June 27, 1967 M. D. MATHIEU 3,328,190

TRANSFER COATING

Filed Dec. 23, 1963

FIG. 1

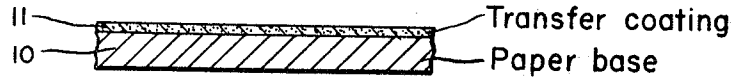


FIG. 2

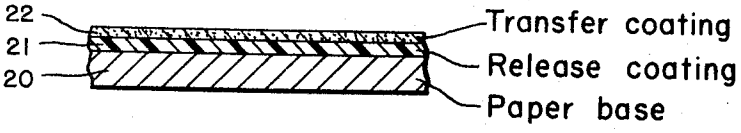
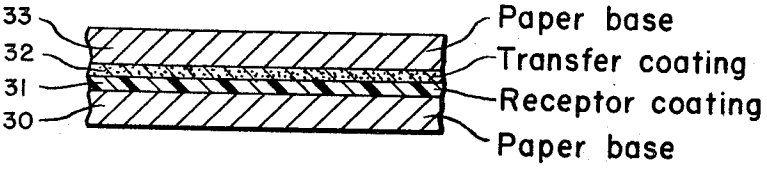


FIG. 3



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TRANSFER COATING

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10 Claims. (Cl. 117—36.1)

This application is a continuation-in-part of application Ser. No. 188,987, filed Apr. 20, 1962, and application Ser. No. 272,246, filed Apr. 11, 1963, both now abandoned.

This invention relates to transfer sheets such as those commonly known in the art as carbon paper. More particularly, the invention relates to transfer sheets prepared by applying a transfer coating to a flexible foundation, the transfer coating comprising closely packed spherical particles which are bound to each other by a sufficient amount of a non-waxy adhesive to hold the spheres together and to the flexible foundation, but insufficient to hold the transfer coating to the foundation when subjected to local pressure, such as by stylus or typewriter keys, etc. The spherical particles used according to this invention are advantageously prepared from organic polymer emulsions.

It has been known in the art that transfer sheets can be prepared by applying to a flexible foundation certain coating compositions which are characterized by their having a waxy nature. The ability of these transfer sheets to transfer an image to a receiving surface in response to local pressure is dependent upon the presence of a waxy material in the coating on the sheets. The waxy material is usually composed of mixtures of waxes, oils and oily substances.

The preparation of such wax-type coatings requires special emulsification equipment and the application thereof to a suitable base by the hot-melt type coating equipment which requires slow coating speeds.

The transfer sheets prepared according to this invention avoid many of the disadvantages inherent in the prior art waxy papers. The transfer sheets of this invention are simple and economical to prepare, and they do not involve the preparation of any rigid emulsification of a waxy material or special emulsification equipment. Using conventional coating equipment, high coating speeds can be used in the manufacture of these transfer sheets. The heat limitation factors, such as those which exist when waxy substances are present, are largely avoided. These tranfser sheets, moreover, are almost completely dry and smear-proof. They possess a high gloss, a high scuff resistance and do not require the presence of a solvent vehicle.

The transfer coating of this invention consists of closely packed, organic-polymer spheres bonded together and to a flexible foundation, such as paper, with an amount of adhesive sufficient to resist ordinary scuff and bending during normal handling, but in an amount less than that which would prevent release of the coating from the paper stock when subjected to local pressure. The transfer coating may advantageously contain at least about 70% by weight of hard organic polymer spheres and, since the void space between a closely packed system of polymer spheres is ordinarily no more than about 27%, the amount of adhesive used should be less than that amount which would completely fill the void space, i.e., less than 27% by weight of the coating. However, at least about 1% by weight of adhesive should be required. The transfer sheets of this invention may be prepared by mixing together an aqueous suspension of a suitable organic polymer; coloring matter such as a pigment or dye; and an adhesive, applying the coating to a flexible foundation, and drying the coating thereon.

