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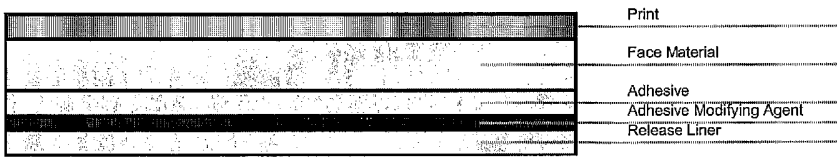
(54) Title: PRESSURE SENSITIVE FILM LABELS FOR REUSABLE CONTAINERS



(57) Abstract: A label which is removable from an article under the action of a hot washing fluid, the label including an adhesive layer comprising a pressure sensitive and heat sensitive adhesive material. The adhesive properties of the adhesive layer are reduced when the article is subjected to heat, so assisting the removal of the label from the article.

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PRESSURE SENSITIVE FILM LABELS FOR REUSABLE CONTAINERS

Field of the Invention

The invention relates to a label for an article, in particular, although not exclusively, for a reusable container, the backing material layer of the label being bondable onto the article by means of an adhesive layer and the label being removable from the article under the effect of hot washing fluid. The articles may be beverage bottles of glass or plastic, repeatedly reusable outer packagings for a multiplicity of individual containers, in particular beverage bottle crates, etc. The invention also relates to a method of making the label, an article to which the label has been applied and a method of removing the label from an article to which it has been applied by the application of a hot washing fluid.

Background of the Invention

In the beverage industry, the containers used, for example bottles, can be subject to a high quota of reuse. The containers are cleaned with each return before refilling, the labels also being detached during washing of the containers. Then the containers are refilled and relabelled corresponding to the beverage type filled. If the vessels are standardised for a particular product group, such as a beer bottle, the bottles returning to the brewery do not need to be resorted according to beer types, as would be the case with permanently pre-decorated bottles. The different labelling usually only occurs after filling. In the case of a direct printing of the bottle which is capable of withstanding the washing process, large warehouse stocks of the appropriate pre-decorated bottles would have to be held in readiness.

In the beverage industry, the washing of the containers is generally carried out via a process of immersion and spraying of the containers with a hot washing fluid, such as dilute caustic soda, heated to between 60°C and 90°C.

Often, paper labels with wet-glue adhesive are used for the labelling of reusable containers. In this case, the wet-glue adhesive is applied to the full surface or in strips, the adhesive only being applied to the paper immediately before labelling. The disadvantage of this label type is that the filler must work with wet glue and the handling of these labels is often more difficult than that of pressure sensitive labels. Pressure sensitive labels are obtained from the label suppliers already provided with adhesive.

Because of the standardised washing conditions in the beverage industry, it has previously only been possible to use paper-based labels for wet-glue or pressure sensitive application. During washing off of the labels in the wash station, the water permeability and swelling nature of paper is exploited with the object that the adhesive comes relatively quickly into full-surface contact with the washing liquid, and is completely detached in the predetermined washing time of the order of some minutes. In the case of wet glue applied labels, the adhesives are usually formulated to go into solution in the washing liquid. In the case of pre-coated pressure sensitive labels, redispersible adhesives are often used and the objective is that the adhesive is removed intact with the paper face to avoid contamination of the washing bath. This high permeability for washing liquid and water is not possessed by the thermoplastic films used for many labels, such as polyolefins, polyesters, polyvinyl chlorides, polystyrenes, etc. Such films prevent the access of the washing liquid to the interface of the adhesive and container surface, so that the impermeable film labels can only be slowly detached from the label edge, which, without additional mechanical support, such as brushes, high-pressure nozzles, etc., does not permit complete removal of the labels within an economically justifiable time span. These mechanical means are undesirable because of the higher outlay.

In the beverage industry, however, there is an increasing demand for film-based labels precoated with adhesive. Such film labels, in contrast to paper labels, can be decorated in an extremely wide range of ways. In contrast to paper, they are also available in transparent form, have excellent wet strength and can be dispensed onto the containers at high speed in standardised machines, without the need to work with wet adhesives, as is the case of the wet-glue paper label. Their mechanical properties such as tensile strength and extensibility are greatly superior to those of paper labels. However, a problem with film labels is that they do not readily wash off with existing washing systems as easily as the paper labels often used until now.

One solution to the problem outlined above is set forth in US Patent No. 6680097 (Steinbeis), the contents of which are hereby incorporated by reference, which states that a label is detachable from the article with little effort. This is achieved by the use of a label in which the backing material layer comprises a plastic film layer that is stretched in at least one direction and shrinks back under the effect of heat, such as the temperature of the washing fluid or/and by thermal radiation, so as to overcome the retention force of the adhesive layer. By virtue of the effect of the heat, a shrinking back of the plastic film occurs, while at the same time the adhesive loses adhesive force. It is claimed that by this means, the label detaches gradually from the article, for

example from the edge or with the formation of channels, and can be easily removed within an extremely short time.

Another prior art reference is WO02/07474, the contents of which are hereby incorporated by reference, which discloses a label in which the film is a biopolymer such as cellulose.

US-A-2003/0150148, the contents of which are hereby incorporated by reference, discloses a label which has embossed or debossed portions to correspond to indicia printed on the label.

Broadly speaking pressure sensitive adhesives that are designed to offer wash-off properties for reusable container applications can be categorised as follows: water wash; alkali sensitive and alkali soluble.

The characteristics of water wash adhesives is that in conjunction with an appropriate facestock, most typically paper due to its inherent water permeability, these adhesives can be removed from an article simply with cold or ambient temperature water. In ambient humidity conditions they retain their permanent adhesive characteristics, but when exposed to a sufficient level of water such as through immersion of the article in a water bath, hosing or spraying the adhesive quickly loses adhesion to the article and the label detaches. Whilst the facestock can be a paper with an appropriate level of wet strength so that it remains intact after the washing process, applications exist where the facestock may require a paper grade that is water dispersible. In addition, water soluble materials also exist for specific applications. In their simplest form water wash adhesives generally contain a given percentage of a water-soluble component such as derivatives of polyvinyl ethers, although water soluble polyacrylate-based pressure sensitive adhesives are also known.

