeaton, Northallerton DL6 2HR (GB). TURNER, Dave [GB/GB]; 15 Meadow Vale, Northumberland Park, Shiremoor, Newcastle-upon-Tyne NE27 OBD (GB). POLACK, Natalie [GB/GB]; 29 Pinewood Road, Eaglescliffe, Stockton-on-Tees TS16 OAJ (GB). KOSUGE, Masahiko [JP/US]; 4718 Valley Overlook Drive, Apt. 302, Midlothian, VA 23112 (US). NUGARA, Peter, Neil [US/US]; 11760 Wexwood Drive, Richmond, VA 23236 (US).

- (74) Agents: COCKERTON, Bruce, Roger et al.; Carpmaels & Ransford, 43-45 Bloomsbury Square, London WC1A 2RA (GB).
- (81) Designated States (unless otherwise indicated, for every kind of national protection available): AE, AG, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BW, BY, BZ, CA, CH, CN, CO, CR, CU, CZ, DE, DK, DM, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ, NA, NI, NO, NZ, OM, PG, PH, PL, PT, RO, RU, SC, SD, SE, SG, SK, SL, SM, SY, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, YU, ZA, ZM, ZW.
- (84) Designated States (unless otherwise indicated, for every kind of regional protection available): ARIPO (BW, GH, GM, KE, LS, MW, MZ, NA, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European (AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HU, IE, IS, IT, LT, LU, MC, NL, PL, PT, RO, SE, SI, SK, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

#### Published:

- with international search report
- before the expiration of the time limit for amending the claims and to be republished in the event of receipt of amendments

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

(54) Title: POLYMERIC FILM SUBSTRATE FOR USE IN RADIO-FREQUENCY RESPONSIVE TAGS

(57) Abstract: A radio-frequency (RF) responsive tag comprising a heat-sealing substrate comprising a polyester layer, and an antenna comprising a patter of conductive material wherein said conductive material is in direct contact with a heat-sealing surface of the substrate, and wherein the shrinkage of the heat-sealing substrate is less than 5% at 190°C over 30 minutes; a method of manufacture of said RF-response tag.

1

# POLYMERIC FILM SUBSTRATE FOR USE IN RADIO-FREQUENCY RESPONSIVE TAGS

This invention relates to a composite film useful as a substrate for radio-frequency responsive tags, including radio-frequency tags (RF tags) used for electronic article surveillance and radio-frequency identification tags (RFID tags), and to the composite structure comprising the substrate and the radio-frequency functional components, and to a process for the production thereof.

10 Radio-frequency (RF) communication systems are finding valuable uses in anti-theft, anticounterfeit and authentication security devices, as well as in control systems for the
storage, movement, maintenance, tracking and sorting of goods or stock. Specific
applications include washable RF-responsive tags which can be sewn into clothing; RFresponsive tags in smart cards and personal identification cards; RF-responsive tags in
medical equipment and supplies; RF-responsive tags in smart labels for logistics and
supply chain applications; and RF-responsive tags for embedding into bank notes. RFresponsive devices can be used to carry unique data, for instance: (i) identifier data, in
which a numeric or alphanumeric string is stored for identification purposes or as an access
key to data stored elsewhere in a computer or information management system; and (ii)
portable data files, in which information can be organised, for communication or as a
means of initiating actions without recourse to, or in combination with, data stored
elsewhere.

The technology uses radio-waves to communicate with an RF-responsive device without the requirement of direct contact or line-of-sight. The RF-responsive device functions by retransmitting or by reflecting or otherwise disrupting a radio frequency signal. There are two main classes of RF-responsive tags.

The first class, referred to herein as "radio-frequency tags" (RF tags), are primarily used for electronic article surveillance (EAS), and typically as anti-theft devices. When the tags are passed through a surveillance zone, which is created by a transmitter sending out defined frequencies to a receiver, the tags create a disturbance in the surveillance field

2

which is detected by the receiver. These types of RF tags typically comprise as essential components a substrate and an antenna such as a metal pattern or coil.

The second class of RF-responsive tags, referred to herein as "radio-frequency identification tags" (RFID tags), comprise not only an antenna and a substrate but also a data-carrying means which is electronically programmable with unique information. Thus, the tag may comprise a microchip or integrated circuit. There are also applications using chip-less tags, and such tags may comprise data-carrying electronic components such as thin-film transistors (TFTs), electromagnetic micro-wires having controlled surface vs bulk characteristics designed to maximise the so-called Barkhausen electromagnetic effect, and components using programmable magnetic resonance technology (PMR) in an acoustic-magnetic detection system.

A typical radio-frequency identification system comprises an antenna; a transceiver (with decoder); and a transponder (the RF-responsive tag). The antenna emits radio signals to activate the tag and read and/or write data to it. The transceiver controls data acquisition and communication, and typically interfaces with an information management system. The antenna may be packaged with the transceiver and decoder to become a reader, which can be configured either as a handheld or a fixed-mount device. The reader emits radio waves in ranges of anywhere from a few centimetres to 30 metres or more, depending upon its power output and the radio frequency used. When the tag passes through the electromagnetic zone, it detects the reader's activation signal. The reader decodes the data encoded in the tag's integrated circuit and the data is passed to the host computer (information management system) for processing.

25

Radio-frequency identification systems operate at various frequency ranges. Low-frequency (typically 30 KHz to 500 KHz) systems have short reading ranges and lower system costs, and are most often used for security access and local asset tracking applications. High-frequency (typically 13.56 MHz) systems are often used in smart cards, libraries, laundries, and track and trace applications. Ultra-high frequency (typically 168 – 950 MHz) and microwave frequency (> 2.4 GHz) offer longer read ranges (greater than 30 metres) and higher reading speeds and are of growing interest in some applications.

RF-responsive tags may be categorized as either active or passive. Active tags are powered by an internal battery and are typically read/write. In a typical read/write work-in-process system, a tag might give a machine a set of instructions, and the machine would then report its performance to the tag, the encoded data then becoming part of the tagged part's history. The battery-supplied power of an active tag generally gives it a longer read range, but such tags are of greater size, greater cost, and have a limited operational life. Passive tags operate without an internal power source, i.e. they have no battery, and obtain operating power from the initial radio signal to transmit a response. Passive tags are consequently much lighter than active tags, less expensive, and offer a virtually unlimited operational lifetime, although they have shorter read ranges and require a higher-powered reader. Read-only tags are typically passive and are programmed with a unique set of data (usually 32 to 256 bits on a chip tag, and less on a chip-less tag) that cannot be modified. Read-only tags most often operate in combination with a database containing modifiable product-specific information, in the same way as a barcóde.