The organic polymer particles, which are in discrete, particular form, closely approximate a system of closely packed spheres. These spheres have substantially no tendency to coalesce, that is, to deform from their essentially spherical geometry under manufacturing and in-use conditions. Therefore, an emulsion polymer system is advantageously employed which has a glass transition temperature higher than the highest temperature likely to be encountered. The term "glass transition" refers to the characteristic change in polymer properties from those of a relatively hard, brittle glassy material to those of a softer, more flexible, rubbery substance as the temperature is raised through the glass transition temperature. In practice, a glass transition temperature for the polymer of about 140° F. is normally satisfactory, since fast-dry production conditions, even at temperatures above 140° F., do not facilitate coalescence of emulsion polymer particles having this property. Also, finished product storage conditions rarely approach this temperature. The organic polymer spheres used according to this invention are therefore defined by their having a glass transition temperature sufficiently high so that the polymer spheres are not substantially deformed or coalesced at the temperatures at which the transfer sheets are prepared and stored. The use of these organic polymer spheres form a stable coating of close-packed, uncoalesced, hard, glassy spherical particles upon their application, as an aqueous emulsion, to the base stock followed by a rapid water removal operation.

A system of close-packed, spherical particles, such as obtained by rapid drying of a polystyrene emulsion film, contains up to about 27% void volume. At a typical coat-weight of 3.3# per ream, a film employing a polystyrene emulsion (manufactured by the Monsanto Chemical Company, under the tradename Lytron S–2, for example) is about 7 microns thick after water removal, corresponding to about 30 layers of close-packed polystyrene particles. It is advantageous to this invention that the average particle size of the emulsion particles be such that the individual voids created as a result of spherical close-packing can easily contain colored pigment particles. For example, use of polystyrene in conjunction with a copper phthalocyanine pigment dispersion, allows formation of a coating such that about 30 copper phthalocyanine pigment particles can be packed in each void formed among the hard polystyrene particles.

The organic polymer spheres employed in this invention are prepared by forming an aqueous emulsion of a resinous material having a glass transition temperature sufficiently high so that the polymer spheres are not substantially deformed or coalesced at the temperatures at which the transfer sheets are prepared and stored. These polymers include the so-called non-film forming thermoplastic resins as well as thermosetting resins. Although it is difficult at times to define a definite borderline between thermosetting resins and thermoplastic resins, the important point is that the resins should be non-film forming at application and drying temperatures and should be capable of being mixed in an aqueous medium with an adhesive and then applied to a suitable base sheet and then dried to form a coating containing loosely bound pigment-like resin particles, this coating being capable of being transferred to a receiving surface in response to local pressure. These polymers are used in emulsion form, and water is the preferred medium in which the polymers are dispersed. It is believed that other mediums could be employed so long as the essential relationship between the polymer spheres and the adhesive is maintained.

Various non-film forming resins can be used according to this invention including polystyrene such as marketed by Monsanto Chemical Company under the trade-

name Lystron S–2, polyacrylic resins such as those marketed by Rohm & Haas under the tradename Rhoplex B–85, polyvinyl chloride such as marketed by the Monsanto Chemical Company under the tradename Opalon 410, polystyrene latex such as marketed by Dow Chemical Company under the tradename Dow Latex 586, polyvinyl chloride latex such as marketed by the Dow Chemical Company under the tradename Dow Latex 700, polyvinyl chloride latex such as marketed by the Dow Chemical Company under the tradename Dow Latex 744–B, and the like. Other systems of spherical particles, of course, can be used along with, or in place of, the organic polymer spheres described above, so long as these spherical particles function in substantially the same way as those prepared from the organic polymer emulsions. It is preferred that these spherical particles have an average, diameter of about 0.05 to 0.3 micron. Depending upon the particular ingredients used and the degree of transferability desired, this range can, of course, vary. Larger polymer particles, for example, will tend to transfer more readily than smaller particles.