The characteristics of alkali sensitive adhesives is that in conjunction with an appropriate facestock, most typically paper due its inherent water permeability, these adhesives cannot be removed easily with immersion of the labelled article in cold or ambient temperature water. They can only be successfully removed from an article, in commercial application terms, with hot alkaline solutions, typical of returnable bottle washing plants. This distinction from water wash adhesives is important for applications where a defined level of water resistance is required as part of the functionality of the label. A typical example is wine and champagne labelling, where a specified level of ice water resistance is required. These adhesives are generally

modified acrylic dispersions. They contain within their formulations components which are balanced in ratio to impart the required level of water resistance, but when in full surface contact with an alkaline solution react and facilitate wash-off.

The characteristics of alkali soluble adhesives category is that in conjunction with an appropriate facestock, most typically paper due to its inherent water permeability, these adhesives will be soluble in hot alkaline solution as against the previously described non-soluble adhesives. These adhesives have been specifically developed to support the paper recycling process. US-A-5229447 discloses an alkali soluble pressure sensitive adhesive which is described as comprising 100 parts by weight of a polymer obtained by polymerisation of a carboxyl-group containing vinyl monomer as an adhesive component and 50-500 parts by weight of a non-ionic surface active agent capable of endowing plasticity as a main additive component. Commercial applications for this particular type of adhesive are reported to be reducing due largely to cost.

A characteristic of pressure sensitive adhesives is that they are permanently tacky at the temperature of use, such as room temperature. Pressure sensitive adhesives adhere to a variety of substrates when applied with pressure, do not require activation by water, heat or solvents, and have sufficient cohesive strength to be handled with fingers. The primary bond for a pressure sensitive adhesive is not chemical or mechanical but rather a polar attraction to the substrate, and requires pressure to achieve sufficient "wet-out" onto the surface to provide adhesion. Further information regarding pressure sensitive adhesives can be found in the textbook Pressure-Sensitive Adhesives Technology by Istvan Benedek, Luc J. Heymans, Istran Benedek (Marcel Dekker 1997).

One would not normally associate pressure sensitive adhesives categorised as having 'permanent' features within the scope of 'wash-off' adhesives. However, it has been found that successful wash-off can be achieved with permanent adhesives that demonstrate the specific properties described herein.

Summary of the Invention

According to the present invention there is provided a label which is removable from an article under the action of a hot washing fluid, which label comprises a backing layer which is a polymeric film and an adhesive layer for bonding the label on to the article, the adhesive layer comprising a pressure sensitive and heat sensitive material.

In the context of the present invention, "heat sensitive" means that the adhesive properties of the adhesive layer degrade or diminish on heating to a temperature in the range 40°C to 120°C, whether or not in the presence of a washing fluid. Preferably, the diminution of adhesive properties occurs at temperatures in the range 40°C to 90°C, and more preferably temperatures in the range 40°C to 70°C. In preferred embodiments, the adhesive properties of the adhesive layer diminish at a temperature in the range 50°C to 70°C and more preferably 55°C to 65°C.

The expression "heat sensitive" is not to be taken as indicating that the adhesive properties of the adhesive layer necessarily diminish instantaneously when subjected to a sufficiently high temperature, but occurs within a time consistent with the requirements of a washing process. Thus, the diminution of adhesive properties preferably occurs in less than 1 minute, and more preferably less than 30 seconds, and not more than 3 minutes after the article carrying the label is subjected to the appropriate temperature. However, it is to be appreciated that longer periods, for example of not more than 10, 8 or 5 minutes after the article is subjected to the elevated temperature would still be regarded as falling within the broad scope of the present invention.

The adhesive property which diminishes when the article is subjected to the elevated temperature is the adhesion, or peel strength, of the adhesive layer. Preferably, the peel strength decreases by at least 30%, and in some cases by at least 40% or even 50%, when the article is subjected to the elevated temperature. In especially preferred embodiments, the peel strength reduces to approximately 20% of the peel strength at ambient conditions, i.e. a reduction of 80%. In general, a reduction in peel strength will be sufficient for the purposes of the present invention if the remaining peel strength of the adhesive layer is inadequate to retain the label on the article when the article is subjected to the mechanical action of a high temperature washing liquid in a normal commercial washing process. An industry recognized test for peel adhesion is described in the FINAT Technical Handbook 6th Edition and is FTM1, Peel Adhesion (180°) at 300mm per minute. The peel test describes the permanence of adhesion or peelability of pressure sensitive adhesives. Test strips of 25mm wide are prepared, adhered to the test plate (glass or metal) and then checked after a specific period (usually 20 minutes and 24 hours). The force required to peel the label material from the test plate is recorded in Newtons and observations on adhesive transfer are noted. For the purposes of the present invention, what is most important is that the peel adhesion to the article under the washing conditions is low, for example no more than

5N/25mm. The peel adhesion at ambient conditions depends on how heat sensitive the chosen adhesive is and also on coat weight, but could be as high as 25N/25mm.

The peel strength to be considered in the context of the present invention is the peel strength of the adhesive layer when the label is applied to the intended article, which may be made from glass, plastics, metal or ceramics, for example.

The pressure sensitive adhesive may be a solvent-borne, water-borne or a hot-melt, , pressure sensitive adhesive having a combination of polymer component and surfactant component, which permit the label to be removed from an article to which the label has been attached, preferably within a period of 10 minutes using an aqueous wash at between 60°C and 90°C. The adhesive material may be rubber-based, acrylic-based or modified acrylic-based. In a preferred embodiment, acrylic-based adhesives are used, modified in order to enhance their heat-sensitive properties to comply with the requirements mentioned above. Modification of the base adhesive material may comprise the addition of a surfactant, which may be anionic, amphoteric, cationic, non-ionic or polymeric surfactants. If an anionic surfactant is used, it may comprise any one of various organic sulphates, sulphonates, sulphonic acids, salts and blends thereof, or sulphosuccinates. If a non-ionic surfactant is used, it may comprise an ethoxylate, an alcohol ethoxylate or an alkoxylate. The surfactant is preferably present in the adhesive material in an amount not more than 5%, preferably not more than 3%, and more preferably not more than 2%. A preferred embodiment is a reactive surfactant that is attached to functional groups in the polymer of the adhesive. This avoids the possibility that the surfactant can wash out of the adhesive layer, for example if the article carrying the label is immersed in water below the temperature at which the adhesive layer loses its adhesion. For example, if the article is a beer or wine bottle, it may be immersed in iced water to cool its contents before consumption.

