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METHOD OF FORMING DECORATIVE BODIES

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This invention relates to a method of forming raised decorative bodies on a surface to be decorated, and more particularly to a method of mass producing greeting cards having a novel raised decorative body of predetermined shape and configuration affixed thereto.

The present application is a continuation-in-part of my copending application, Serial No. 592,272, filed June 19, 1956, now abandoned, entitled Method of Forming Decorative Bodies, which application is in turn a continuation-in-part of my then copending application, Serial No. 497,721, filed March 29, 1955, now abandoned, entitled Decorative Articles of Manufacture, which application was in turn a continuation-in-part of my then copending application, Serial No. 457,131, filed September 20, 1954, now abandoned, entitled Composition for Simulating Precious Stones and Method of Applying the Same.

Conventionally, greeting cards are formed with a decorative printing of predetermined shape and design. Numerous printing techniques well known in the art have been used to form designs of exotic color and form. In more expensive greeting cards, card decorating techniques have been used by which materials other than ink such as, for example, felt fibers, may be utilized to provide more unusual types of greeting cards.

Greeting card manufacturers also applied synthetic jewels or sequin-like materials to the preprinted surfaces of greeting cards to satisfy the more esoteric demands of those customers who desire something different in the greeting card line.

The conventional techniques of applying flocking material and synthetic jewels are time consuming, laborious and expensive. Conventionally, flocking is applied by first applying to the greeting card an adhesive in a predetermined pattern, spraying or otherwise applying a flocked material to the greeting card and then allowing the adhesive to dry and thereby hold the flocked material to it. Synthetic gems are conventionally applied by hand and the gems are adhered to the surface of the greeting card by means of an adhesive.

Prior to the present invention, many manufacturers of greeting cards attempted to devise a method by means of which raised decorative bodies which simulate gems or jewels could be applied to greeting cards on a mass production basis. It was their desire to manufacture a card which had the appearance of luxury at a relatively moderate price. The method conventionally used required the use of a guide such as a stencil plate or the like through which a hardenable mass was introduced to the surface of an object to be decorated. This mass was then hardened as by heat to form a decorative body. In order to maintain the mass in a predetermined shape the guide was left in position on the surface until the mass had sufficiently hardened to prevent the running thereof and then the stencil plate was removed. The necessity of retaining the stencil plate or the like on the surface to be decorated until the mass had at least partially hardened increased the cost of production and limited the output per stencil during a given time period. Thus, conventional techniques by which gem-like objects were applied to a surface to be decorated were expensive, time consuming and complicated.

It is, therefore, an object of this invention to provide a method by means of which raised decorative bodies simulating gems or jewels may be applied readily and eco-

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nomically on a mass production basis to surfaces to be decorated.

It is another object of this invention to provide a method for forming raised decorative bodies from a liquid composition in which guide means are required for only a short period of time.

It is still another object of this invention to provide a method for forming raised decorative bodies on surfaces to be decorated which permits the decoration of a great number of surfaces during a given time period.

It is a further object of this invention to provide a method of forming decorative bodies which eliminates the need for an adhesive and expensive hand processing.

Still another object of this invention is to provide a method by means of which decorative bodies of predetermined size and shape may be readily and conveniently applied to greeting cards.

A more particular object of this invention is to provide a method by means of which clear plastic bodies having an entrapped substantially uniform layer of reflective material simulating thereby a jewel-like appearance may be readily and conveniently secured to the surface of a greeting card in any predetermined pattern or position.

Other objects and advantages of the present invention will, it is submitted, be apparent to those skilled in the art from the following detailed description of a preferred embodiment thereof.

Generally, the method which is the subject of this invention contemplates the formation of a substantially opaque resinous composition containing a thixotropic agent, the application of the resinous material to a surface to be decorated in predetermined shape and form, and the curing of the material whereby a clear, transparent resinous material having a gem-like quality is formed which firmly adheres to the surface.