The transfer properties of the transfer sheets of this invention, as operational products, are directly related to the strength of the adhesive bonds between the polymer spheres and to the adhesion of the coating to the paper. In order to obtain clear, sharply-defined, transferred images, the adhesive need only bind the hard, spherical particles together in the vicinity of the contact points resulting from their close packing. Adhesive contents beyond this are conducive to a less sensitive transfer and adhesive levels below this lead to a powdery, ill-defined image, lacking desirable handling properties of the coated sheet.

Emulsion polymer systems can also be used as adhesives in this invention and it is advantageous that its particles deform readily within a short-drying time (5–15 sec.) at elevated temperatures. For example, an emulsion polymer, whose glass transition temperature is less than about 140° F., preferably less than 100° F. may be used. The drying conditions employed in this invention can involve the use of forced hot-air drying, up to 250° F. ambient temperature, at web speeds such that water evaporation takes place within 5 to 15 seconds dwell-time. Under these conditions the coated web temperature rarely rises above about 140° F. Emulsion polymers having a glass transition temperature less than about 100° F., can, under these conditions, be defined as film-forming systems, that is, their properties in a coating are characteristic of the polymer contained originally in the emulsion particles, rather than of the particles themselves. Any other well-known drying means can, of course, be used.

Preferably, the adhesive emulsion particles should be small compared to the close-packed, spherical particles, that is a $\frac{1}{10}$ ratio in diameters, in order to easily enter the voids between the latter and in order to migrate to the polystyrene contact points and thus exercise their adhesive nature. Use of appropriate concentrations of such film-forming emulsion polymers as adhesives, results in bonding between the hard polymer sphere not unlike that obtained with use of water soluble adhesives.

The objectives of this invention can also be met by use of large-particle-size emulsion polymer systems as adhesives (average diameters greater than $\frac{1}{10}$ that of the hard component), provided that these also fulfill the criterion of ability to flow, or coalesce, under the drying conditions usually employed. In the situation where emulsion adhesive particles approach the size of the hard polymer spheres, they initially contribute to the formation of the close-packed system (i.e., in the early stages of the water removal), as if by replacing some of the hard polymer spheres, but because of their deformability flow into intimate contact with, and act as a binder for, their nearest neighbors. Such emulsion polymers which form adhesive particles approaching the size of the hard polymer spheres act in a dual role in assuming part of the function of an adhesive and part of the function of the organic polymer spheres. Therefore, in a case where the adhesive particles approach the size of the polymer spheres and are partially coalesced during manufacturing conditions such that only about one-half of their adhesive properties are utilized, more adhesive can be used than when the adhesive particles are small in relation to the organic polymer spheres.

Various adhesives can be employed according to this invention. As stated above, the adhesives can either be of the emulsion type or the type soluble in aqueous systems. A combination or mixtures of the adhesives can also be used sometimes to advantage. Representative examples of suitable adhesives which can be used include polyvinyl alcohol such as that marketed by Du Pont under the tradename Elvanol 72–60, starches including starch ethers and chlorinated or oxidized starches, carboxymethylcellulose, alkali soluble partial pentaerythritol ester of wood rosin marketed by Hercules Powder Company under the tradename Pentalyn 255, gelatin, polyacrylamide, vinyl acetate and copolymers thereof such as marketed by The Borden Chemical Company under the tradenames Lemas 541–10 and Polyco 678–W. Further examples include hydrocarbon resins such as methylated paraffinic chain hydrocarbon resins of petroleum origin such as marketed by Pennsylvania Industrial Chemical Corp. under the tradename Piccopale A–22, acrylic polymers such as that available from Rohm & Haas marketed under the tradename Rhoplex B–15, as well as various butadiene-styrene copolymers (40–60 ratio) such as those marketed by The Borden Chemical Company under the tradenames Polyco 350–W, Polyco 2414, Polyco 2419 and by Dow Chemical Company under the tradename Dow Latex 630, acrylic-vinylidene chloride copolymers such as marketed by Rohm & Haas under the tradename Rhoplex R–9, as well as natural resins such as casein. Other non-waxy adhesives can also be used as will be apparent to those skilled in the art.

