April 28, 1964 F. W. MACKENZIE 3,131,106

ADHESIVE TRANSFERS

Filed June 22, 1961

FIG. 1

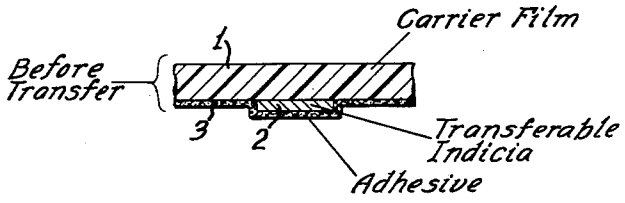


FIG. 2

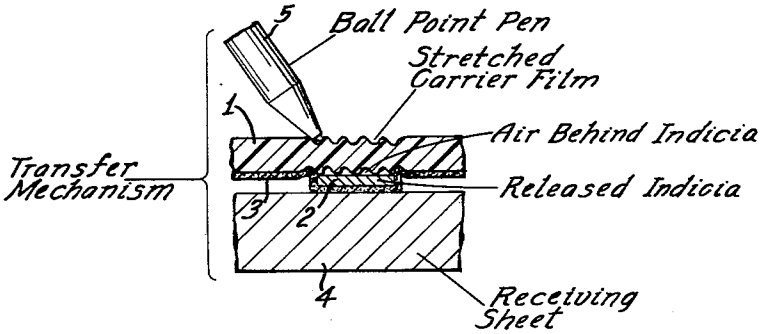
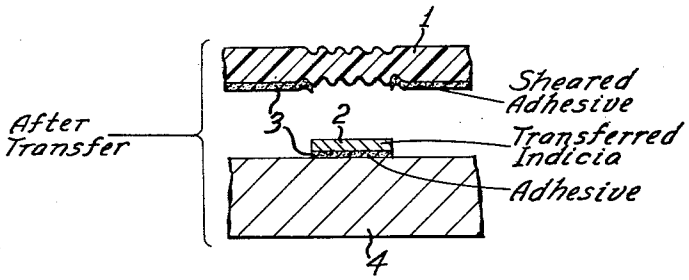


FIG. 3



United States Patent Office 3,131,106 Patented Apr. 28, 1964

3,131,106

ADHESIVE TRANSFERS

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Filed June 22, 1961, Ser. No. 118,765
Claims priority, application Great Britain June 24, 1960
5 Claims. (Cl. 156—230)

This invention relates to adhesive transfers (decalcomanias) and more particularly to a form of transfer material in which an image, design or printed matter (hereinafter generally referred to as "indicia") may be transferred from a carrier sheet to a further support. The invention includes transfer materials, their production and the processes of their use.

Transfer materials consisting of a carrier sheet carrying an indicia which can be transferred bodily from the carrier sheet to a further support are very well known indeed and a great deal of effort has been directed to the production of such materials which will permit transfer, in close register, of any indicia with ease, speed and reliability and which will give consistently good results.

In the earlier days of the art, attention was mainly directed to transfer materials which required the application of water to release the indicia. Such transfer materials were usually difficult to make, complicated in construction, and difficult to store without deterioration. Moreover the necessity for using a liquid to effect the transfer gave difficulties in their application and in some cases rendered them useless for desired purposes.

In more recent years attention has been directed to the production of transfer materials from which the indicia could be transferred without the use of any treatment liquid, so called "dry transfer materials." In some such materials heat is required in order to release the indicia from the carrier support, e.g. for the purpose of softening a waxy release layer. It has also been proposed, more recently, to apply to the surface of the indicia a pressure sensitive adhesive so that the indicia, being very firmly adherent to the pressure sensitive adhesive layer would release from the carrier support and adhere, with the said adhesive, to another support.