The adhesive material is preferably non-resinated, that is to say it lacks any additional tackifier or other added resin.

The thickness of the adhesive layer may be lower than is conventionally used for pressure sensitive adhesive layers on labels. Preferably, the thickness of the adhesive layer is not more than 15 g/m^2, and may be not more than 14 g/m^2, although it is envisaged that in most applications, the adhesive layer will have a thickness not less than 10 g/m^2.

According to another aspect of the present invention, there is provided a method of removing from an article a label as defined above, the method comprising:

(i) subjecting the article to a preheating step, in which the article is heated to a temperature in excess of 40°C, whereby the peel strength of the adhesive is reduced by not less than 40%, and, subsequently,

(ii) subjecting the article to a washing process in which the article is (a) immersed in an agitated washing fluid and/or (b) sprayed with a washing fluid, whereby the mechanical action of the agitation and/or spraying separates the label from the article.

The preheating step may comprise immersing the article in a hot water bath having a temperature in excess of 50°C. In the washing step, the washing fluid may comprise an alkaline solution, for example an aqueous solution of NaOH at a concentration in the range 1% to 2.5%.

Preferably the label is removed from the article with the adhesive layer remaining captive on the label.

A label in accordance with the present invention may also incorporate the features disclosed in our co-pending British patent applications 0405271.8 and/or 0412663.7.

Thus, in accordance with the invention of British patent application 0405271.8, the pressure sensitive adhesive layer may have an inner surface which faces towards the backing layer and an outer surface for adhering to the surface of the article, wherein the outer surface of the adhesive layer, prior to its application to an article, includes a plurality of micro-channels which extend to the periphery of the label.

Other features of the co-pending application which may be incorporated in the label of the present invention are as follows:

- the micro-channels extend all or part way across the label, when viewed in the plane of the label.
- the channels only extend part way into the label.
- the channels have a depth of no greater than about 15μm.
- the channels have a depth of no greater than about 10μm.
- the channels have a depth of at least 1μm.
- the channels have a depth of at least 5μm.

- the channels have a width of no greater than about 250μm.
- the channels have a width of no greater than about 150μm.
- the channels have a width of no less than about 10μm.
- the channels have a width no less than about 50μm.
- the minimum distance between adjacent channels is about 10μm
- the maximum distance between adjacent channels is 10mm.
- the polymeric film of the label is selected from thermoplastic films such as polyolefins, polycarbonates, polyesters, polyvinyl chlorides and polystyrenes; and biopolymers such as cellophane or polylactic acid (PLA).
- the polymeric film is a polypropylene film.
- the polymer film has a thickness of from 15μm to 100μm.

The method of the British patent application 0405271.8 may be used to make labels of the present invention. This method comprises

providing a composite structure comprising a backing layer which is a polymeric film and a pressure sensitive adhesive layer for bonding the label onto the article, the pressure sensitive adhesive layer having an inner surface which faces and adheres to the backing layer and an outer surface for adhering to the surface of the article; and

forming micro-channels in the outer surface of the adhesive layer, which micro-channels extend to the periphery of the label.

In the method the micro-channels may be formed by embossing a surface of the composite structure.

In accordance with the invention disclosed in our co-pending British patent application 0412663.7, the outer surface of the adhesive layer, prior to its application to an article, is provided with a coating of an adhesive modifying agent.

The label may be made by a method as disclosed in our co-pending British patent application 0412663.7, in a coating of an adhesive modifying agent is applied to the surface of the adhesive layer away from the backing layer.

One example of an adhesive modifying agent is a UV-cured adhesive deadening varnish.

In preferred embodiments incorporating an adhesive modifying agent, the adhesive modifying agent serves to reduce the adhesive strength of the adhesive layer in the region or regions at which the adhesive modifying agent is applied. However, the expression "adhesive modifying agent" is used in a broad sense to mean any agent

which has an effect on the chemical, physical or mechanical properties of the adhesive layer.

The adhesive modifying agent may be applied to the adhesive surface through direct printing by a conventional print process i.e. screen, flexographic, letterpress or gravure. To effect this, the face material and adhesive part of the construction is first de-laminated from the release liner to allow the printing of the adhesive modifying agent onto the now exposed adhesive face and then re-laminated after printing through a nip roller structure. Such release liners are invariable provided on labels with pressure-sensitive adhesive, to cover the adhesive during transport and handling so as to prevent the adhesive sticking unintentionally to itself or to other articles. The release layer is removed just before the label is affixed to the intended article.

The adhesive modifying agent may be applied to the full surface of the label or in patterns appropriate to the label profile.

The adhesive modifying agent may be applied as a series of round dots with the percentage coverage appropriate to the functional requirements of the individual label. Percentages in the range of 3% to 10% have been found preferable. The printing structure may also take the form of square dots, parallel or intersecting lines and other structures known to those skilled in the art.

The purpose of the adhesive modifying agent is to facilitate the lifting of the label edges within the washing bath without any mechanical means other than the stiffness (flexural modulus) of the label material overcoming the adhesion to the article and the turbulence created by the water streams. This then allows the washing solution to penetrate behind the label and effect the washing-off of the label, so enhancing the reduction in adhesion achieved by the use of a heat sensitive adhesive.

Brief Description of the Drawings

For a better understanding of the present invention, and to show how it may be put into effect, reference will now be made, by way of example, to the accompanying drawings in which:

Figure 1 represents the cross-sectional primary composite structure of a pressure sensitive label, where through the mechanical process of embossing and via the properties of the label material described herein, the print layer, face material and

adhesive layer have been physically altered to replicate the pattern of the embossing tool used;

Figure 2 shows the front face of the label of Figure 1.