More particularly, the method contemplates the use of particular polyester monomer systems containing a thixotropic agent, a thermal responsive catalyst and certain stabilizing agents; the application of the modified monomeric material to a greeting card, the said material being formed and positioned by a stencil of predetermined shape and thickness; and the conversion of the opaque thixotropic monomer to a solid polymer by means of an accurately controlled time-temperature cure cycle.

According to the present invention the guide means such as a stencil plate is made from a material to which the composition of the decorative bodies will not adhere when in liquid form. It has been found that this condition is best met by using Teflon as the material of the stencil plate. Holes in the predetermined shape desired for the raised decorative bodies in accordance with the overall design of the Christmas cards or the like are then cut into the stencil plate.

The entire stencil plate is preferably of standard length and width, so as to permit insertion into a frame which is hingedly attached to a working surface. The paper sheet or the like which is to be decorated is then placed on the working surface, the frame with the Teflon plate therein is lowered so that the stencil plate comes in contact with the upper surface of the sheet of paper or the like.

Suitable guides for the sheet of paper are provided on the working surface so as to obtain proper registration between the Teflon plate or guide means and the underlying sheet of paper. The liquid composition of which the raised decorative bodies are to be formed is then introduced from the upper free face of the stencil plate so as to fill the holes which were previously cut into the stencil plate.

Care has to be taken that these holes are completely filled with the liquid composition. This can be done in any suitable manner known in the art such as by running a squeegee over the upper free surface of the stencil plate.

As soon as the flowable thixotropic composition filled into the holes in the stencil plate is sufficiently hardened to retain its shape, which happens practically immediately, the frame with the stencil plate is again lifted. While the frame and stencil plate arrangement has been described as hingedly connected with the working surface, it is of course within the scope of the present invention to provide any other suitable means for raising and lowering the stencil plate or the like in such a way as to obtain proper registration with the underlying surface to be decorated.

After removal of the guide means or stencil plate, the paper card or other object with the raised decorative body adhering thereto is then removed from the working surface. The raised objects or bodies are then further hardened or dried as will be described below.

In order to obtain substantially immediate shape-retaining semihardness of the composition which is applied to the surface to be decorated through the holes in the stencil plate or the like, the method of the present invention employs a hardenable thixotropic composition. Because of its thixotropic nature this composition will become rigid and shape-retaining immediately upon being motionless, that is immediately after being filled into the holes of the stencil plate, and because it contains a hardenable substance it can be subsequently hardened so as to become permanently shape-retaining.

Thus, according to the present invention, initial shape-retention is achieved by the thixotropic nature of the composition and permanent shape-retention by the subsequent hardening process. During the hardenable process, the thixotropic properties of the composition are lost. However, the thixotropic rigidity and shape-retention will be retained for a sufficient length of time until the progressing permanent hardening process has sufficiently advanced to maintain the shape of the raised decorative body.

There are generally two steps in forming the finished plastic material; the first is the formation of the polymerizable unsaturated resinous composition. Second, the resulting polyester is then polymerized with a compound containing a polymerizable $\text{CH}_2=\text{C}<$ group.

The polyesters may be defined as polymerizable ethylenically unsaturated polyhydric alcohol-polycarboxylic acid polyesters. These polyesters are generally prepared by the reaction of unsaturated polycarboxylic acids and polyhydric alcohols under an inert atmosphere and at elevated temperatures. These polyesters are well known in the art, and have been described in numerous plastic encyclopedias and patents. The reaction is generally carried out until a desired acid number or a predetermined extent of esterification is realized. The resulting unsaturated polyesters obtained are polymerizable.

In the preparation of the polymerizable polyester resinous compositions ethylenically unsaturated polycarboxylic acids such as maleic, fumaric, aconitic, itaconic, and the like may be used. In addition, acids such as phthalic, malonic, succinic, glutaric, sebacic and the like, may be used. The anhydrides of these acids may also be used, e.g., maleic anhydride, phthalic anhydride, and as used herein polycarboxylic acids include the polycarboxylic anhydrides when available. Also, mixtures of the acids and anhydrides may be used in the preparation of the polyester resin.