In general, with this latter type of product it has been necessary to provide a protective sheet over the pressure sensitive adhesive, which sheet was peeled away as a step immediately prior to the transfer of the indicia to another support. The protective sheet was necessary since otherwise the transfer material was impossible to handle as a commercial article owing to its readiness to stick to anything placed in contact with it, i.e. sheets of such material could not be stacked for packing purposes. Moreover the necessity for removing the protective sheet immediately before using the transfer material led itself to two major difficulties. On the one hand the actual operation of removing the protective sheet not infrequently pulled away from the carrier sheet the pressure sensitive adhesive and part, or the whole, of the indicia and, on the other hand, even if the protective sheet was peeled off without damage to the other layers it left a material which because of its readiness to stick to anything on the lightest contact could not easily be manipulated to position it for accurate register of the indicia to the surface where it was required.

If it is provided that the pressure sensitive adhesive is present only in register with the elements of the indicia this difficulty is overcome but in practice it is extremely difficult to maintain exact register in successive printing operations and it usually arises that the indicia transfers with an edging of imperfectly registered adhesive. This edge is unsightly and, being tacky, tends to pick up dirt

after transfer so that the transferred indicia may become spoilt in a very short time by the accretion of dirty marks round the elements of the indicia.

Absolute accuracy of register is difficult to achieve so that such methods have usually only been employed where the indicia are of very simple form, e.g. straightforward geometrical designs.

Furthermore, dry transfer materials of the type just discussed have usually used a paper carrier support, sometimes a laminar product of a fair degree of complication, which has had a very limited transparency or translucency (if indeed it was not opaque) so that accurate positioning of the transfer material, when the transfer is being effected, is the more difficult. Moreover, such carrier sheets based on paper have often suffered from the disadvantage of dimensional variation with atmospheric conditions, and have often had a serious tendency to curl.

As a result of very considerable research and experiment it has been found possible to overcome the aforesaid difficulties and it is a primary object of the present invention to provide a form of transfer material which is stable on storage, very simple indeed to use and which gives consistently good transfers.

According to the present invention there is provided a transfer material comprising a light transmitting extensible base sheet printed with indicia on one face and said indicia supercoated with an adhesive, the adhesive being of a character which will not adhere to another surface under light finger pressure but which will adhere to another surface when a substantial pressure is applied to the reverse side of the base sheet, whereby when the base sheet is subsequently lifted away the indicia remains, stuck by the adhesive, on said other surface. In this aspect of the present invention, this application is a continuation-in-part of my copending application Ser. No. 787,756, filed January 19, 1959, now abandoned.

More specifically according to the present invention there is provided a transfer material comprising a carrier sheet consisting of a sheet of transparent or translucent film of high dimensional stability under normally varying conditions of temperature and humidity but which is readily capable of stretching on application of tension thereto, indicia in printing ink carried by said carrier sheet, said printing ink being based essentially on a polymeric material and containing a plasticiser therefor, and a thin layer of a pressure sensitive adhesive in register with said indicia or extending over the whole of the printed area of the carrier sheet on the printed side, the adhesion between the said indicia and the carrier sheet being reducible by local stretching of the carrier sheet in the region thereof, and the pressure sensitive adhesive being substantially non-bonding at pressures less than 50 lbs. per square inch.

It is to be noted that if, in a transfer material according to the present invention the pressure sensitive adhesive layer is omitted, then the application of local pressure to the reverse side of the carrier causes the printed indicia to separate integrally from the carrier sheet. This result is achieved by selecting a carrier sheet which is readily capable of stretching on application of tension thereto and formulating the ink so that the indicia will so separate on the local application of stretching tension to the carrier sheet, i.e. the effect of such stretching must be to apply a force to the indicia which is greater than the adhesive force between the indicia and the support.

There is thus a fundamental difference between the transfer materials employed in the application of an important aspect of the present invention and those of the prior art referred to above. The prior art materials have relied essentially on the use of a surface adhesive of

greater adhesive power to the transfer surface and to the indicia than the adhesive power of the indicia to the carrier, i.e. the surface adhesive has acted to pull the indicia away from the support and hold it to the transfer surface. In the preferred materials of the present invention, in contrast, the separation of the indicia from the support derives essentially from a manipulation of the support and not by the pulling power of some applied adhesive, and the thus-loosened indicia are simply stuck by the high pressure adhesive to the transfer surface. In consequence it is made possible to employ an adhesive which is not a bonding adhesive at low pressures.