Figure 3 represents the cross-sectional primary composite structure of an alternative pressure sensitive label, which has passed through the process of printing a modifying agent onto the adhesive surface; and

Figure 4 shows examples of patterns in which the modifying agent can be applied to the rear face of the label of Figure 3.

Detailed Description

Polymeric film

The polymeric film of the label of the present invention may be any suitable polymer film from which labels may be made. For the avoidance of doubt, the polymeric film used in the invention is not a paper. Examples of polymer films are thermoplastic films such as polyolefins, polycarbonates, polyesters, polyvinyl chlorides and polystyrenes. Biopolymers such as cellophane or polylactic acid (PLA) may also be used. Particularly preferred are water-impermeable, heat-stable, polyolefin films, most preferably polypropylene films, such as biaxially-oriented polypropylene (BOPP).

The polymer film may be a laminate or a single layer. It may include additional non-polymer layers such as a metallized layers.

The polymer film is preferably transparent to provide a "no-label" look. It may or may not be coloured to match the colour of an article to which the label is to be attached.

The polymer film may have a thickness typical of films for labels, such as from 15μm to 100μm, preferably 40 to 50μm.

The polymer film may be treated by known methods in the art and may be coated in a manner which is known per se.

Although the polymeric film may shrink or otherwise deform when subjected to heat, or water, or more particularly to a hot caustic washing liquid, it is not necessary for the film to behave in this way. A non-shrinking and non-deforming film may be used.

Pressure sensitive adhesive

The adhesive used in the present invention may be a water borne, , pressure sensitive adhesive, although it is known that the mechanisms that facilitate the label wash-off and are described herein, could equally be incorporated into a solvent or hot-melt pressure sensitive adhesive system. By "water-borne" is meant that water is used as the carrier in which the polymers of the pressure sensitive adhesive are dispersed in the manufacture of the adhesive. Typically, water-borne pressure sensitive adhesives are made by a process in which the polymer component of the adhesive is dispersed in water as a solvent and surfactant(s), for example in amounts of about 50:50 by weight of the polymer to the solvent used. The water is subsequently removed. An analogous process is used for making solvent-based adhesives, although a solvent other than water, usually a volatile organic solvent, is used. It is preferable that the pressure sensitive adhesive is also typically not one which is "resinated". By this is meant that the pressure sensitive adhesive has not had any additional resins added to it, i.e. tackifiers.

The mechanisms by which wash-off is facilitated by the preferred adhesive is that when the label is heated above 40°C, more preferably above 60°C, as would be typically encountered by the labeled container within a washing environment, for example during a preheating stage before the article enters the washing tank, it loses adhesion (i.e. peel strength) to the container by as much as, or in excess of, 50% i.e. it can be peeled off the article more readily, although the adhesive does however retain some tack. The adhesion loss occurs fairly rapidly, preferably within 1 minute of immersion at the elevated temperature. This loss in adhesion facilitates the edges of the label being lifted from the container and the washing solution is then able to ingress behind the label, between the adhesive layer and the article and complete the full removal of the label through a combination of physical and chemical means.

The physical removal of the label is effected by the turbulence created within the washer by its motion and more specifically the high liquid flow which is generated by the pumps at the label removal points of the machine. The force of the water movement being greater than that of the adhesion to the container, shears the label from the container.

The chemical removal of the label is effected by the adhesive coming into contact with the hot washing fluid, allowing chemical interactions to take place between the washing solution and additives in the adhesive, such as surfactants, that have the effect of eliminating the tack and therefore preventing the label from re-adhering to the container surface or to any other parts within the machine system.

It will thus be appreciated that a fundamental property of the adhesive material is its significant sensitivity to heat. This is in contrast to known pressure sensitive adhesives used for labels on articles such as bottles which are to be removed in a washing process. Known adhesives for securing labels to articles such as bottles have been developed on the basis that conventional washing processes utilize an alkaline washing fluid such as an aqueous solution of NaOH, and have therefore been developed so as to be dissolved or otherwise made ineffective in the presence of such an alkaline fluid. Such known adhesives are not sufficiently heat sensitive, in that their properties are not sufficiently affected by the application of heat alone (for example by immersion in hot water) to achieve separation of the label from the article within a short enough time to provide a commercially viable label removal system.

Other preferred properties of the pressure sensitive adhesive are as outlined below;

The adhesion of the label to the container under chilled or even ice water immersion conditions should remain at a level consistent with that under ambient conditions and in the case of full ice water immersion, ingress of water behind the label should not occur within the timescales that are commonly required for labeled beverage containers. Preferably resistance to ice water immersion should be a minimum of 72 hours, more preferably 7 days and in some cases as much as 21 days. The adhesive should demonstrate minimal water whitening within this period. In addition, labeled containers that have been immersed in ice water during the product's use should show no significant loss in their subsequent washing performance. This can be effected by the surfactants being reacted i.e. bonded, within the adhesive layer's structure and not therefore being 'flushed' out when water immersed, a problem that has been known to arise with paper based pressure sensitive labels.

It is preferred that when the label is removed from the article, the adhesive layer remains captive on the label, thereby minimising physical pollutants remaining within the washing machine.

It is also preferred that the washing performance of the adhesive will not deteriorate significantly as a consequence of aging during the product's life cycle, typically this can be a period of up to 12 months from the time of labeling and includes exposure of the labelled container to varying temperature, varying humidity and UV light.

In achieving the functionality of the label as described herein it is preferable that the pressure sensitive adhesive is coated onto the filmic label facestock at a coat weight of between 10gsm and 20gsm, most preferably 12 – 14gsm. Preferably the adhesive is applied to the label material covering the full surface or in regions with gaps, or if appropriate in patterns.

The pressure sensitive adhesive used in the present invention, when tested in accordance with the "water whitening" test method that is described on Page 558 of Pressure Sensitive Adhesive Technology, Istvan Benedek & Luc J Heymans, may have a performance that is classified as 'fair' i.e. the adhesive film does not show any signs of water whitening within 10 seconds. Indeed, for examples of pressure sensitive adhesives for use in the invention, whitening may not occur for 90 seconds and even up to 300 seconds.