The carboxylated organic, water-soluble polymers, those which possess periodic carboxylic acid groups along the polymer chain, such as carboxymethylcellulose, or carboxy containing acrylic polymers are advantageously used, preferably in the range of about 1–5% by weight, as the adhesive and serve as a very flexible adhesive which tends to render the resulting transfer coating resistant to any deleterious effects which may be caused by storing the transfer sheets under humid atmospheric conditions.

Transfer coatings based on emulsion polymer adhesives sometimes suffer from deterioration in image quality and density when stored at temperatures between 60 and 140° F. and relative humidities from 40 to 100%. This effect is due to slight flow, or creep, of the emulsion adhesive during storage, thereby causing an increased adhesive contact. An increased adhesive contact in turn results in decreased image transfer sensitivity. Therefore, it is frequently advantageous to employ, in emulsion form, a so-called self-cross-linking adhesive, and adhesive which, by virtue of its latent chemical reactive groups, undergoes a chemical cross-linking reaction among and between the adhesive particles when the coating, containing the emulsion, is dried under normal drying conditions. Examples of self-cross-linking adhesives are vinyl acrylic copolymer emulsions manufactured by the National Starch and Chemical Company under the tradename X-Link 2833; aqueous dispersions of acrylic copolymers manufactured by the Rohm & Haas Company under the tradename Rhoplex K–3; compositions formed of 1–30% of an acrylic and/or methacrylic ester, a methylol compound and/or methylol-ether of (meth)acrylamide and optionally up to 20% of other copolymerizable monomers, obtained by polymerization in emulsion or solution as more fully described in Belgian Patent No. 617,606.

Any suitable dye or coloring matter, i.e., colored pigment particles, may be mixed in with the coated composition. Examples of such dyes which have been advantageously employed include phthalocyanine pigment dye

such as phthalocyanine blue marked by American Cyanamid under the tradename Calcotone D–100, and carbon black such as that manufactured under the tradename Aquablak B by Columbia Carbon Company. Advantageously, the particle size of the solid colored pigment is less than about 0.5 micron.

Various compatible plasticizers may be incorporated into the coating composition in small amounts as hereinafter described such as butyl benzyl phthalate, manufactured under the tradename Santicizer 160 as well as other well-known plasticizers which will be apparent to those skilled in the art.

The most advantageous proportions of spherical particles to adhesive used can be determined by routine experimentation and will vary depending upon the particular polymer and adhesive used, the roundness of the resinous polymer spheres, and also depending upon the quantity of coloring material present in the coating. For example, if the adhesive is water soluble, the amount of adhesive used is preferably about 1–5% by weight, whereas, if the adhesive is in emulsion form, the amount of adhesive used is preferably about 3–10% by weight. If the particle size of the organic polymer spheres is small, less adhesive will be necessary than if the particle size is large, and if the particle sizes are more evenly distributed, more adhesive is expected to be necessary than if the particle sizes are unevenly distributed. The amount of adhesive used should be such that together with the coloring matter, and any additives, the voids between the polymer spheres are not completely filled.

The amount of adhesive which can be used according to this invention would therefore normally be no greater than about 27% by weight of the dry transfer coating, but the amount used should be sufficient to bind the spheres together and to the flexible foundation during normal handling conditions. The optimum ratio in a polystyrene/polystyrene-butadiene coating has been found to be approximately 9:1 on a dry weight basis. The method of applying and drying the coating will also affect the resin/adhesive ratio. Lesser amounts of adhesive may be used under conditions of rapid drying since adhesive losses through penetration are not as high. When using the polystyrene resin (Lytron) a styrene-butadiene adhesive (Polyco 2414), and the phthalocyanine blue dye, the optimum ratio is found to be 12.5:1.3:0.3. When using the same adhesive and pigment with carbon black (Aquablak B), the styrene-butadiene content was advantageously increased to 1.5 instead of 1.3.