Polyhydric alcohols which may be used in the preparation of the polyesters include such compounds as ethylene glycol; diethylene glycol; propylene glycol; dipropylene glycol; butanediol-1,4; butanediol-1,3; butanediol-1,2; pentanediol-1,4; pentanediol-1,5; hexanediol-1,6; glycerol, 1,1'-isopropylidenebis(p-phenylenoxy)di-2-propanol, pentaerythritol, dipentaerythritol.

In preparing the polyester, mixtures of one or more of the carboxylic acids or the anhydrides thereof listed above may be mixed with one or more of the polyhydric alcohols and reacted in an inert atmosphere and at ele-

vated temperatures to provide the polyesters. These techniques and compositions are well known in the art.

The polymerizable unsaturated polyesters thus formed are then admixed with a monomeric compound containing the polymerizable $\text{CH}_2=\text{C}<$ group to give a composition that may be cured to a stable thermoset condition. Monomeric materials which may be used include such compounds as styrene, side-chain alkyl and halo-substituted styrenes such as alpha-methylstyrene, alpha-chlorostyrene, alpha-ethylstyrene and the like or alkyl and halo ring-substituted styrene such as ortho-, meta- and para-alkyl styrenes such as o-methylstyrene, p-ethylstyrene, m-propylstyrene, 2,4-dimethylstyrene, 2,5-diethylstyrene, bromostyrene, chlorostyrene, dichlorostyrene and the like. Still further, one can make use of the allyl compounds such as diallyl phthalate, tetrachlorodiallyl phthalate, allyl alcohol, methallyl alcohol, allyl acetate, allyl methacrylate, diallyl carbonate, allyl lactate, allyl alpha-hydroxyisobutyrate, allyl trichlorosilane, allyl acrylate, diallyl malonate, diallyl oxalate, diallyl gluconate, diallyl methylgluconate, diallyl adipate, diallyl sebacate, diallyl citraconate, the diallyl ester of muconic acid, diallyl itaconate, diallyl chlorophthalate, diallyl dichlorosilane, the diallyl ester of endomethylene tetrahydrophthalic anhydride, triallyl tricarballylate, triallyl aconitate, triallyl cyanurate, triallyl citrate, triallyl phosphate, trimethallyl phosphate, tetraallyl silane, tetraallyl silicate, hexallyl disiloxane and the like.

After admixing the polyester and monomer, the latter of which may include one or more of the compounds above set forth, a catalytic agent is used to effect the reaction between the polyester and monomer. As is well known in the art, a catalyst of the peroxide class may be utilized. Examples of catalysts that may be used are such as benzoyl peroxide, succinyl peroxide, acetyl peroxide, methyl ethyl ketone peroxide, cumene hydroperoxide, tertiarybutyl hydroperoxide, cyclohexanone peroxide, perbenzoic acid, peracetic acid, anisoyl peroxide, toluyl peroxide, p-bromobenzoyl peroxide, tertiarybutyl perbenzoate, p-methane hydroperoxide, pinane hydroperoxide, diisopropylbenzene hydroperoxide, tertiarybutyl peracetate, 1-cyclohexanol-1-hydroperoxide, furoyl peroxide and chloracetyl peroxide or any organic ozonide, such as diisopropylene ozonide, diisobutylene ozonide or a mixture of such substances. Other free radical type polymerization catalysts that may also be used are such as α,α -azodiisobutyronitrile, the salts of inorganic peracids, e.g., ammonium persulfate, sodium persulfate and the like.

It will be apparent from the above description of the polyester, monomer and catalyst that by the use of the term "polymerizable unsaturated polyester resin" I mean that resinous material which is the mixture of a polymerizable unsaturated polyester, a monomeric compound containing a polymerizable $\text{CH}_2=\text{C}<$ group and a suitable peroxide catalyst. The polymerizable unsaturated polyester is the reaction product of a selected unsaturated polycarboxylic acid and a polyhydric alcohol.