The pressure sensitive adhesive used in the present invention, when tested in accordance with the "loss of transparency" test method that is described on Page 558 of Pressure Sensitive Adhesive Technology, Istvan Benedek & Luc J Heymans, may have a performance that is classified as 'fair' i.e. the adhesive film demonstrates a difference in transparency between wet and dry laminate of <8%.

The pressure sensitive adhesive used in the present invention, when tested in accordance with the "wet anchorage" test method that is described on Page 558 of Pressure Sensitive Adhesive Technology, Istvan Benedek & Luc J Heymans, may have a performance such that the adhesive layer does not rub off the intended substrate after immersion in distilled water for 7 minutes.

The pressure sensitive adhesive used in the present invention, when tested in accordance with the "wet adhesion on glass" test method that is described on Page 558 of Pressure Sensitive Adhesive Technology, Istvan Benedek & Luc J Heymans, may have adhesive recovery properties over time after immersion of the sample in

distilled water for 7 minutes in excess of 60 minutes when coated onto a polypropylene material and between 30 minutes and 60 minutes on regenerated cellulose film.

A preferred pressure sensitive adhesive for use in the present invention is SE5279, commercially available from HB Fuller Limited.

The pressure sensitive adhesive used in the present invention is one which has a combination of polymer component and surfactant component that offers all of the mechanisms outlined and which permit the label to be removed from an article to which the label has been attached within a period of 10 minutes using an aqueous wash at 80°C.

More specifically, the pressure sensitive adhesive is preferably one which, when used as the adhesive for a label in which the facestock is a biaxially oriented polypropylene (BOPP) film layer (which may have a water vapour transmission rate of about 3.3 g/m^2 per 24 hours at 38°C and 90% relative humidity) having a thickness of 50µm, the label is removed from a glass substrate within 10 minutes or less under the action of a wash water at 80°C, wherein the wash water has a 2% NaOH concentration and a 0.1% addition of a surfactant washing additive, such as P3 Stabilon Plus, available from Ecolab, Inc. Preferably in this test, the label should be removed from the glass substrate in less than 8 minutes, more preferably within 5 minutes and most preferably within 3 minutes.

With other more water permeable facestock materials, such as regenerated cellulose film (which may have a water vapour transmission rate of around 370 g/m^2 per 24 hours at 30 °C and 90% relative humidity), the label will be removed in a shorter period of time than noted above. Such materials may have a thickness of about 45µm.

In the above noted test, the labelled article may be allowed a minimum of 24 hours, standing time at ambient conditions (23°C, 50% RH) prior to undertaking the washing test, to allow the associated cold creep of the pressure sensitive adhesive and adhesion to the glass surface to fully develop. The glass article may be a returnable glass bottle such as used for beverages. In the test, the label to be tested may be prepared to dimensions of 50mm (width) x 70mm (height) and hand applied to the glass article using a wiper blade mechanism to simulate automatic application. The washing test may be conducted in a stainless steel bath filled with 12 litres of hot washing fluid agitated at least 15cm in depth via an overhead stirrer at 250 rpm,

simulating the conditions within the main soak of an industrial washing environment. The bath is maintained at a temperature of 80°C, 2.0% NaOH concentration and with a 0.1% addition of P3 Stabilon Plus, a washing additive supplied commercially by Ecolab.

Example

A water-borne, permanent pressure sensitive adhesive available from HB Fuller against the reference SE5279 was coated onto a permeable facestock material (Regenerated Cellulose Film of thickness 45 μm) and a non-permeable facestock (Biaxially Oriented Polypropylene of thickness 50 μm), at a coat weight of 20gsm.
The samples were prepared to dimensions of 50mm (width) x 70mm (height) and hand applied to a returnable glass beer bottles using a wiper blade mechanism to simulate automatic application. The labelled bottles were then allowed a minimum of 7 days standing time at ambient conditions (23°C, 50% RH) prior to undertaking the washing tests, to allow the associated cold creep of the PSA and adhesion to the glass surface to fully develop. The washing tests were conducted in a stainless steel bath filled with 12 litres of hot washing fluid agitated via an overhead stirrer at 250 rpm, simulating the conditions within the main soak of an industrial washing environment. The bath was maintained at a temperature of 80°C, 2.0% NaOH concentration and with a 0.1% addition of P3 Stabilon Plus, a washing additive supplied commercially by Ecolab.

It was determined that in conjunction with the Regenerated Cellulose Film, the wash-off performance of this adhesive was less than 180 seconds.

It was also determined that in conjunction with the BOPP film, the wash-off performance of this adhesive was under 480 seconds.

As previously stated, the outer surface of the adhesive layer, prior to application to an article, may include a plurality of micro-channels which extend to the periphery of the label. Thus surface micro-channels are provided within the adhesive channels layer which may extend all or part way across the label, when viewed in the plane of the label. In some embodiments, the channels may only extend part way into the label, it being sufficient for the channels to extend to the edge of the label and permit the hot washing water to lift the outer edge of the label from the article to which the label is affixed. Once the outer edges of the label have been lifted, this will expose the inner areas of the label to the hot water which, in some circumstances, will be sufficient to facilitate complete removal of the label from the article.

The size and density of the micro-channels in the adhesive layer may depend upon a number of factors, such as the elasticity of the face material and adhesive. However, as a general guideline, the channels will normally have a depth of no greater than about 15μm, no less than 3μm and most preferably in the range of 5μm to 10μm. The channels will normally have a width of no greater than about 300μm and no less than about 10μm and preferably in the range of about 50μm to 150μm. At least some of the channels may cross each other to form a network of inter-linked channels, such as a regular grid. Alternatively, a series of generally parallel channels may be provided which do not interlink. The arrangement of channels may be in the form of a regular pattern, but this is not essential and labels in which the channels are randomly arranged are also considered to be within the scope of the present application.

The precise number and disposition of such channels can be determined empirically by routine experimentation. Normally, a minimum distance between the micro-channels will be about 10μm, preferably about 250μm, to ensure a sufficient amount of exposed adhesive is provided to bond to the article satisfactorily. A maximum distance between adjacent channels may be of the order of up to about 10mm, and normally will be no more than 5mm.