An RF-responsive tag may comprise analogue circuitry (including the antenna, and for instance a capacitor) for data transfer and power supply, and in chip tags, a digital lowpower integrated circuit (or microchip), and optionally a battery. Typical transponders are 20 described in, for example, US Patents 5,541,399, 4,730,188 and 4,598,276. In chip tags, the microprocessor interfaces with the transponder memory, which may comprise readonly (ROM), random access (RAM) and non-volatile programmable memory (typically electrically erasable programmable read only memory (EEPROM)) for data storage depending upon the type and sophistication of the device. ROM-based memory is used to 25 accommodate security data and the transponder operating system instructions which, in conjunction with the processor or processing logic, deals with the internal functions such as response delay timing, data flow control and power supply switching. RAM-based memory is used to facilitate temporary data storage during transponder interrogation and response. Non-volatile programmable memory is used to store the transponder data and 30 ensures that data are retained when the device is in a quiescent or power-saving state. The transponder antenna is the means by which the device senses the interrogating field (and, where appropriate, the programming field) and also serves as the means of transmitting the transponder response to interrogation. The antenna typically consists of a coil of wire or a

4

conductive pattern, typically aluminium, copper or silver, disposed on a dielectric or insulating substrate. The antenna may be formed in a number of ways, for instance by stamping or embossing the metal component onto the substrate; by etching the conductive pattern in a foil substrate; by using conductive paints, inks or pastes; by electroless or 5 electroplating onto a metallic seeding ink (for instance in accordance with the technology of RT circuits); or by bonding pre-cut patterns onto the substrate. In one process, an antenna is formed by screen-printing a substrate with silver ink, and optionally copperplating over the ink. Typically, an antenna is formed by etching, or printing a silver ink. Interface circuitry between antenna and microchip directs and accommodates the 10 interrogation field energy for powering purposes in passive transponders and triggering of the transponder response. One problem with the use of conventional substrates, particularly those used for the manufacture of RF-responsive tags in which the conductive pattern of the antenna is formed using conductive inks or pastes, has been flex-cracking of the conductive pattern (which can adversely affect the range and functioning of the antenna, as 15 well as the adhesion strength to the conductive pattern of other components of the RFresponse tag which are mounted over the conductive pattern).

As used herein, the term "RF-responsive tag" refers to an article which reacts to a radio-frequency signal and which comprises as essential functional components a substrate and an antenna, and optionally further comprises a battery, and/or a data carrying means such as an integrated circuit. The term "RF-responsive tag" includes both RF tags and RFID tags, as defined hereinabove. As used herein the term "antenna" refers to a metal pattern or coil receptive to radio-frequencies.

Previously, RF-responsive tags have been manufactured by providing a polymeric film substrate, and adhering one or more of the transponder components thereto, including the antenna component, by using a layer of adhesive. The adhesive layer is typically coated off-line, i.e. in a process step which is separate from the step of manufacture of the polymeric film substrate and which involves the use of hazardous and environmentally unacceptable solvents. The adhesive layer is typically 20 to 30 microns thick in the prior art RF-responsive tags. It is an object of this invention to provide a more economical and thinner RF-responsive tag, which may also be flexible. It is also an object of this invention to provide good delamination resistance between the polymeric film substrate and the

antenna component. It is also an object of this invention to provide such an RF-responsive tag (or a composite film comprising a substrate and the conductive pattern suitable as a precursor in the manufacture of the RF-responsive tag) which exhibits reduced flex-cracking of the conductive pattern, particularly wherein the conductive pattern is formed using conductive inks or pastes.

According to the present invention, there is provided an RF-responsive tag comprising a heat-sealing substrate comprising a polyester layer, and an antenna comprising a pattern of conductive material wherein said conductive material is in direct contact with a heat-sealing surface of the substrate, and optionally a data-carrying means in electrical communication with the antenna, and wherein the shrinkage of the heat-sealing substrate is less than 5% at 190°C over 30 minutes.

The conductive material of the antenna is preferably metallic, and is preferably selected from metals such as copper, aluminium, silver, gold, zinc, nickel and tin, preferably copper, aluminium, silver and gold, preferably aluminium and copper, and preferably copper.

The RF-responsive tag of the present invention is advantageous in that it dispenses with the need for a layer of adhesive between the antenna and the substrate, and is therefore more economical, efficient and environmentally acceptable to produce, and the RF-responsive tag may also be made thinner.

The substrate is a self-supporting film or sheet by which is meant a film or sheet capable of independent existence in the absence of a supporting base. The substrate is preferably uniaxially or biaxially oriented, preferably biaxially oriented. The substrate comprises a polyester film. Linear polyesters are preferred. Suitable polyesters include those derived from one or more dicarboxylic acids, such as terephthalic acid, isophthalic acid, phthalic acid, 1,4-, 2,5-, 2,6- or 2,7-naphthalenedicarboxylic acid, succinic acid, sebacic acid, adipic acid, azelaic acid, 1,10-decanedicarboxylic acid, 4,4'-diphenyldicarboxylic acid, hexahydro-terephthalic acid or 1,2-bis-p-carboxyphenoxyethane (optionally with a monocarboxylic acid, such as pivalic acid), and from one or more glycols, particularly an aliphatic or cycloaliphatic glycol, such as ethylene glycol, 1,3-propanediol, 1,4-butanediol,

6

neopentyl glycol, diethylene glycol and 1,4-cyclohexanedimethanol. An aliphatic glycol is preferred. A preferred substrate polyester is selected from polyethylene terephthalate (PET) and polyethylene naphthalate (PEN). PET or a copolyester thereof is particularly preferred.

5

15

The substrate layer may be a monolayer substrate or may be a multilayer substrate, the functional requirement being that it is heat-sealable to the conductive material of the antenna. As used herein, the term "heat-sealing substrate" therefore refers to a substrate layer which has been heat-sealed to the conductive material of the antenna. Similarly, the 10 term "RF-responsive tag comprising a heat-sealing substrate and an antenna" refers to an RF-responsive tag in which an adhesive heat-seal bond has been formed between a heatsealable substrate and the antenna. Similarly, the term "composite film comprising a layer of conductive material and a heat-sealing substrate" refers to a composite film in which a heat-seal bond has been formed between the heat-sealable substrate and the conductive material.

The heat-sealable polymeric material should soften to a sufficient extent that its viscosity becomes low enough to allow adequate wetting for it to adhere to the surface to which it is being bonded. The heat-sealable polymer is preferably a copolyester derived from one or 20 more of the dicarboxylic acid(s) or their lower alkyl diesters with one or more of the glycol(s) referred to herein.

In one embodiment, the substrate comprises one layer, hereinafter referred to as Embodiment A, wherein the polyester is selected from those recited hereinabove and is 25 preferably selected from a copolyester derived from an aliphatic glycol and at least two dicarboxylic acids, particularly aromatic dicarboxylic acids. Preferably, the dicarboxylic acids are terephthalic acid and one other dicarboxylic acid which is preferably an aromatic dicarboxylic acid, and preferably isophthalic acid. A preferred copolyester is derived from ethylene glycol, terephthalic acid and isophthalic acid. The preferred molar ratios of the 30 terephthalic acid component to the isophthalic acid component are in the range from 50:50 to 90:10, preferably in the range from 65:35 to 85:15. In a preferred embodiment, this copolyester is a copolyester of ethylene glycol with about 82 mole % terephthalate and about 18 mole % isophthalate.