The particular proportions of ingredients to be used to form the final resin will be dictated by the particular compounds selected, and is well within the skill of the art.

The uncatalyzed polymerizable unsaturated polyester resin thus formed is relatively fluid and would, of its own accord, tend to run, or flow. In order to hold the material in a predetermined shape during the curing operation, various thixotropic materials well known in the art may be added to the unpolymerized material to increase the apparent viscosity. Typical thixotropic agents are hydrogenated castor oil, aluminum palmitate, aluminum stearate, bentonite and calcium stearate. These thixotropic agents convert the relatively fluid polymerizable resin into a substantially stable gel which will at least for the curing times involved herein, hold the gel in a predetermined shape and form.

The use of a thixotropic agent adversely affects the clarity and quality of the polymerizable unsaturated polyester resin. The mixture of the resin and the thixotropic agent is cloudy and at least semiopaque. I have discovered, and as a part of this invention, that if this mixture is cured under certain time-temperature conditions, the thixotropic agent loses its efficacy and permits the formation of a clear, transparent body. Accordingly, I have discovered a method by which a thixotropic agent may be utilized to hold a polymerizable resin in a predetermined shape during curing and still obtain a clear, transparent cured decorative body which firmly adheres to the surface to be decorated.

In the catalytic curing of the resulting thixotropic mixture, air tends to inhibit the surface cure resulting in a residual tackiness. This surface tackiness may be prevented or reduced by the addition of certain compounds well known in the art. Examples thereof are cobalt naphthene, paraffin wax and silicon fluid. These additive compounds tend to improve the quality of the ultimate product but are not necessary if air inhibition of the cure can be otherwise prevented.

To the thixotropic composition used in the method of the present invention, coloring materials which are soluble in the polyester resin, as well as solid materials such as metal flakes, tinsel, powder of mother of pearl and the like may be added. The solid materials added to the polyester resin are in the unpolymerized resin substantially uniformly distributed throughout the mixture. The false viscosity given the mixture by the thixotropic agent holds these solid materials in suspension.

I have discovered that if the resin is cured rapidly as, for example, by immediately subjecting the unpolymerized decorative bodies to a temperature in excess of 200° F., the solid materials are permanently entrapped and uniformly distributed throughout the decorative body. This uniform distribution does not result in a decorative body having a pleasing appearance. However, and as a further part of the present invention, I have discovered that if the unpolymerized resin is cured under predetermined time-temperature conditions, the thixotropic agent begins to lose its efficacy at a temperature below the point where catalytic curing proceeds rapidly. As the thixotropic agent loses its efficacy, apparent or false viscosity of the uncured resin decreases allowing the solid materials to settle to a substantially uniform layer. By substantially increasing the temperature, rapid curing is effected whereby a decorative body having a realistic gem-like quality is formed.

After the raised ornamental body has been formed on the surface to be decorated, the same is exposed to such hardening conditions, for instance elevated temperatures, as will—in connection with the accelerators, catalysts and the like embodied in the composition—insure proper hardening of the raised decorative body. Preferably a short period such as between 2 and 4 minutes of prehardening at a temperature of between 100° F.—120° F. precedes the final hardening step which may be executed at a temperature of between 150° F.—220° F. and which, depending on the heating method employed, may last for between about 2 and 6 minutes. When determining the actual hardening conditions, one has also to take into consideration the size and shape of the raised ornamental objects, e.g., the relationship between volume and exposed surface of these objects and thus the speed of heat penetration therethrough.

The hardening conditions, or curing cycle is, as indicated above, performed in two steps. In the first step the card carrying the unpolymerized resinous material is subjected to a temperature between 100° F. and 120° F. for a period long enough to destroy the efficacy of the thixotropic agent and permit the settling of the added solid materials, if any, in the unpolymerized body. The temperature is then, in the second step of curing, raised rapidly to a temperature above 150° F. to effec-

uate a final cure. It will be understood by those skilled in the art that the precise curing temperature to be used both in the first and second stages of cure will be dictated by (1) the temperature required to cause the thixotropic agent to lose its efficacy, and (2) the temperature at which the catalytic cure of the polymerizable resin may be achieved most expeditiously.