Transfer materials according to the present invention are easy to handle since the pressure sensitive adhesive will not bond to anything with which it comes into contact unless a substantial pressure is applied. It is accordingly not necessary to provide a protective sheet semi-permanently stuck to the adhesive surface. It is desirable in practice to interleave the transfer materials with, for example, silicone-treated interleaving paper but this paper does not stick tightly to the adhesive layer and will usually separate under its own weight.

Furthermore, the transparency or translucency of the support sheet enables the transfer material to be accurately positioned for use since the indicia can be seen through the back of the sheet. The whole product is stable under normal storage conditions and when required for use it is only necessary to apply it to the surface on to which the transfer is to be made, and apply a pressure in excess of 50 lbs. per square inch to the back of the carrier. The indicia then release from the support sheet and become adherent to the said surface. In practice it is usually necessary only to apply pressure to the back of the sheet with a hard object in the area of the indicia, covering at least part of the perimeter of the elements of the indicia, using a sufficient pressure to effect the release and simultaneous bonding.

When the adhesive is superimposed on the printed indicia only, and in accurate register therewith, there is no danger of its being transferred to form an edging to the transferred indicia which may pick up dirt.

If the adhesive is applied as a thin layer over the whole surface of the printed side of the carrier it is found that, provided it has a lower tensile strength than that of the printing ink and good adhesion to the non-printed areas of the carrier, it will shear round the elements of the printed indicia so that only the adhesive coated on the printed indicia will transfer.

Whilst various types of translucent or transparent film materials are suitable for use as carrier sheets in the present invention, it is found that particularly good results are obtained by the use of self-supporting films formed of polyalkylene derivatives, e.g. polyethylene, polypropylene and copolymers containing at least 90% of either of these polyalkylenes.

The preferred material for use is polyethylene film made of polyethylene of density 0.96 g./ml., melt index 0.2 (method of BS 1972); softening point 122° C. for 30° deflection (BS 1493); tensile strength 4200 p.s.i. (BS 2571); elongation at break 100–500%; Young modulus 2.0×10^5 p.s.i.; and water absorption less than .01% after 30 days immersion. The above polymer has a different molecular structure to conventional polyethylene, having linear and more regular polymer chains with very little side chain branching, which impart a high degree of crystallinity of about 93%. This structure provides the physical and chemical properties required for use in the present invention namely, stiffness, strength and elongation in very thin film form, and resistance to temperature, water and solvents.

The film support may be, for example, of thickness 0.001 to 0.008 inch and may have a glossy, matt or semi-matt surface and the transferred indicia will have a corresponding surface. A matt surface to the transferred indicia is sometimes of value where the transferred image is to be used for photographic reproduction.

The indicia are applied to the carrier support sheet by a printing operation. The formulation of the ink is of importance in order that it should meet the requirements set forth above, i.e. that it should adhere sufficiently to the support and yet be freed from the support by localised stretching of the support. The ink consists essentially of a high polymer and a plasticiser therefor. Generally it is found desirable to select a high polymer which, if used alone, would release spontaneously from the support and then to add to it sufficient of a plasticiser as to achieve the desired level of adhesion to the support.

Thus, for example, a clear ink medium may be prepared from a high polymer of cellulose nitrate of extra low, low or medium viscosity type, being an ester soluble grade, of nitrogen content of 11.8–12.3%, or a spirit soluble grade of nitrogen content 10.5% to 11.2%. The cellulose nitrate may be employed damped in butanol, or the like, with 65 to 70% polymer solids. The cellulose nitrate is dissolved in a solvent suitable in volatility for the particular printing process, and which does not dissolve, curl or distort the carrier sheet. Aliphatic or aromatic hydrocarbon solvents are undesirable, but esters, ether-esters, ketones, alcohols, ether-alcohols, ketone-alcohols are suitable on the polyethylene carrier support referred to above. A particularly suitable solvent for screen process printing, having low odour and a low evaporation rate, is ethylene glycol monoisopropylether acetate. A high polymer solution as thus formulated provides the necessary tensile strength of the dry ink film, but used without the inclusion of plasticiser would give indicia which would spontaneously release from the carrier sheet.