In a further embodiment, hereinafter referred to as **Embodiment B**, the substrate comprises more than one layer, provided that at least one of the layers (i.e. the layer adjacent the metallic antenna layer) is heat-sealable. In this embodiment, the layer(s) other than the heat-sealable layer adjacent the metallic antenna layer is/are referred to herein as "base layer(s)" of the substrate layer.

In embodiment B, the base layer may be any layer compatible with the heat-sealable polymer, in order to provide adequate interlayer adhesion. The base layer is preferably a synthetic linear polyester selected from those mentioned herein above, particularly a polyester derived from one dicarboxylic acid, preferably an aromatic dicarboxylic acid, preferably terephthalic acid or naphthalenedicarboxylic acid, more preferably terephthalic acid, and one glycol, particularly an aliphatic or cycloaliphatic glycol, preferably ethylene glycol. PET or PEN, particularly PET, is particularly preferred as the base layer, particularly for the embodiments B1, B2, B3 and B4 described hereinbelow.

In one preferred embodiment, hereinafter referred to as **Embodiment B1**, the heat-sealable layer comprises a copolyester derived from an aliphatic glycol and two or more dicarboxylic acids, preferably two or more aromatic dicarboxylic acids. Preferably, the dicarboxylic acids are terephthalic acid and one other dicarboxylic acid, preferably one other aromatic dicarboxylic acid, and preferably isophthalic acid. A preferred copolyester is derived from ethylene glycol, terephthalic acid and isophthalic acid. The preferred molar ratios of the terephthalic acid component to the isophthalic acid component are in the range of from 50:50 to 90:10, preferably in the range from 65:35 to 85:15. In a preferred embodiment, this copolyester is a copolyester of ethylene glycol with about 82 mole % terephthalate and about 18 mole % isophthalate.

In an alternative preferred embodiment, hereinafter referred to as **Embodiment B2**, the copolyester of the heat-sealable layer comprises an aromatic dicarboxylic acid and an aliphatic dicarboxylic acid. A preferred aromatic dicarboxylic acid is terephthalic acid. Preferred aliphatic dicarboxylic acids are selected from sebacic acid, adipic acid and azelaic acid. The concentration of the aromatic dicarboxylic acid present in the copolyester is preferably in the range from 45 to 80, more preferably 50 to 70, and

particularly 55 to 65 mole % based on the dicarboxylic acid components of the copolyester. The concentration of the aliphatic dicarboxylic acid present in the copolyester is preferably in the range from 20 to 55, more preferably 30 to 50, and particularly 35 to 45 mole % based on the dicarboxylic acid components of the copolyester. Particularly 5 preferred examples of such copolyesters are (i) copolyesters of azeleic acid and terephthalic acid with an aliphatic glycol, preferably ethylene glycol; (ii) copolyesters of adipic acid and terephthalic acid with an aliphatic glycol, preferably ethylene glycol, preferably butylene glycol. Preferred polymers include a copolyester of sebacic acid/terephthalic acid/butylene glycol (preferably having the components in the relative molar ratios of 45-55/55-45/100, more preferably 50/50/100) having a glass transition point (Tg) of -30°C and a melting point (Tm) of 117°C), and a copolyester of azeleic acid/terephthalic acid/ethylene glycol (preferably having the components in the relative molar ratios of 40-50/60-50/100, more preferably 45/55/100) having a Tg of -15°C and a Tm of 150°C.

15

In an alternative embodiment, hereinafter referred to as **Embodiment B3**, the heat-sealable layer comprises a copolyester derived from an aliphatic diol and a cycloaliphatic diol with one or more, preferably one, dicarboxylic acid(s), preferably an aromatic dicarboxylic acid. Examples include copolyesters of terephthalic acid with an aliphatic diol and a cycloaliphatic diol, especially ethylene glycol and 1,4-cyclohexanedimethanol. The preferred molar ratios of the cycloaliphatic diol to the aliphatic diol are in the range from 10:90 to 60:40, preferably in the range from 20:80 to 40:60, and more preferably from 30:70 to 35:65. In a preferred embodiment this copolyester is a copolyester of terephthalic acid with about 33 mole % 1,4-cyclohexane dimethanol and about 67 mole % ethylene glycol. An example of such a polymer is PETG<sup>TM</sup>6763 (Eastman) which comprises a copolyester of terephthalic acid, about 33% 1,4-cyclohexane dimethanol and about 67% ethylene glycol and which is always amorphous. In an alternative embodiment, the heat-sealable layer polymer may comprise butane diol in place of ethylene glycol.

Formation of the copolyesters is conveniently effected in known manner by condensation, or ester-interchange, at temperatures generally up to 275°C.

PCT/GB2005/000999

In a further alternative embodiment, hereinafter referred to as **Embodiment B4**, the heat-sealable layer comprises an ethylene vinyl acetate (EVA). Suitable EVA polymers may be obtained from DuPont as Elvax<sup>TM</sup> resins. Typically, these resins have a vinyl acetate content in the range of 9% to 40%, and typically 15% to 30%.

5

The thickness of the heat-sealable layer in embodiment B is generally between about 1 and 30%, preferably about 10 and 20% of the thickness of the substrate. The heat-sealable layer may have a thickness of up to about 25μm, more preferably up to about 20μm, more preferably up to about 15μm, more preferably up to about 10μm, more preferably between about 0.5 and 6μm, and more preferably between about 0.5 and 4μm. The overall thickness of the substrate is preferably up to about 350 μm, more preferably up to about 100 μm, more preferably up to about 75 μm, more preferably between about 12 and 100 μm, and more preferably between about 20 and 75 μm.

Preferably, the substrate exhibits a heat-seal strength to itself of at least 300 g/25mm<sup>2</sup>, preferably from about 400 g/25mm<sup>2</sup> to about 1000 g/25mm<sup>2</sup>, and more preferably from about 500 to about 850 g/25mm<sup>2</sup>.

Preferably, the substrate exhibits a heat-seal strength to the metallic layer of at least about 200 g/25mm<sup>2</sup>, preferably at least about 400 g/25mm<sup>2</sup>, preferably at least about 600 g/25mm<sup>2</sup>, and preferably at least about 800 g/25mm<sup>2</sup>. Typical bond strengths are in the range of from about 400 to about 1000 g/25mm<sup>2</sup>. The bond strength to the metal should be high enough so that the film destructs if attempts are made to separate the metallic antenna from the polymeric substrate. In one embodiment, the adhesive strength of the substrate to 25 the metallic layer exceeds the Ultimate Tensile Strength (UTS) of the substrate.