In one presently preferred embodiment of the invention, the micro-channels comprises two series of parallel channels which intersect each other substantially at 90°.

The cross-sectional shape of the channels is not critical. For example, the channels may each be defined by a base and a pair of generally parallel side walls which extend away from the base. Alternatively, the channels may have a concave cross-sectional shape. This latter arrangement is typically obtained with most pressure sensitive adhesives.

The micro-channels in the outer surface of the adhesive layer may be formed by a process in which the composite material is subjected to a mechanical forming step in which the desired pattern of channels is formed on a resilient surface as a series of recessed areas on the block, which resilient surface is then used to press the surface of the composite to mechanically deform the layers of the composite. This process is otherwise referred to as "embossing". Embossing is a technique that produces raised or depressed sections on a surface in accordance with the shape and contours of the desired design. Known embossing techniques typically utilize a pair of dies having the design to be embossed found in them, one die generally being the negative of the other. When the pair of dies are brought together under pressure, with a piece of work

material between them, the design is transferred to the work material by deforming the material in the region of the design away from the original plane. Typically, in the present invention, the dies will be constituted by a pair of rollers, one of which has the desired pattern of channels provided on its surface, and the other of which is a backing roller against which the embossing roller presses.

The embossing of the composite structure may be carried out so that the pattern is applied to the face material or to the adhesive layer surface. It is preferred that the embossing is carried out so that the pattern is applied through the face material.

In the embossing method of the invention, the embossing roller may be formed of a metal in which the desired pattern of channels is etched. The depth of the channels, and their desired shape and disposition will correspond to the desired depth and disposition of channels in the adhesive layer, as described above. When the composite structure is embossed, the pattern of channels is transferred directly to the composite layer. The backing layer of the structure is a polymeric film, and in some instances, for example where the film is a polyolefin film such as polypropylene, the polymer film will deform elastically and shortly thereafter return to its original form, without any pattern of channels remaining in it. However, the material of the pressure sensitive adhesive has a very high viscosity and does not flow appreciably under the conditions under which the labels are made or used. Thus, the pattern of channels will remain in the surface of the adhesive layer for a prolonged period of time.

An example of an embossed label is shown in Figures 1 and 2. The label is embossed over its entire surface with a pattern appropriate to create a network of micro-channels with interlinking in both directions of the label and that would extend to the periphery of the cut labels. The depth of the embossing was 7-10μm and the width of the channels prior to application was 100 – 125μm.

Adhesive modifying agent

As previously stated, the outer surface of the adhesive layer, prior to application to an article, may include a printed modification to the adhesive surface.

In one embodiment of the invention, the adhesive modifying agent serves to deaden the adhesive where printed. In other words, the adhesive modifying agent lowers the level of adhesion to the article and also may allow ingress of washing fluid, such as a caustic solution through the voids between the adhesive layer and the article.

In another embodiment of the invention, the adhesive modifying agent incorporates a component which is soluble in the washing fluid, for example is soluble in a hot caustic solution. Thus, the adhesive modifying agent, where printed, lowers the level of adhesion to the article and allows ingress of caustic through the channels created by the printed pattern which is readily solubilised in contact with the hot washing fluid

In another embodiment of the invention, the adhesive modifying agent incorporates a component which swells when exposed to heat or water and, in particular, swells when exposed to hot washing fluid such as a caustic solution. Thus, the adhesive modifying agent, where printed, swells in the presence of the washing fluid and so lowers the level of adhesion to the article and allows ingress of caustic through the channels created by the printed pattern.

A label as shown in Figures 3 and 4 may be produced by printed on a narrow web printing press. By de-laminating the web it was possible to flexographically print onto the adhesive surface with a UV-curing adhesive deadening varnish (reference L301, supplier Paragon Inks) before re-laminating the web through a nip roller assembly. The web was printed with a range of patterns as shown in Figure 4 using a round dot printing structure;

An allover pattern was applied at percentages of 3%, 5%, 10%, 20%, 30% & 40%

A 5mm border to the label was applied at percentages of 3%, 5%, 10%, 20%, 30% & 40%

A 10mm border to the label was applied at percentages of 3%, 5%, 10%, 20%, 30% & 40%

The above percentages refer to the area of the original artwork occupied by the dots. In practice, variations that occur during the preparation of the printing plate form the artwork, and then in the printing process, result in an increase in the size of the dotsof adhesive modifying agent applied to the adhesive. By way of example, a density of 3% in the original artwork can result in a density of approximately 15% on the adhesive.

The varnish was applied using an anilox roller of line count 650 and volume 3.5 cc/m2.

It will be appreciated that the embossing technique and the application of an adhesive modifying agent are optional features which are not essential to the broadest scope of the present invention. With appropriate formulation of the adhesive, labels will wash off adequately in many circumstances. The use of embossing and/or an adhesive

modifying agent may nevertheless be appropriate in some conditions if accelerated washing off of the label is required.

CLAIMS

1. A label which is removable from an article under the action of a hot washing fluid, which label comprises a backing layer which is a polymeric film and an adhesive layer for bonding the label on to the article, the adhesive layer comprising a pressure sensitive and heat sensitive adhesive material.

2. A label as claimed in claim 1, in which the adhesive material is a water-borne adhesive material.

3. A label as claimed in claim 1, in which the adhesive material is a solvent-borne adhesive material.

4. A label as claimed in claim 1, in which the adhesive material is a hot melt material.

5. A label as claimed in any one of the preceding claims, in which the adhesive material is non-resinated.

6. A label as claimed in any one of the preceding claims, in which the adhesive material contains a surfactant.

7. A label as claimed in any one of the preceding claims, in which the adhesive thickness is not greater than 15 g/m^2.

8. A label as claimed in claim 7, in which the thickness of the adhesive layer is not less than 12 g/m^2 and not more than 14 g/m^2.

9. A label as claimed in any one of the preceding claims, which, prior to its application to an article, includes a plurality of micro-channels which extend to the periphery of the label.

10. A label according to claim 9, wherein the micro-channels extend all or part way across the label, when viewed in the plane of the label.