In addition to the organic polymers, adhesives, and coloring matter, various additives may be used to improve the handling characteristics or release characteristics. Such additives include calcium stearate, potassium stearate, ammonium stearate, polyoxyethylene glycols, lecithin, polybutenes, polyethylene emulsions, or soft waxes. The only requirement is that these additives, together with the adhesive and coloring matter, should not completely fill the voids between the organic polymer spheres, i.e., the organic polymer spheres should comprise at least about 70% by weight of the transfer coating.

The plasticizer which may be used should only be present in small amounts. The ratio of resin to plasticizer is advantageously about 100:1.25. This ratio can, however, go as high as about 100:5. The use of plasticizers in excessive amounts will substantially interfere with the transfer properties of the transfer sheets and must be avoided.

If desired, small amounts of semi-hard polymers up to about 5% by weight can be used to improve the aging characteristics of the transfer sheets. These semi-hard polymers are defined by their having a glass transition temperature between that of the organic polymer spheres and that of the adhesive. These semi-hard polymers partially deform under manufacturing and drying conditions.

The temperature at which the coating composition is blended together, of course, depends upon the particular components employed. A temperature of about 122° F. to 140° F. may be advantageously employed in most instances. In drying the coating, it is desirable to supply a suitable degree of heat in order to speed up the drying process. The only requirement as to this temperature is that it should not be sufficient to significantly soften the organic polymer spheres and destroy the particle nature of the non-film forming resin which would interfere with the transfer properties desired in the resulting sheet. In non-plasticized polystyrene formulations, for example, dryer temperatures should be less than about 212° F. Somewhat lower temperatures should generally be used with plasticized polystyrene since the softening point of the plasticized polystyrene is generally lower than that of the non-plasticized polystyrene. The specific drying temperature used with any particular formulation can be determined by routine experimentation by those skilled in the art.

The limitation on the drying temperature is quite advantageous and one aspect of the invention includes the step of heating of the transfer coatings to temperatures above the softening point of the organic polymer spheres so as to deactivate or destroy the transfer properties of the film. The fact that the transfer properties of the film can be destroyed by the simple expedient of heat, permits the transfer coatings to be rendered non-transferable after the transfer coating has served its purpose, and results in an original record (typewritten, for example) having a nontransferable coating on the back. In addition, the image transferred to a receptor paper can also be deactivated by heating the transferred image, thus improving the smudge-proofness and permanence of the transferred image.

The coating may be applied to any suitable backing material, usually paper. A coating applied at a coating weight of about 2 to 4 lbs. per ream 25" x 38"—500 (sheets) is desirable. This weight, however, could vary from about 1 to 7 lbs. per ream. The coating may be applied at any suitable solids content, generally ranging from about 10 to 40%. On an airknife coater, optimum results were obtained on a solids content in excess of 30%. The coating may be applied at a machine speed ranging from about 100 to 500 ft. per minute. Even higher machine speeds would probably be suitable.

The receiving surface, which accepts the transferred image from the transfer sheets of this invention, is a surface which advantageously has been treated with a "receptor coating." A specially treated paper, having a receptor coating, which can be used, is one which is coated with a composition containing, for example, a liquid dispersion of calcium carbonate having a relative sedimentation volume of between about 2 and 6 in admixture with an adhesive, the composition having a pigment/adhesive ratio of about between 7:1 and 2:1. Such a specially treated receptive paper is described more particularly in the copending application of William H. Hoge and Marshall S. Barbour, Ser. No. 79,980, filed Jan. 3, 1961, now U.S. Patent No. 3,118,782, assigned to the same assignee as the present application.