The substrate exhibits a low shrinkage, and preferably less than 3% at 190°C over 30 minutes, preferably less than 2%, preferably less than 1%, and preferably less than 0.5%, preferably less than 0.2%.

3.0

Formation of the substrate may be effected by conventional techniques well-known in the art. Conveniently, formation of the substrate is effected by extrusion, in accordance with the procedure described below. In general terms the process comprises the steps of

extruding a layer of molten polymer, quenching the extrudate and orienting the quenched extrudate in at least one direction.

The substrate may be uniaxially oriented, but is preferably biaxially oriented by drawing in two mutually perpendicular directions in the plane of the film to achieve a satisfactory combination of mechanical and physical properties. Orientation may be effected by any process known in the art for producing an oriented film, for example a tubular or flat film process. A flat film process may involve either sequential or simultaneous drawing.

In the preferred flat film process, the substrate-forming polyester is melted and extruded through a slot die and rapidly quenched onto a chilled casting drum to ensure that the polyester retains the disordered, amorphous structure of the melt. Orientation on the molecular scale is then effected by reheating the extrudate or cast film above its glass transition temperature (Tg) and stretching it in at least one direction. Typically, stretching will be carried out on film whose temperature has been raised to between 70 and 150°C in the case of polyethylene terephthalate (PET). For polyethylene naphthalate (PEN), higher temperatures are required, typically between 110 and 170°C. As a general rule, the preferred stretching temperatures are in the range of from about (Tg+10°C) to about (Tg+60°C).

20

Biaxial orientation may be produced by stretching sequentially a flat, quenched extrudate firstly in one direction, usually the longitudinal direction or forward, machine direction (MD) of the process, and then in the transverse direction (TD). Forward stretching of the cast film is conveniently performed over a set of rotating rolls which are driven at different speeds. Although the details of this process step may vary, the principle of the technology, which is to heat and accelerate the cast film in the process direction, is characteristic of all designs. Transverse stretching is then performed in a stenter oven. In the stenter stage of the process, the edges of the film are gripped by clips and led along rails, which provide support during a reheating step and then diverge to cause the material to be stretched for a second time. Alternatively, the cast film may be stretched simultaneously in both the forward and transverse directions in a biaxial stenter. Stretching is performed to an extent determined by the nature of the polyester, for example PET is usually stretched so that the MD and/or TD dimensions of the oriented film are from 2 to 5, more preferably 2.5 to 4.5

11

times, that of its original dimension. Greater draw ratios (for example, up to about 8 times) may be used if orientation in only one direction is required. It is not necessary to stretch equally in the machine and transverse directions although this is preferred if balanced properties are desired.

5

In the preferred flat film process, the final stage involves stabilising the stretched film by heat-setting, still at the elevated temperatures of the stenter oven and under a controlled dimensional restraint. The film is heated at a temperature above its glass transition temperature but below the melting temperature thereof, to enable crystallisation of the 10 polyester. Some dimensional relaxation (or "toe-in") in either MD or TD or both is permitted at this stage to improve further the final thermal shrinkage or dimensional stability of the finished film. Relaxation of the film in the transverse direction is carried out by converging the paths of the clips holding the film in the stenter. In a sequential stretching process, the relaxation in MD is made possible when the winding speed of the 15 film is lower than its exit speed from the stenter. A simultaneous biaxial stretching process allows for longitudinal (MD) relaxation inside the stenter by the controlled deceleration of the linear motor-driven clips during or after heat-setting so that the speed of the film exiting the stenter oven is slower than the maximum speed within the stenter frame. In applications where dimensional stability is not of significant concern, the film may be heat-20 set at relatively low temperatures or not at all. In contrast, as the temperature at which the film is heat-set is increased, other properties such as elongation to break and tear-resistance may change. Thus, the actual heat-set temperature and time will be chosen depending on the composition of the film and the balance of final properties desired, as appropriate to the end-use application of the film. Within these constraints, the maximum temperature of the 25 film passing through the heat-set stage of the process will generally be from about 135° to about 250°C, as described in GB-A-838708. The film is then cooled under controlled tension and temperature and wound into rolls.

An optional step in the manufacture of the substrate is to subject it to further heat-30 stabilisation by heating it under minimal physical restraint at a temperature above the glass transition temperature of the polyester but below the melting point thereof, in order to allow the majority of the inherent shrinkage in the film to occur (relax out) and thereby produce a film with much lower residual shrinkage and consequently higher dimensional

stability. The film shrinkage or relaxation which occurs during the further heat-stabilisation stage is effected either by controlling the line tension experienced by the film at elevated temperature or by controlling the line-speed. The tension experienced by the film is a low tension and typically less than 5 kg/m, preferably less than 3.5 kg/m, more preferably in 5 the range of from 1 to about 2.5 kg/m, and typically in the range of 1.5 to 2 kg/m of film width. For a relaxation process which controls the film speed, the reduction in film speed (and therefore the strain relaxation) is typically in the range 0 to 2.5 %, preferably 0.5 to 2.0%. There is no increase in the transverse dimension of the film during the heatstabilisation step. The temperature to be used for the heat stabilisation step can vary 10 depending on the desired combination of properties from the final film, with a higher temperature giving better, i.e. lower, residual shrinkage properties. A temperature of 135°C to 250°C is generally desirable, preferably 150 to 230°C, more preferably 170 to 200°C. The duration of heating will depend on the temperature used but is typically in the range of 10 to 40 sec, with a duration of 20 to 30 secs being preferred. This heat stabilisation 15 process can be carried out by a variety of methods, including flat and vertical configurations and either "off-line" as a separate process step or "in-line" as a continuation of the film manufacturing process. In one embodiment, heat stabilisation is conducted "offline". The heat-stabilisation step promotes very low shrinkage, typically less than 1% over 30 minutes in an oven at 190°C, particularly less than 0.5%, and particularly less than 20 0.2%. The heat-stabilisation step is particularly suitable in the manufacture of coated multilayer substrates such as Embodiments B2 and B4, and would be conducted on the base layer prior to the off-line coating of the heat-sealable layer.

Formation of a multi-layer substrate comprising a heat-sealable layer and a base layer may

25 be effected by conventional techniques. The method of formation of the multi-layer

substrate will depend on the identity of the heat-sealable layer. Conventional techniques

include casting the heat-sealable layer onto a preformed base layer. Conveniently,

formation of the heat-sealable layer and the base layer is effected by coextrusion, and this

is suitable for Embodiments B1 and B3 described herein. Other methods of forming the

multi-layer substrate include coating the heat-sealable polymer onto the base layer, and this

technique would be suitable for Embodiments B2 and B4 described herein. Coating may

be effected using any suitable coating technique, including gravure roll coating, reverse

roll coating, dip coating, bead coating, extrusion-coating, melt-coating or electrostatic

spray coating. Coating may be conducted "off-line", i.e. after the stretching, heat-setting and optional heat-stabilisation steps employed during manufacture of the substrate, or "in-line", i.e. wherein the coating step takes place before, during or between any stretching operation(s) employed. In one embodiment, the coating of the heat-sealing layer is conducted off-line. Prior to application of a heat-sealable layer onto the base layer, the exposed surface of the base layer may, if desired, be subjected to a chemical or physical surface-modifying treatment to improve the bond between that surface and the subsequently applied layer. For example, the exposed surface of the base layer may be subjected to a high voltage electrical stress accompanied by corona discharge.