Preferably, after completion of the hardening process, the cards or other decorated objects are permitted to cool for between 5 and 10 minutes prior to being packed or otherwise handled.

The heat-curing process may be executed on trays which are placed in an oven in which air of desired elevated temperature circulates, or, preferably, the decorated objects such as Christmas cards are placed on a conveyor belt which travels through the heating zone at such speed that the raised decorative bodies are exposed to the desired temperature for the desired length of time.

It is also within the scope of the present invention to employ a curing process which is actuated by light instead of heat. In this case the cards or the like with the raised decorative bodies thereon are preferably placed on a conveyor belt and exposed thereon for the desired length of time to ultra-violet rays.

As the thixotropic agent loses its efficacy during the final stages of cure, the outer surfaces of the portions of unpolymerized material applied to the card become relatively fluid and tend to flow. The amount of flow I have discovered is very small but is sufficient to allow a thin layer of unpolymerized resin to become closely incorporated with or absorbed by the fibers of the paper card. When curing is completed, the resin material which has become absorbed in the paper joins the applied decorative body and the card. The body thus virtually becomes a part of the card and cannot be removed without physically tearing it off the surface of the card. It will be apparent that this is superior to the methods previously used in applying decorative bodies to cards where a surface adhesive was used which did not permit the decorative body to be physically incorporated within at least a portion of the card.

In applying the decorative bodies to the cards the following steps are followed:

(1) The polyester resin is prepared.
(2) The thixotropic agent and other desirable additives are thoroughly mixed with the polyester resin.

(3) A stencil having predetermined shape and form is applied to the card to be decorated.

(4) The polyester resin is placed on the stencil and the excess removed.

(5) The stencil is removed, leaving uncured polyester resin in a plurality of predetermined positions and shapes on the card, the thixotropic agent preventing "running" of the resin.

(6) The card is introduced into a heating oven where it is first heated to a temperature below its generally accepted cure point, and where the thixotropic agent loses its efficacy, and the added solid material falls to form a uniform layer. The heat is then increased steadily for a predetermined time until thorough cure is achieved. The resulting card has decorative bodies firmly fixed thereto.

The following are specific examples of the method of forming decorative bodies. In these examples the materials are given in parts by weight.

EXAMPLE NO. I

76 parts propylene glycol, 49 parts maleic anhydride and 74 parts phthalic anhydride were introduced into a steam-jacketed reaction vessel. The vessel was covered with a pressure cap having a gas inlet and a gas outlet nozzle. Nitrogen gas was introduced into the gas inlet nozzle until the area above the added mixture contained only nitrogen at which time the inlet and outlet nozzles

were closed to maintain an inert atmosphere in the vessel. The reactants were stirred constantly and the vessel heated until approximately 30 parts by weight of water had been evolved from the reactants. The temperature of the reactants was maintained below 200° F. Following the evolution of the water, the resulting polyester was cooled to 100° F. and 0.005 part of the reaction catalyst methyl ethyl ketone peroxide was added to the polyester and thoroughly admixed therewith. To the resulting mixture was next added 80 parts styrene which in turn was thoroughly mixed throughout the polyester. The resulting mixture of the polyester, catalyst and styrene monomer was a polymerizable unsaturated polyester resin having a viscosity of approximately 1000 centipoises. The resin was relatively fluid and accordingly would not retain its shape when placed on a surface to be decorated.

To the resin was added 0.5 part hydrogenated castor oil, a thixotropic agent which increased the apparent viscosity of the resin and substantially eliminated its tendency to flow. The resulting thixotropic mixture, which was cloudy or milky in appearance, could be stored or used for periods up to three to four hours at room temperature without hardening, the speed of the reaction being very slow at temperatures below approximately 150° F.