11. A label according to claim 9, wherein the channels only extend part way into the label.

12. A label according to any one of claims 9 to 11 wherein the channels have a depth of no greater than about 15μm.

13. A label according to claim 12, wherein the channels have a depth of no greater than about 10μm.

14. A label according to any one of claims 9 to 13 wherein the channels have a depth of at least 1μm.

15. A label according to claim 14, wherein the channels have a depth of at least 5μm.

16. A label according to any one of claims 9 to 15 wherein the channels have a width of no greater than about 250μm.

17. A label according to claim 16, wherein the channels have a width of no greater than about 150μm.

18. A label according to any one of claims 9 to 17 wherein the channels have a width of no less than about 10μm.

19. A label according to claim 18, wherein the channels have a width no less than about 50μm.

20. A label according to any one of claims 9 to 19 wherein the minimum distance between adjacent channels is about 10μm

21. A label according to any one of claims 9 to 20 wherein the maximum distance between adjacent channels is 10mm.

22. A label as claimed in any one of the preceding claims, in which the outer surface of the adhesive layer, prior to its application to an article, is provided with a coating of an adhesive modifying agent.

23. A label as claimed in claim 22, in which the adhesive modifying agent is a UV-cured adhesive deadening varnish.

24. A label as claimed in claim 22 or 23, in which the adhesive modifying agent is applied to the adhesive surface through direct printing.

25. A label as claimed in claim 24, in which the adhesive modifying agent is applied to the adhesive surface by a screen, flexographic, letterpress or gravure print process.

26. A label as claimed in any one of claims 22 to 25, in which the adhesive modifying agent is applied to the full surface of the label.

27. A label as claimed in any one of claims 22 to 25, in which the adhesive modifying agent is applied in a pattern.

28. A label as claimed in any one of claims 22 to 27, in which the adhesive modifying agent is applied as a series of dots.

29. A label as claimed in any one of claims 22 to 28, in which the adhesive modifying agent is applied at a density in the range of 5% to 10%.

30. A label as claimed in any one of claims 22 to 28, in which the adhesive modifying agent is applied in a pattern of parallel or intersecting lines.

31. A label as claimed in claim 1, and substantially as described herein.

32. An article carrying a label in accordance with any one of the preceding claims.

33. An article as claimed in claim 32 which is a bottle.

34. A method of removing from an article a label as claimed in claim 1, the method comprising:

(i) subjecting the article to a preheating step, in which the article is heated to a temperature in excess of 40°C, whereby the peel strength of the adhesive is reduced by not less than 40%, and, subsequently,

(ii) subjecting the article to a washing process in which the article is (a) immersed in an agitated washing fluid and/or (b) sprayed with a washing fluid, whereby the agitation and/or spraying separates the label from the article.

35. A method as claimed in claim 34, in which the preheating step comprises immersing the article in hot water at a temperature not less than 50°C.

36. A method as claimed in claim 34 or 35, in which the washing fluid comprises an alkaline solution.

37. A method as claimed in claim 36, in which the alkaline solution comprises an aqueous solution of sodium hydroxide.

38. A method as claimed in any one of claims 34 to 37, in which the washing fluid includes a surfactant.

39. A method as claimed in any one of claims 34 to 37, in which the washing fluid is at a temperature in excess of 60°C.

40. A method as claimed in claim 34 and substantially as described herein.

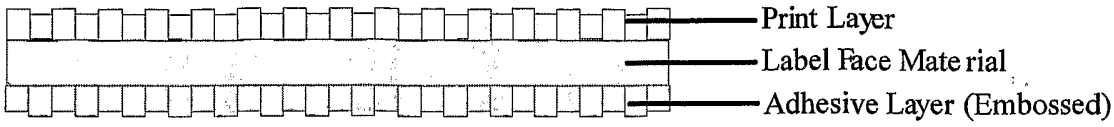

Figure 1

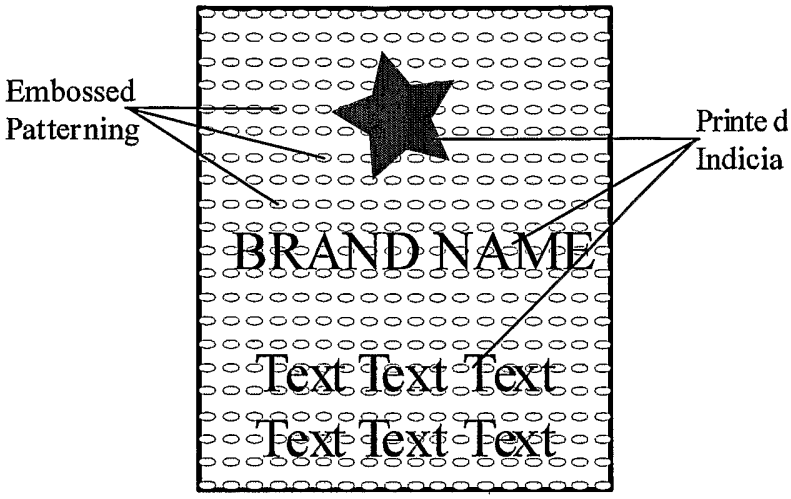

Figure 2

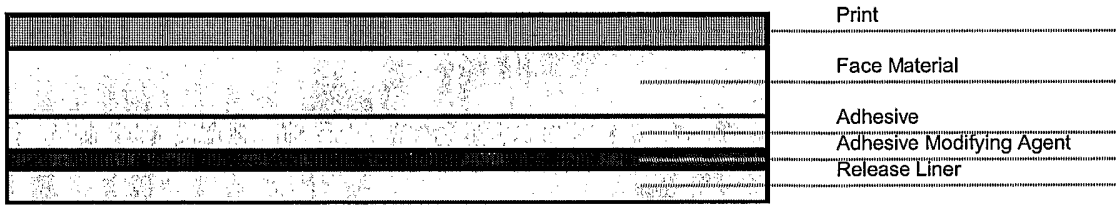

Figure 3

INTERNATIONAL SEARCH REPORT

ional Application No
PCT/GB2005/000898

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 C09J7/02 G09F3/10 B08B9/08 G09F3/02

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

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 C09J G09F B08B

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

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

EPO-Internal, WPI Data, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 03/099953 A (3M INNOVATIVE PROPERTIES COMPANY; YANG, JIE; CALLAHAN, KENNETH J.,; LU) 4 December 2003 (2003-12-04) page 1, line 29 - page 3, line 5 page 8, line 30 - page 10, line 11 page 21, line 11 - page 23, line 5; figures	1-30, 32-39
X	WO 02/074874 A (UCB, S.A; GAVEL, THIERRY; MIDDLETON, WAYNE; NASIB, VELI; FALETTI, GIAN) 26 September 2002 (2002-09-26) abstract; claims	1-8,22, 32-40

-/--

[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

° Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than 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.