10 Alternatively, the base layer may be pretreated with an agent known in the art to have a solvent or swelling action on the base layer, such as a halogenated phenol dissolved in a common organic solvent e.g. a solution of p-chloro-m-cresol, 2,4-dichlorophenol, 2,4,5- or 2,4,6-trichlorophenol or 4-chlororesorcinol in acetone or methanol.

In one preferred embodiment, the substrate is a multilayer coextruded substrate comprising a heat-sealable layer and a base layer, preferably according to embodiments B1 and B3. In this embodiment in particular, the thickness of the heat-sealable layer is preferably from about 10 to about 20% of the thickness of the substrate, and preferably up to about 20μm preferably thinner as described herein.

20

In a further preferred embodiment, the substrate is a multilayer coated substrate comprising a heat-sealable layer and a base layer, preferably according to embodiments B2 and B4, preferably according to embodiment B2, and particularly wherein said base layer is heat-stabilised as described herein.

25

The antenna may be formed on the substrate by a conventional method, for instance according to a method as described hereinabove, and comprises the step of contacting the metallic material of the antenna with a heat-sealable surface of the substrate under conditions of elevated temperature (i.e. at a temperature above room temperature at which the polymeric material of the heat-sealable layer softens to an extent sufficient to adhere the metallic layer), and optionally pressure. In one embodiment, metal wire in a preformed configuration may be heat-sealed to the substrate. In a further embodiment, a metallic foil is laminated to a heat-sealable surface of the substrate by contacting the foil

with the heat-sealable surface of the substrate under elevated temperature and optionally pressure. The conductive pattern of the antenna is then produced by a conventional technique, such as etching. Techniques for etching conductive patterns onto a substrate are well-known in the art and are disclosed for instance in "The Art of Electronics" by Horowitz and Hill (2<sup>nd</sup> Edition, 1989, Cambridge University Press; Section 12.04) and also in US-6623844, US- 6621153 and US-2002/015002-A, the disclosures of which are incorporated herein by reference. In one embodiment of an etching process, once the metallic layer has been applied to the substrate, an etching-resist pattern is applied to the metallic layer, for instance by printing a suitable ink on the surface of the metallic layer in the shape of the desired conductive pattern. Any suitable printing technique may be used, for instance gravure printing. The etching-resist ink may need to be cured, for instance by heat or UV irradiation, in order to ensure that it is adhered to the underlying metallic layer sufficiently strongly to withstand the subsequent etching step. Next the substrate/metallic layer/resist pattern laminate is then etched using a suitable reagent to form the desired conductive pattern. For instance, the removal of the exposed portions of a copper layer may be effected using a solution of iron chloride FeCl<sub>2</sub> at around 50 °C. The final step in the process is the removal of the material of the resist pattern by a suitable chemical reagent to leave the metallic conductive pattern imprinted on the substrate. In a second embodiment of an etching process, a liquid or dry film resist (such as Riston® from 20 DuPont) is applied in the form of a continuous coating or layer to the metallic layer. A photographic film with a negative image of the conductive pattern (a "photo-tool") is then superposed over the substrate/metallic layer/photo-resist laminate, and the layer of photoresist is then exposed through the negative using UV light. The exposed areas of the photoresist are thereby cross-linked or otherwise chemically changed. A developer is then used 25 to remove the unchanged regions of the photo-resist, to leave a protected positive pattern on the copper substrate. The laminate is then etched, and the final step is the removal of the remaining photo-resist to leave the metallic conductive pattern imprinted on the substrate.

The thickness of the conductive metallic pattern is typically between about 2 and 100 μm, and particularly between about 10 and 50 μm, although thicknesses of less than 10 μm are becoming more common.

The antenna may be electrically connected to an optional data-carrying means, such as an integrated circuit, by conventional means, for instance using solder or conductive adhesive. If necessary, the data-carrying means may be affixed to the substrate using additional adhesive (including pressure sensitive adhesive and non-conductive adhesive).

5

The RF-responsive tag may comprise further optional layers. In RF-responsive tags where an integrated circuit is required to be located substantially over the antenna, an insulating layer may be disposed over at least part of the antenna. A cover layer may be present over the antenna and integrated circuit, and may be formed from any suitable layer-forming or 10 film-forming material, including the polyester film described herein. The cover may be printable and optionally comprises an ink-receptive layer. The surface of the substrate opposite the surface on which is disposed the antenna may comprise a layer of adhesive, optionally with a cover or release layer that may be peeled away when the RF-responsive tag is to be affixed to an article. In an alternative embodiment, the RF-responsive tag may 15 be affixed to an article by formation of a heat-seal bond. In that embodiment, a mono-layer substrate may itself be capable of forming a heat-seal bond to the article, or an additional heat-sealable layer may be present. A multilayer substrate comprising a base layer and on a first surface thereof a first heat-sealable layer for bonding to the antenna, as described herein, may comprise a second heat-sealable layer on the second surface thereof for 20 forming a heat-seal bond to an article, wherein the second heat-sealable layer may be the same as or different to the first heat-sealable layer.

According to a further aspect of the present invention, there is provided a method of manufacture of an RF-responsive tag comprising a substrate, an antenna comprising a pattern of conductive material, and optionally a data-carrying means, said method comprising the following steps:

- (i) providing a heat-sealable substrate comprising a polyester layer and wherein the shrinkage of said substrate is less than 5% at 190°C over 30 minutes;
- (ii) disposing the conductive material of the antenna directly onto at least part of a heat sealable surface of the substrate;
  - (iii) effecting heat-sealing between the heat-sealable substrate and the conductive material;

16

(iv) optionally providing a data-carrying means in electrical communication with the conductive material.

Where the conductive material of the antenna has not been pre-formed into the conductive pattern of the antenna, step (ii) in the process defined above is the first stage of antenna formation, the second stage of antenna formation being the step of forming a pattern in the conductive material, this second stage being carried out after heat-sealing step the conductive material to the substrate (step (iii) in the process defined above). Formation of the conductive pattern may be effected by a conventional method as described herein, typically by an etching process comprising the steps of forming an etching-resist having a wiring pattern on the surface of the conductive layer, forming a conductive pattern on the surface of the substrate by etching, and removing the resist.