Thus one feature of this invention resides in the use of a combination of two coated papers, the transfer paper overlaying a receptor paper, wherein the transfer coating of the transfer paper is in contact with the receptor coating of the receptor paper. Local pressure applied to the transfer paper therefore results in a transferred image on the receptor paper corresponding in area to the area at which the local pressure was applied to the transfer paper. Alternatively, a transfer coating can be applied to one side of a paper and a receptor coating applied to the other side and then two or more of these sheets can be placed together so that the transfer coating of one sheet is in contact with the receptor coating of another sheet.

The transfer sheets of this invention can also be used

in conjunction with ordinary copy paper for reception of the image to be transferred.

A release coating between the paper base and the transfer coating can be used in this invention if desired. Such a release coating can advantageously be used to facilitate transfer of the transfer coating to ordinary copy papers or papers not having an image receptive coating applied thereto.

Various release coatings can be used according to this invention such as those composed predominantly of vinyl resins. Cellulosic, polyamide, acrylic or methacrylic resins and so forth can also be used. Further examples of release coatings include cellulosic resins such as hydroxylethyl cellulose polyamide, acrylic or methacrylic resins as well as inorganic materials such as sodium silicate or colloidal silica (e.g. silica manufactured by Monsanto Chemical Company under the tradename Syton). The release coating can be applied by solvent or water techniques as will be apparent to those skilled in the art. Specific examples of release coatings include 20 parts by weight vinylidene chloride, vinyl-chloride copolymer dissolved in 80 parts by weight methylethyl ketone, 5 parts by weight polyvinyl butyral dissolved in 80 parts by weight ethanol and 6 parts by weight of a silica filler.

FIGS. 1–3 of the accompanying drawing are diagrammatic, enlarged representations of cross-sections of three typical embodiments of the transfer sheets of this invention.

FIG. 1 shows a paper base **10** having a transfer coating **11** prepared according to the present invention adhered to the paper base. FIG. 2 shows a paper base **20** with a release coating **21** adhered thereto and the transfer coating **22** prepared according to this invention adhered to the release coating. FIG. 3 shows a combination of a paper base **30** having a receptor coating **31** adhered thereto and the receptor coating contacting the transfer coating **32** prepared according to this invention adhered to the paper base **33**. The application of pressure by means of a stylus or typewriter key on the top or uncoated side of paper base **33** results in a transfer of the transfer coating to the receptor coating on paper base **30** in areas coextensive with the application of the pressure.

In the following detailed example, which are given for illustrative purposes only, specific embodiments of this invention are shown. Parts are by weight.

EXAMPLE 1

100 parts of a non-film forming polystyrene aqueous emulsion containing 50% solids (Lytron S–2) were thoroughly mixed together with 89 parts of an aqueous suspension of carbon black containing 13.2% carbon black solids, 2.5 parts of butyl benzyl phthalate, 11.2 parts of a butadiene-styrene copolymer aqueous emulsion containing 48% copolymer solids and 120 parts of water. The copolymer contained a butadiene styrene ratio of 40–60. The coating composition thus prepared was then applied onto a base paper at a coat weight of 3 pounds per ream and the coating dried at a temperature of 140° F. The dry transfer coating possessed a high gloss, high scuff resistance and was substantially smudge-proof. The coating on the base paper was then placed in contact with a sheet of conventional bond white paper and pressure applied by means of a stylus to the backside of the paper having the transfer coating applied thereto. The coating readily transferred to the bond paper receiving surface underlying the coating in the areas coextensive with the applied pressure. The image transferred to the bond receiving surface was sharp and distinct, possessed a high scuff resistance, was substantially smudge-proof and dry.

The coated paper, to which the stylue had been applied, was heated to a temperature above the softening point of the polystyrene particles contained therein for a sufficient time to substantially eliminate the particle nature of the non-film forming polystyrene pigment particles. This heating step rendered the coating non-transferable.

The bond paper, having the transferred image thereon, was heated to a temperature above the softening point of the polystyrene particles in the same manner as the sheet having the transfer coating thereon. The image was completely smudge-proof and dry and possessed a high degree of permanence.