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
13 June 2005	20/06/2005
Name and mailing address of the ISA European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Authorized officer Meier, S

Form PCT/ISA/210 (second sheet) (January 2004)

INTERNATIONAL SEARCH REPORT

ıtional Application No
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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 2003/207065 A1 (SHER FRANK T ET AL) 6 November 2003 (2003-11-06) abstract paragraph '0008! - paragraph '0009! paragraph '0016! - paragraph '0022! paragraph '0052! - paragraph '0057!; figures; examples	1–30, 32-39
X	US 6 680 097 B1 (AMBERGER WERNER ET AL) 20 January 2004 (2004-01-20) cited in the application column 2, line 60 - column 4, line 37 column 6, line 45 - column 7, line 44 claims 35-46	1–8, 22-30, 32-39
X	EP 1 055 715 A (OJI-YUKA SYNTHETIC PAPER CO. LTD; YUPO CORPORATION) 29 November 2000 (2000-11-29) abstract paragraph '0043! - paragraph '0048!; claims 1-6,14-18	1–30, 32-39
X	WO 03/087251 A (3M INNOVATIVE PROPERTIES COMPANY) 23 October 2003 (2003-10-23) abstract page 3, line 14 - page 6, line 32; figures	1–30, 32-39

Continuation of Box II.2

Claims Nos.: 31, 40

Present claims 31 and 40 relate to an extremely large number of possible products and methods. In fact, the claims contain so many options, variables, possible permutations and provisos since the refer to the description that a lack of clarity (and conciseness) within the meaning of Article 6 PCT arises to such an extent as to render a meaningful search of the claims impossible. In addition, according to R. 6.2(a) PCT and PCT-Guidelines 5.10 "The claims must not,....,rely on references to the description or drawings except where absolutely necessary". Consequently, no search has been carried out for claims 31 and 40 of the present application.

The applicant's attention is drawn to the fact that claims relating to inventions in respect of which no international search report has been established need not be the subject of an international preliminary examination (Rule 66.1(e) PCT). The applicant is advised that the EPO policy when acting as an International Preliminary Examining Authority is normally not to carry out a preliminary examination on matter which has not been searched. This is the case irrespective of whether or not the claims are amended following receipt of the search report or during any Chapter II procedure. If the application proceeds into the regional phase before the EPO, the applicant is reminded that a search may be carried out during examination before the EPO (see EPO Guideline C-VI, 8.5), should the problems which led to the Article 17(2) declaration be overcome.

INTERNATIONAL SEARCH REPORT

...ernational application No.
PCT/GB2005/000898

Box II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)

This International Search Report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. [ ] Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:

2. [X] Claims Nos.: 31, 40
because they relate to parts of the International Application that do not comply with the prescribed requirements to such an extent that no meaningful International Search can be carried out, specifically:
see FURTHER INFORMATION sheet PCT/ISA/210

3. [ ] Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. [ ] As all required additional search fees were timely paid by the applicant, this International Search Report covers all searchable claims.

2. [ ] As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.

3. [ ] As only some of the required additional search fees were timely paid by the applicant, this International Search Report covers only those claims for which fees were paid, specifically claims Nos.:

4. [ ] No required additional search fees were timely paid by the applicant. Consequently, this International Search Report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

[ ] The additional search fees were accompanied by the applicant's protest.

[ ] No protest accompanied the payment of additional search fees.

INTERNATIONAL SEARCH REPORT

Information on patent family members

ional Application No
PCT/GB2005/000898

Patent document cited in search report		Publication date	Patent family member(s)			Publication date
WO 03099953	A	04-12-2003	AU	2003251312	A1	12-12-2003
			EP	1532220	A1	25-05-2005
			WO	03099953	A1	04-12-2003
WO 02074874	A	26-09-2002	CA	2440240	A1	26-09-2002
			CN	1501967	A	02-06-2004
			WO	02074874	A1	26-09-2002
			EP	1381654	A1	21-01-2004
			JP	2004525414	T	19-08-2004
			US	2004197512	A1	07-10-2004
US 2003207065	A1	06-11-2003	US	2003124293	A1	03-07-2003
			US	6197397	B1	06-03-2001
			AU	737778	B2	30-08-2001
			AU	3290897	A	31-07-1998
			BR	9714191	A	28-03-2000
			CA	2274750	A1	09-07-1998
			CN	1242037	A ,C	19-01-2000
			DE	69715250	D1	10-10-2002
			DE	69715250	T2	30-04-2003
			DK	951518	T3	06-01-2003
			EP	0951518	A1	27-10-1999
			ES	2183190	T3	16-03-2003
			JP	2001507732	T	12-06-2001
			JP	2003145660	A	20-05-2003
			NZ	336083	A	29-06-2001
			WO	9829516	A1	09-07-1998
US 6680097	B1	20-01-2004	AU	4261199	A	14-11-2000
			CA	2370038	A1	19-10-2000
			WO	0062273	A1	19-10-2000
EP 1055715	A	29-11-2000	EP	1055715	A1	29-11-2000
			US	6576325	B1	10-06-2003
			WO	0036040	A1	22-06-2000
			TW	467948	B	11-12-2001
WO 03087251	A	23-10-2003	US	2003190458	A1	09-10-2003
			AU	2003219744	A1	27-10-2003
			BR	0308439	A	18-01-2005
			EP	1492851	A1	05-01-2005
			WO	03087251	A1	23-10-2003