According to a further aspect of the invention, there is provided the use of a heat-sealing film comprising a polyester layer wherein the shrinkage of said film is less than 5% at 190°C over 30 minutes, as described herein, as a substrate in the manufacture of an RF-responsive tag comprising said film as a substrate, an antenna comprising a pattern of conductive material, and optionally a data-carrying means in electrical communication with the antenna, wherein the conductive material is in direct contact with a heat-sealing surface of the film.

According to a further aspect of the invention, there is provided the use of a heat-sealing film comprising a polyester layer wherein the shrinkage of said film is less than 5% at 190°C over 30 minutes, as described herein, for the purpose of reducing flex-cracking in a composite film suitable as a precursor in the manufacture of an RF-responsive tag, said composite film comprising a layer of conductive material and said heat-sealing film wherein the conductive material is in direct contact with a heat-sealing surface of the substrate, and particularly wherein said layer of conductive material is formed using conductive inks or pastes, and particularly wherein said heat-sealing film is a coated film such as those according to embodiments B2 and B4. The RF-responsive tag comprises said composite film as a substrate, an antenna comprising a pattern of conductive material, and optionally a data-carrying means in electrical communication with the antenna.

According to a further aspect of the invention, there is provided the use of a composite film comprising a layer of conductive material and a heat-sealing substrate comprising a polyester layer wherein the shrinkage of said heat-sealing substrate is less than 5% at 190°C over 30 minutes, as described herein, as a precursor in the manufacture of an RF-5 responsive tag comprising said heat-sealing substrate, an antenna comprising a pattern of said conductive material, and optionally a data-carrying means in electrical communication with the antenna, wherein the conductive material is in direct contact with a heat-sealing surface of the substrate.

10 According to a further aspect of the invention, there is provided a composite film suitable as, or for use as, a precursor in the manufacture of an RF-responsive tag, said composite film comprising a layer of conductive material and a heat-sealing substrate comprising a polyester layer wherein the shrinkage of the heat-sealing substrate is less than 5% at 190°C over 30 minutes, as defined herein, wherein the conductive material is in direct contact with a heat-sealing surface of the substrate.

One or more of the layers of the substrate may conveniently contain any of the additives conventionally employed in the manufacture of polymeric films. Thus, agents such as cross-linking agents, dyes, pigments, voiding agents, lubricants, anti-oxidants, radical 20 scavengers, UV absorbers, thermal stabilisers, anti-blocking agents, surface active agents, slip aids, optical brighteners, gloss improvers, prodegradents, viscosity modifiers and dispersion stabilisers may be incorporated as appropriate. In particular the substrate may comprise a particulate filler which may, for example, be a particulate inorganic filler or an incompatible resin filler or a mixture of two or more such fillers. Such fillers are wellknown in the art.

Particulate inorganic fillers include conventional inorganic fillers, and particularly metal or metalloid oxides, such as alumina, silica (especially precipitated or diatomaceous silica and silica gels) and titania, calcined china clay and alkaline metal salts, such as the carbonates and sulphates of calcium and barium. The particulate inorganic fillers may be of the voiding or non-voiding type. Suitable particulate inorganic fillers may be homogeneous and consist essentially of a single filler material or compound, such as titanium dioxide or barium sulphate alone. Alternatively, at least a proportion of the filler may be

25

heterogeneous, the primary filler material being associated with an additional modifying component. For example, the primary filler particle may be treated with a surface modifier, such as a pigment, soap, surfactant coupling agent or other modifier to promote or alter the degree to which the filler is compatible with the polymer layer. Preferred particulate inorganic fillers include titanium dioxide and silica.

The inorganic filler should be finely-divided, and the volume distributed median particle diameter (equivalent spherical diameter corresponding to 50% of the volume of all the particles, read on the cumulative distribution curve relating volume % to the diameter of the particles - often referred to as the "D(v,0.5)" value) thereof is preferably in the range from 0.01 to 5 μm, more preferably 0.05 to 1.5 μm, and particularly 0.15 to 1.2 μm. Preferably at least 90%, more preferably at least 95% by volume of the inorganic filler particles are within the range of the volume distributed median particle diameter ± 0.8 μm, and particularly ± 0.5 μm. Particle size of the filler particles may be measured by electron microscope, coulter counter, sedimentation analysis and static or dynamic light scattering. Techniques based on laser light diffraction are preferred. The median particle size may be determined by plotting a cumulative distribution curve representing the percentage of particle volume below chosen particle sizes and measuring the 50th percentile.

The components of the composition of a layer may be mixed together in a conventional manner. For example, by mixing with the monomeric reactants from which the layer polymer is derived, or the components may be mixed with the polymer by tumble or dry blending or by compounding in an extruder, followed by cooling and, usually, comminution into granules or chips. Masterbatching technology may also be employed.

25

In one embodiment, the substrate is optically clear, preferably having a % of scattered visible light (haze) of <10%, preferably <6%, more preferably <3.5 % and particularly <2%, measured according to the standard ASTM D 1003. Preferably, the total light transmission (TLT) in the range of 400-800 nm is at least 75%, preferably at least 80%, and more preferably at least 85%, measured according to the standard ASTM D 1003. In this embodiment, filler is typically present in only small amounts, generally not exceeding 0.5% and preferably less than 0.2% by weight of the polymer of a given layer.

5

10

25

30

The following test methods may be used to characterise the polymeric film:

- (i) The clarity of the film may be evaluated by measuring total light transmission (TLT) and haze (% of scattered transmitted visible light) through the total thickness of the film using a Gardner XL 211 hazemeter in accordance with ASTM D-1003-61.
- (ii) Heat-seal strength of the heat-sealable substrate to itself is measured in an Instron Model 4301 by positioning together and heating the heat-sealable layers of two samples of polyester film at 140°C for one second under a pressure of 43 psi (approx. 296 kPa). The sealed film is cooled to room temperature, and the heat-seal strength determined by measuring the force required under linear tension per unit width of seal to peel the layers of the film apart at a constant speed of 4.23 mm/second.
- (iii) Heat-seal strength of the heat-seal bond between the conductive material and the substrate was measured in an Instron Series IX Automated Materials Testing System machine by positioning together and heating the conductive layer and substrate at 140°C for one second under a pressure of 43 psi (approx 296 kPa). The composite film is cooled to room temperature, and the heat-seal strength determined by measuring the force required under linear tension per unit width of seal to peel the layers of the film apart at a constant speed of 50 mm/minute.
- 20 (iv) Ultimate tensile strength at destruction (UTD) and elongation at destruction (ETD) are measured using the ASTM D882-88 test modified as described herein.
  - (v) Shrinkage at a given temperature is measured by placing the sample, unrestrained, in a heated oven at that temperature for the allotted period of time (typically 30 minutes). The % shrinkage is calculated as the % change of dimension of the film in a given direction before and after heating.
  - (vi) Flex-cracking can be assessed qualitatively by the repeated bending (through a given angle and about a fulcrum point) of the composite film comprising the heat-sealable substrate and the conductive layer, and assessing by eye whether any cracks have developed in the conductive layer.