The coated surface of another portion of the transfer sheet prepared according to this example was placed against a sheet of paper having a receptive coating thereon prepared according to Example 1 in application Ser. No. 79,980, filed Jan. 3, 1961. When pressure was applied by means of a stylus to the backside of the paper having transfer coatings adhered thereto, the coating transferred event more readily to give an extremely clear and sharp image on the receptive coating of the underlying sheet.

EXAMPLE 2

A transfer sheet was prepared in the same manner as set forth in Example 1 except that the transfer coating composition was formed by thoroughly mixing together 25 parts of a non-film forming polystyrene aqueous emulsion containing 50% solids, 1.7 parts of an aqueous alkylketene dimer containing 30% dimer solids marketed under the tradename Aquapel 380 by the Hercules Powder Company, 22.3 parts of an aqueous carbon black containing 13.2% carbon black solids, 2.8 parts of the same butadiene-styrene copolymer referred to in Example 1 containing 48% copolymer solids and 30 parts of water.

EXAMPLE 3

A transfer sheet was prepared in the same manner as set forth in Example 1 except that the coating composition was formed by thoroughly mixing together 25 parts of a non-film forming polystyrene aqueous emulsion containing 50% solids, 2.8 parts of the same butadiene-styrene copolymer used in Example 1 containing 48% copolymer solids, 15 parts of highly dispersed phthalocyanine blue pigment paste containing 2% solids and 30 parts water.

EXAMPLE 4

A transfer sheet was prepared in the same manner as set forth in Example 3 except that 25.5 parts of a non-film forming polystyrene latex containing 49.5% solids (Dow Latex 586) were substituted for the 25 parts of the polystyrene used in Example 3.

EXAMPLE 5

A transfer sheet was prepared in the same manner as set forth in Example 3 except that 25 parts of a non-film forming polyvinyl chloride latex containing 60% solids (Dow Latex 700) were substituted for 25 parts of the non-film forming polystyrene used in Example 3.

EXAMPLE 6

A transfer sheet was prepared in the same manner as set forth in Example 3 except that 33 parts of a non-film forming polyacrylic aqueous emulsion containing 38% solids (Rhoplex B–85) was substituted for the 25 parts of the polystyrene and 21.5 parts of water was used instead of 30 parts of water.

EXAMPLE 7

A transfer sheet was prepared in the same manner as set forth in Example 3 except that 25 parts of a non-film forming polyvinyl chloride aqueous emulsion containing 50% solids (Opalon 410) were substituted for the 25 parts of the non-film forming polystyrene.

EXAMPLE 8

A transfer sheet was prepared in the same manner as set forth in Example 1 except that the coating composition was formed by thoroughly mixing together 55.5 parts of a polystyrene emulsion containing 50% solids and manufactured under the tradename Polyco 2306; 6.2 parts of a self-cross-linking vinyl acrylic copolymer emulsion manu-

factured by the National Starch and Chemical Company under the trade name X-Link 2833; 60.0 parts of a 2% solution of phthalocyanine blue and 5 parts water.

All of the transfer sheets prepared according to the above Examples 2 to 8 were observed and tested in the same manner as set forth in Example 1. The dry transfer coatings possessed a high gloss, high scuff resistance and were substantially smudge-proof and possessed substantially the same transfer and deactivation properties as described with respect to the transfer sheet prepared in Example 1. The transfer sheet prepared according to Example 8 possessed particularly good aging properties.

In the above examples it was noted that there was a proper amount of adhesive present in the coating compositions, i.e., there was a sufficient amount of adhesive to hold the resin particles together and adhere the coating to the paper, but the amount of adhesive was insufficient to hold the coatings to the paper when subjected to the pressure such as from a stylus or a typewriter key under normal operating conditions.