A plasticiser must be added to the high polymer solution to impart flexibility according to the known principles of lacquer formulation and also to control the release to precisely the required value by a most careful control of concentration. Two types of plasticiser may be used; one type is the so called solvent plasticiser which may provide part of the plasticiser content and is usually a non-polymeric material of low volatility, usually an ester, which is compatible with the high polymer and has a softening or dissolving action on it. Examples are, dioctyl phthalate, tributylcitrate, dimethylcyclohexyl adipate, trixyleneylphosphate. The second type of plasticiser is a non-drying oil (i.e., non-oxidising) modified polyester, compatible with the high polymer. The following oils, alcohols and acids may be variously employed in the production of such polyesters:

Oil	Alcohol	Acid
Castor oil.	Ethylene glycol.	Phthalic (or anhydride).
Hydrogenated castor oil.	Glycerol.	Isophthalic.
Coconut oil.	Pentaerythritol.	Terephthalic.
	Trimethylopropane.	Adipic.
		Sebacic.

A particularly advantageous plastiser is a polyglyceryl sebacate, of 72% castor oil content and having a viscosity *x*—*y*, Gardner-Holdt scale.

In general terms, using cellulose nitrate as the polymer there may be used, for 100 parts of cellulose nitrate, 20 to 150 parts of plasticiser, e.g. 55 to 150 parts of a castor-oil modified alkyd resin or hydrogenated castor-oil modified alkyd resin.

The printing inks used may be clear or pigmented or dyed and it is found to be particularly advantageous to formulate them so that they may be applied by silk-screen printing methods.

It will generally be found sufficient to so formulate the ink that it will adhere to the carrier sheet at pressures below 2 lbs. per square inch but will release from the

carrier sheet at pressures of 50 lbs. per square inch or more.

If desired the printed indicia, which may be a clear non-pigmented ink, may be overprinted with a normal type printing ink applied by any conventional method, e.g. by screen process, letterpress, offset litho, gravure, flexographic or bronzing processes, and this overprinting may constitute the means whereby colouration or opacity is imparted to the indicia.

The pressure sensitive adhesive is preferably applied as a solution, dispersion, or emulsion in organic solvent or water of a mixture of a high tack pressure-sensitive adhesive component and a non-tacky low tensile strength deformable component. The tacky component may be, for example, an acrylic, methacrylic ester or acid polymer or copolymer, a vinyl ether or ester polymer or copolymer, poly isobutylene or polybutene.

The non-tacky component may be a saturated long chain hydrocarbon or carboxylic acid ester or amide thereof or polymer of any of these or a long chain alcohol or a poly glycol.

The following example will serve to illustrate the invention:

Example

A clear ink which has excellent screen printing properties is formulated as follows:

	Parts by weight
33% solids cellulose nitrate low viscosity nitrogen 11.8–12.3% in ethylene glycol mono isopropyl ether acetate	12,500
72% castor oil glyceryl sebacate	2545
Dimethylcyclohexyl adipate	255

The polymer to total plasticiser ratio (which controls the release properties) is 100:67 in this ink.

A modification of this ink to include pigment is achieved by the addition of 6000 parts of rutile titanium dioxide and adjustment of the polymer to total plasticiser ratio to 100:81.

Such clear or pigmented ink medium is printed through a 180 stainless steel or nylon mesh screen, the design being formed by a hand cut stencil for simple designs and a photostencil for complex designs, to provide a dry ink film thickness of 0.0005±0.0002 inch, on a polyethylene film (polyethylene density 0.96 referred to above).