The invention is further illustrated by the following examples. It will be appreciated that the examples are for illustrative purposes only and are not intended to limit the invention

20

as described above. Modification of detail may be made without departing from the scope of the invention.

## **EXAMPLES**

#### 5 Example 1

A bi-layer polyester film comprising a substrate layer of clear PET and a copolyester heat-sealable layer was prepared as follows. A polymer composition comprising PET was co-extruded with a copolyester comprising terephthalic acid/isophthalic acid/ethylene glycol (82/18/100), cast onto a cooled rotating drum and stretched in the direction of extrusion to approximately 3 times its original dimensions. The film was passed into a stenter oven at a temperature of 100°C where the film was stretched in the sideways direction to approximately 3 times its original dimensions. The biaxially-stretched film was heat-set at a temperature of about 230°C by conventional means. The total thickness of the final film was 23 μm; the heat sealable layer was approximately 4 μm thick.

15

A copper foil (12  $\mu$ m) was disposed directly onto the surface of the film by contacting the foil with the heat-sealable surface of the coextruded film and effecting lamination at 140°C for one second under a pressure of 43 psi (about 296 kPa). The heat-seal strength of the metal/film bond was 450 g/25mm<sup>2</sup>.

20

A pattern was etched in the copper layer according to the techniques described herein by forming an etching-resist having a wiring pattern on the surface of the copper foil by gravure printing, curing the etching-resist ink by UV irradiation, forming a conductor wiring pattern on the surface of the substrate by ferric chloride etching at 50°C, and removing the etching-resist material by dipping the etched film into a sodium hydroxide solution at room temperature.

#### Example 2

Example 1 was repeated except that the total thickness of the final film was 75 μm; the 30 heat sealable layer being approximately 11 μm thick. In addition, a copper foil of 20 μm in thickness was laminated to the coextruded film at 160°C for one second under a pressure of 40 psi (about 275 kPa). The heat-seal strength of the metal/film bond was 1323 g/25mm². The shrinkage of the film was 2% in both MD and TD directions.

21

#### Example 3

Example 1 was repeated except that the total thickness of the final film was 75 μm; the heat sealable layer being approximately 11 μm thick. In addition, an aluminium foil of 13 μm in thickness was laminated to the coextruded film at 160°C for one second under a pressure of 40 psi (about 275 kPa). The heat-seal strength of the metal/film bond was 343g/25mm<sup>2</sup>.

## Example 4

10 Example 1 was repeated except that the total thickness of the final film was 30 μm; the heat sealable layer being approximately 5 μm thick. In addition, a copper foil of 20 μm in thickness was laminated to the coextruded film at 160°C for one second under a pressure of 40 psi (about 275 kPa). The heat-seal strength of the metal/film bond was 537 g/25mm<sup>2</sup>.

## 15 Example 5

Example 1 was repeated except that the total thickness of the final film was 30  $\mu$ m; the heat sealable layer being approximately 5  $\mu$ m thick. In addition, an aluminium foil of 13  $\mu$ m in thickness was laminated to the coextruded film at 160°C for one second under a pressure of 40 psi (about 275 kPa). The heat-seal strength of the metal/film bond was  $213g/25mm^2$ .

## Example 6

The procedure of Example 1 was repeated using a copolyester of terephthalic acid/1,4-cyclohexane dimethanol/ethylene glycol (100/33/67) as the heat-sealable layer.

25

20

#### Example 7

A polymer composition comprising polyethylene terephthalate was extruded and cast onto a cooled rotating drum and stretched in the direction of extrusion to approximately 3 times its original dimensions. The film was passed into a stenter oven at a temperature of 100°C where the film was stretched in the sideways direction to approximately 3 times its original dimensions. The biaxially stretched film was heat-set at about 230°C by conventional means. The heat-set film was then coated off-line using conventional coating means with a

copolyester of azeleic acid/terephthalic acid/ethylene glycol (45/55/100) to give a dry coating thickness of 2  $\mu m$ . The total film thickness was 25  $\mu m$ .

An aluminium foil (13 µm) was disposed directly onto the surface of the film by contacting the foil with the heat-sealable surface of the coextruded film and effecting lamination at 160°C for one second under a pressure of 40 psi (about 275 kPa). The heat-seal strength of the metal/film bond was 528g/25mm<sup>2</sup>. A pattern was formed in the foil, as described in Example 1.

#### 10 Example 8

The procedure of Example 7 was repeated except that the thickness of the base layer was 75µm; the coating thickness was approximately 12µm; and prior to coating with the azelaic acid-containing copolyester the heat-set biaxially stretched film was first heat-stabilised by unwinding the film and passing it through a series of four flotation ovens and allowing it to relax by applying the minimum line tension compatible with controlling the transport of the web. The heat-stabilised film was then wound up. Each of the four ovens had three controlled temperature zones in the transverse direction (left, centre and right):

		Left	Centre	Right
20	Oven 1	170	180	170
	Oven 2	170	180	170
	Oven 3	170	180	170
	Oven 4	165	180	165

25 The line speed of the film during the heat-stabilisation step was 15m/min. The tensions used for the film (1360mm original roll width) were 24-25N.

An aluminium foil was laminated to the film at 160°C for one second under a pressure of 60 psi (about 413 kPa), to form a heat-seal bond having a strength of 2028g/25mm<sup>2</sup>. A pattern was formed in the foil, as described in Example 1.

## Example 9

23

The procedure of Example 8 was repeated except that a copper foil was used instead of the aluminium foil. The bond strength was 2824 g/25mm<sup>2</sup>.

## Example 10

5 The procedure of Example 7 was repeated except that the base film was coated with EVA copolymer of 10  $\mu$ m in thickness, the total film thickness being 33  $\mu$ m. The metal/polymer bond strength was 471 g/25mm<sup>2</sup>.

# Example 11

10 The procedure of Example 10 was repeated except that copper foil was laminated to the composite film, the metal/polymer bond strength being 833 g/25mm<sup>2</sup>.

#### **CLAIMS**

30

- 1. A radio-frequency responsive tag comprising a heat-sealing substrate comprising a polyester layer, and an antenna comprising a pattern of conductive material wherein said conductive material is in direct contact with a heat-sealing surface of the substrate, wherein the shrinkage of the heat-sealing substrate is less than 5% at 190°C over 30 minutes.
  - 2. A tag according to claim 1 wherein the shrinkage is 3% or less.
- 10 3. A tag according to claim 1 or 2 wherein the substrate is a monolayer substrate.
  - 4. A tag according to claim 1, 2 or 3 wherein the substrate comprises a copolyester.
- 5. A tag according to claim 4 wherein the copolyester is derived from ethylene glycol, 15 terephthalic acid and isophthalic acid.
  - 6. A tag according to claim 1 or 2 wherein the substrate comprises a heat-sealable layer and a base layer.
- 20 7. A tag according to claim 6 wherein the base layer comprises polyethylene terephthalate.
  - 8. A tag according to claim 6 or 7 wherein the heat-sealable layer is a copolyester.
- 25 9. A tag according to claim 8 wherein the copolyester is derived from ethylene glycol, terephthalic acid and isophthalic acid.
  - 10. A tag according to claim 8 wherein the copolyester is derived from terephthalic acid, ethylene glycol and 1,4-cyclohexanedimethanol.
  - 11. A tag according to claim 10 wherein the molar ratio of 1,4-cyclohexanedimethanol to ethylene glycol is in the range from 30:70 to 35:65.