I claim:

1. The method of preparing a transfer sheet which comprises coating a suitable flexible foundation with a coating composition comprising an admixture of a non-waxy adhesive and an aqueous emulsion of organic polymer spheres which have a glass transition temperature of more than about 140° F. and a particle size of between about 0.05 to 0.3 microns, heating the coating to a temperature which will not substantially deform the organic polymer spheres, but which will substantially deform the adhesive so that a transfer coating is obtained which is equivalent to a coat weight of about 1 to 7 lbs. of coating per ream of paper, the organic polymer spheres comprising at least about 70% by weight of the coating, and the adhesive comprising between about 1–27% by weight of the coating.

2. The method of claim **1** in which the flexible foundation is paper and the coating is of a coat weight of about 2–4 lbs. per ream of paper.

3. The method of claim **1** in which the admixture contains a pigment.

4. A transfer sheet comprising a flexible foundation and a coating adhered thereto, said coating comprising at least 70% by weight of discrete, non-coalesced, organic polymer spheres, and from 1–27% by weight of a non-waxy adhesive which is in an amount sufficient to hold the polymer spheres together and to the flexible foundation, but insufficient to hold the polymer spheres to the flexible foundation when subjected to local pressure, said organic polymer spheres having a glass transition temperature of at least about 140° F. and having a particle size of about 0.05 to 0.3 microns, said coating being in an amount equivalent to the amount of coating on a coated paper having from 1 to 7 lbs. of coating per ream of paper.

5. The transfer sheet of claim **4** in which the flexible foundation is paper and the coating is of a coat weight of about 2 to 4 lbs. per ream of paper.

6. The transfer sheet of claim **4** in which the coating contains coloring matter.

7. The transfer sheet of claim **4** in which the adhesive is a carboxylated organic, water-soluble polymer containing periodic carboxylic acid groups along the polymer chain.

8. The transfer sheet of claim **4** in which the adhesive is a self-cross-linking adhesive.

9. The method of transferring images from a transfer sheet to a receptor sheet which comprises coating a base sheet with a composition comprising an admixture of a non-waxy adhesive and an aqueous emulsion of organic polymer spheres which have a glass transition temperature of more than about 140° F. and a particle size of between about 0.05 to 0.3 microns, heating the coating to a temperature which will not substantially deform the organic polymer spheres, but which will substantially deform the adhesive so that a transfer coating is obtained which is equivalent to a coat weight of about 1 to 7 lbs. of coating per ream of paper, the organic polymer spheres comprising at least about 70% by weight of the coating, and the adhesive comprising between about 1–27% by weight of the coating, placing the transfer coating of the transfer sheet in contact with a receptor sheet, applying local pressure to the assembly to effect the transfer of an image from the coating to the receptor sheet in areas coextensive with the applied pressure, and subsequently heating the transfer sheet to a temperature above the softening point of the polymer spheres for a sufficient length of time to destroy the transfer properties of the transfer coating adhered thereto.

10. The method of transferring images from a transfer sheet to a receptor sheet which comprises coating a base sheet with a composition comprising an admixture of a non-waxy adhesive and an aqueous emulsion of organic polymer spheres which have a glass transition temperature of more than about 140° F. and a particle size of between about 0.05 to 0.3 microns, heating the coating to a temperature which will not substantially deform the organic polymer spheres, but which will substantially deform the adhesive so that a transfer coating is obtained which is equivalent to a coat weight of about 1 to 7 lbs. of coating per ream of paper, the organic polymer spheres comprising at least about 70% by weight of the coating, and the adhesive comprising between about 1–27% by weight of the coating, placing the transfer coating of the transfer sheet in contact with a receptor sheet, applying local pressure to the assembly to effect the transfer of an image from the coating to the receptor sheet in areas coextensive with the applied pressure, and subsequently heating the receptor sheet having the image adhered thereto to a temperature above the softening point of the polymer spheres for a sufficient length of time to render the transferred image non-transferable.

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3,036,924	5/1962	Newman	106—14.5

MURRAY KATZ, *Primary Examiner.*