A high-pressure sensitive adhesive is formulated as follows. An aqueous emulsion is formed of 2 parts of an ester of a polyhydric alcohol and a fatty acid (e.g. ethylene glycol and lauric, palmitic or stearic acid) in 12 parts of water. To this is added 5 parts of an emulsion made up of:

	Parts
Water	45.27
Non-ionic surface active agent	1.2
Anionic surface active agent	0.3
Hydroxyethyl cellulose	0.55
Potassium persulphate	0.33
Borax	0.35
Copolymer of butyl acrylate (80%) and methyl methacrylate (20%)	52.0

to form a stable paste product. This is printed by the screen process and dried by evaporation. The resulting layer is of very low tack.

This adhesive may be printed in register with the indicia, but this is unnecessary, and excellent release, bonding and shear around the perimeter of any detail is obtained with an overall coat over the whole sheet or as a number of simple panels over the individual designs. A suitable wet coating weight is 1.1–2.2 grams per square foot which corresponds to a wet thickness of .0005-.001 inch.

A discontinuous coating, in which the coating is applied in a large number of small discreet dots covering say 70% of the sheet and with a frequency of 180 per linear inch provides ease of release and excellent shearing even at thick coats or with adhesives of higher shear strength than that just described.

The resulting transfer material may be used in the following manner:

The transfer material is placed, adhesive side down, onto the surface which is to receive the indicia, such as a sheet of drawing paper or film. The indicia is registered with the surface and then pressure is applied using a standard ball-point pen having a .040 inch diameter ball, using a light writing load of 2 ozs. This constitutes an average pressure of 1600 p.s.i., since the pressure band is of .010 inch width, and causes a noticeable stretch in the carrier sheet which exceed the yield point of the film and can be seen and felt when the pressure is released. The pressure is applied as a series of strokes over at least part of th perimeter of each ink area, when uni-directional lifting away of the carrier film is desirable. If pressure is applied over the whole of the perimeter of ink area the carrier film may be lifted away in any direction.

In practice a load of 1 to 4 ozs. is adequate, the lightest load being adequate when the final surface is very soft and permits a large stretching of the carrier sheet and the higher loads when the final support is hard, e.g. glass.

The presently preferred form of dry transfer of the present invention, and its method of use, are shown in the accompanying drawing in which

FIGURE 1 shows a diagrammatic section of the dry transfer materials;

FIGURE 2 shows in section the method and mechanism of transfer;

FIGURE 3 shows the result of the transfer operation, illustrating the residual elements of the transfer material in their new location.

Referring to these drawings, in FIGURE 1 there is shown the carrier film **1** carrying indicia **2** (one such is shown) and adhesive **3**. It will be noted that the adhesive extends beyond the margins of the indicia to overlap onto the carrier film **1**.

To effect the transfer, the dye transfer material is laid down with the surface of adhesive layer **3** in contact with a receiving sheet **4**. Pressure is applied to the area of the carrier film **1** behind the indicia **2** by means of a ball point pen **5**. The effect is to stretch the film **1** as shown and thus release the indicia **2** from the film **1**. Air thus enters between the indicia **2** and the film **1**.

The dry transfer material is then lifted away as shown in FIGURE 3. The area of adhesive **3** which overlapped onto the film **1** remains in position on the carrier film **1** due to shearing of the adhesive around the edges of the indicia. The indicia **2** however is adherent to the receiving sheet **4** by means of that part of the adhesive **3** which lay over the indicia **2**.

I claim as my invention:

1. As a new article of manufacture, a dry transfer sheet for applying printed indicia to a receiving sheet at a selected position on such receiving sheet under control of a user, said dry transfer sheet comprising a carrier sheet of polyethylene having high-release properties; indicia on said polyethylene carrier sheet in the form of ink films strong enough to retain their shape during transfer and having a mechanically breakable bond to said carrier sheet; a dry adhesive coating extending over the indicia, said coating being adherable to a selected position on a receiving sheet at ambient conditions upon rubbing with high localized force-per-unit-area over such position but said coating being immune to transfer-effecting adhesion upon application of light finger pressure at such position or over other portions of the transfer sheet, said adhesive coating having a greater affinity for a receiving sheet when subjected to high pressure as by rubbing with a stylus over a selected area thereof than the affinity between the indicia and the carrier sheet after application of such pressure, so that the transfer sheet can be placed against a receiving sheet and adjusted to accu-

rately locate indicia to be transferred in any selected position on the receiving sheet without adhesion of the adhesive coating to the receiving sheet and thereafter the indicia can be transferred dry by rubbing as aforesaid while in the selected position.