- 12. A tag according to any of claims 6 to 11 wherein the heat-sealable layer and base layer are coextruded.
- 13. A tag according to claim 5 or 9 wherein the copolyester is derived from ethylene glycol, terephthalic acid and isophthalic acid, wherein the molar ratio of the terephthalic acid component to the isophthalic acid component is in the range from 65:35 to 85:15.
- 14. A tag according to claim 8 wherein the copolyester is derived from terephthalic acid, an aliphatic dicarboxylic acid and a glycol.
  - 15. A tag according to claim 8 wherein the copolyester is derived from terephthalic acid, azeleic acid and ethylene glycol.
- 15 16. A tag according to claim 15 wherein the copolyester is a copolyester of ethylene glycol with about 55% terephthalic acid and about 45% azeleic acid.
  - 17. A tag according to claim 5 or 6 wherein the heat-sealable layer comprises ethylene vinyl acetate (EVA).
- 18. A tag according to any of claims 6, 7, 8 or 14 to 17 wherein the heat-sealable layer is coated on to the base layer.

20

25

- 19. A tag according to any of claims 14 to 18 wherein said shrinkage is less than 1%.
- 20. A tag according to any of claims 14 to 18 wherein said shrinkage is less than 0.5%.
  - 21. A tag according to any preceding claim wherein the conductive material comprises a metallic material.
- 22. A tag according to any preceding claim wherein the conductive material comprises copper.

26

- 23. A tag according to any preceding claim which further comprises a data-carrying means in electrical communication with the antenna.
- A method of manufacture of a radio-frequency responsive tag comprising a substrate, an antenna comprising a pattern of conductive material, and optionally a data-carrying means, said method comprising the following steps:
  - (i) providing a heat-sealable substrate comprising a polyester layer wherein the shrinkage of said substrate is less than 5% at 190°C over 30 minutes;
  - (ii) disposing the conductive material of the antenna directly onto at least part of a heat-sealable surface of the substrate;
    - (iii) effecting heat-sealing between the heat-sealable substrate and the conductive material;
    - (iv) optionally providing a data-carrying means in electrical communication with the conductive material.

15

10

- 25. A method according to claim 24 wherein the method further comprises, subsequent to step (iii), formation of a pattern in the conductive material
- 26. Use of a heat-sealing film comprising a polyester layer wherein the shrinkage of said film is less than 5% at 190°C over 30 minutes as a substrate in the manufacture of a radio-frequency responsive tag comprising said film as a substrate, an antenna comprising a pattern of conductive material, and optionally a data-carrying means in electrical communication with the antenna, wherein the conductive material is in direct contact with a heat-sealing surface of the film.

25

30

27. Use of a heat-sealing film comprising a polyester layer wherein the shrinkage of said film is less than 5% at 190°C over 30 minutes, for the purpose of reducing flex-cracking in a composite film suitable as a precursor in the manufacture of an RF-responsive tag, said composite film comprising a layer of conductive material and said heat-sealing film wherein the conductive material is in direct contact with a heat-sealing surface of the substrate.

27

28. Use of a composite film comprising a layer of conductive material and a heat-sealing substrate comprising a polyester layer wherein the shrinkage of said heat-sealing substrate is less than 5% at 190°C over 30 minutes, as a precursor in the manufacture of a radio-frequency responsive tag comprising said heat-sealing substrate, an antenna comprising a pattern of said conductive material, and optionally a data-carrying means in electrical communication with the antenna, wherein the conductive material is in direct contact with a heat-sealing surface of the substrate.

5

- 10 29. A composite film suitable for use as a precursor in the manufacture of a radio-frequency responsive tag, said composite film comprising a layer of conductive material and a heat-sealing substrate comprising a polyester layer wherein the shrinkage of the heat-sealing substrate is less than 5% at 190°C over 30 minutes, wherein the conductive material is in direct contact with a heat-sealing surface of the substrate.
  - 30. A composite film according to claim 29 wherein the heat-seal bond strength between the conductive material and the substrate is at least 200 g/25mm<sup>2</sup>.

## INTERNATIONAL SEARCH REPORT

Application No PCT/GB2005/000999

	· · · · · · · · · · · · · · · · · · ·	101/402005/	000555	
A. CLASSII IPC 7	FICATION OF SUBJECT MATTER G06K19/077 H05K3/02 B32B31/	00 G08B13/24 H01Q1/	/22	
According to	h International Patent Classification (IPC) or to both national classific	cation and IPC		
	SEARCHED			
Minimum do IPC 7	cumentation searched (classification system followed by classificat G06K H05K B32B G08B H01Q	tion symbols)		
	ion searched other than minimum documentation to the extent that		rched	
i	ata base consulted during the international search (name of data b	ase and, where practical, search terms used)		
C. DOCUME	ENTS CONSIDERED TO BE RELEVANT			
Category °	Citation of document, with indication, where appropriate, of the re	elevant passages	Relevant to claim No.	
X	WO 03/028416 A (INFINEON TECHNOL GUNDLACH, HARALD; MUELLER-HIPPER S) 3 April 2003 (2003-04-03) page 1, line 11 - line 23 page 5, line 30 - page 7, line 3 page 10, line 7 - line 35 figures 2a,2b	, ANDREAS;	1-30	
Furti	her documents are listed in the continuation of box C.	Patent family members are listed in a	annex.	
"A" docume consid "E" earlier of filing d "L" docume which citation "O" docume other of docume of the reference of the refere	ent defining the general state of the art which is not lered to be of particular relevance document but published on or after the international late state of the art which may throw doubts on priority claim(s) or is clied to establish the publication date of another n or other special reason (as specified) ent referring to an oral disclosure, use, exhibition or means ent published prior to the international filing date but nan the priority date claimed	T' later 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.		
Date of the	actual completion of the international search	Date of mailing of the international search	h report	
1	1 July 2005	05/08/2005		
Name and r	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) 440–3016	Authorized officer  de Ronde, J.		

## INTERNATIONAL SEARCH REPORT

Application No PCT/GB2005/000999

WO 03028416 A			member(s)	date
	03-04-2003	DE WO TW US	10145749 A1 03028416 A1 546996 B 2005034995 A1	24-04-2003 03-04-2003 11-08-2003 17-02-2005