2. As a new article of manufacture, a dry transfer sheet for applying printed indicia to a receiving sheet at a selected position on such receiving sheet under control of a user, said dry transfer sheet comprising a light-transmitting carrier sheet at least one surface of which has high-release properties; indicia on the high-release surface of said carrier sheet in the form of ink films strong enough to retain their shape during transfer and having a mechanically breakable bond to said high-release surface of said carrier sheet; and a dry adhesive coating extending over the indicia, said coating being adherable to a selected position of another sheet at ambient conditions upon rubbing with high localized force-per-unit-area over such posiition but said coating being immune to transfer-effecting adhesion upon application of light finger pressure at such position or over other portions of the transfer sheet; said adhesive coating having a greater affinity for a receiving sheet when subjected to high pressure as by rubbing with a stylus than the affinity between the indicia and the carrier sheet after application of such pressure, so that the dry transfer sheet can be placed against a receiving sheet and adjusted to accurately locate indicia to be transferred in any selected position on the receiving sheet without adhesion of the adhesive coating to the receiving sheet and thereafter the indicia can be transferred dry while in the selected position by rubbing with high localized pressure over the indicia.

3. A transfer sheet, including a carrier sheet having a surface of a bond-resisting nature, indicia formed of coherent films capable of retaining their shape during transfer and having a mechanically breakable bond to the carrier sheet, and a coating of a flexible light-transmitting adhesive substance on said indicia having inadequate adhesion to a receiving sheet to effect transfer of said elements under light finger pressure but having transfer-effecting adhesion to the receiving sheet under heavy localized pressure equal to that produced by rubbing with a stylus while other elements on the transfer sheet not subjected to such heavy localized pressure remain on said carrier sheet, whereby said transfer sheet can be freely moved about relative to a receiving sheet to an accurately adjusted position while in contact wtih the receiving sheet, and heavy localized pressure can then be applied for transferring the indicia to an accurately determined position on the receiving sheet by localized pressure as aforesaid.

4. The method of applying printed indicia to a receiving sheet, including the steps of printing indicia on a carrier sheet with printing material that forms indicia

that are strong enough to retain their shape during transfer, the carrier sheet and the printing material being related to provide a mechanically breakable bond between the indicia and the surface of the carrier sheet; coating the indicia with an adhesive substance that does not cause transfer of the indicia to said receiving sheet under light finger pressure; thereafter placing a receiving sheet in confronting relation to the carrier sheet and adjusting the indicia to be transferred to a desired position on said receiving sheet and with the adhesive substance which covers the indicia being in contact with the receiving sheet; rubbing a stylus over the indicia to be transferred and separating said sheets to effect the dry transfer of said indicia to the desired position on the receiving sheet.

5. The method of applying printed indicia to a receiving sheet, including the steps of printing indicia on a carrier sheet with printing material that forms indicia strong enough to retain their shape during transfer, the carrier sheet and the printing material being related to provide a mechanically breakable bond between the indicia and the surface of the carrier sheet; coating the printed indicia with an adhesive substance that does not cause transfer of the indicia to said receiving sheet under light finger pressure; thereafter placing a receiving sheet in confronting relation to the carrier sheet with the indicia to be transferred at a desired position of said receiving sheet and with the adhesive substance which covers the indicia being in contact with the receiving sheet, said sheets being chosen so that at least one sheet is light-transmitting to facilitate the adjustment of the indicia to be transferred to the desired position on the receiving sheet, rubbing a stylus over the indicia to be transferred and separating said sheets to effect the dry transfer of the indicia to the desired position on the receiving sheet.

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