A stencil having openings of predetermined size and shape was then placed on a surface to be decorated, more specifically, a Christmas card. The surface of the stencil was covered with the polymerizable resin to completely fill all openings in the stencil. The excess resin was removed with a squeegee and the material allowed to set for a few moments. The stencil was then removed and the card was placed in an oven having a temperature of 120° F. for 3 minutes. During the heating at this temperature the decorative bodies on the cards maintained the shape imparted to them by the stencil and as the thixotropic agent began to lose its efficacy, the individual resin bodies cleared. At the end of 3 minutes the composition became clear and entrapped air bubbles present in the individual bodies because of the previous agitation were eliminated from the body. After the preliminary heating at 120° F. the temperature was raised rapidly to 200° F. for 4 minutes at which temperature the catalytic curing of the resin occurred very rapidly. The resulting decorative bodies were hard and clear, i.e., transparent and firmly adhered to the surface of the card. In removing one of the bodies from the card the card was torn.

EXAMPLE NO. II

The polymerizable polyester resin was prepared as in Example No. I except that 0.01 part of a 50 percent benzoyl peroxide was used as a catalyst. To the thixotropic mixture was added a red soluble dye to tint the cloudy mixture a deep red. To this mixture was added and uniformly distributed throughout a small amount of finely divided tinsel material. The decorative bodies were applied to the card as in Example No. I and the following curing cycle was used.

The card was first subjected to a temperature of 120° F. for 5 minutes and 6 seconds. The temperature was then rapidly increased to 145° F. and the card kept at that heat for 3 minutes and 49 seconds. The temperature was again increased to 200° F. for 2 minutes and 33 seconds, and finally, the temperature was increased to 250° F. for 2 minutes and 33 seconds. At 120° F. the apparent viscosity of the mixture obviously decreased as the tinsel, which was uniformly distributed throughout the applied body, began to settle and the body to become transparent. At the end of the 145° F. step the body was clear and a brilliant red, all of the added tinsel having settled to a uniform layer adjacent the card. No change in physical appearance was observed after the 200° F. step and after final curing at 250° F. a decorative body having a realistic jewel-like appearance was formed.

Other decorative bodies may be formed in the manner

set forth in Examples I and II utilizing the polymerizable unsaturated polyester resins previously described.

It will be apparent that the precise curing times to be used will be dependent upon the resin selected, the catalytic agent used, the dimensions of the particle applied and the relative humidity.

In general, the decorative body is heated at a temperature and for a time sufficient to permit the thixotropic agent to lose its efficacy and the body to clear. The temperature of the body is then promptly raised to affect rapid catalytic curing of the resin.

The following are general examples which tend to illustrate the quantities of materials involved.

EXAMPLE No. III

Heat Hardenable Composition

	Minimum quantity, g.	Maximum quantity, g.	Preferred quantity, g.
Basic thixotropic composition including polyester resin.....	1,000	1,000	1,000
Solution of 6% cobalt-naphthenate in petroleum spirits.....	2.5	10.0	7.5
Paraffin wax (colorless, to be added in hot, liquid condition).....	0.1	1.0	1.0
50% benzoyl peroxide in tricresyl phosphate.....	10.0	30	15
Coloring materials (dyes) compatible with and soluble in polyester resins, preferably of the "Orasole" (Ciba) type.....	(1)	(1)	20
Metal flakes, tinsel or the like.....	(1)	(1)	250

¹ As desired.

The thus-obtained milky heavy liquid material can be used for 3 to 4 hours without hardening. It should not be stored for any longer periods of time. The decorative raised bodies formed of the above composition are to be heated for between 2 and 4 minutes at about 100° F.—120° F. Thereby the composition becomes clear and transparent, air bubbles disappear, the tinsel or metal flakes sink to the bottom. Subsequently, complete hardening of the composition is achieved by heating for about 4 minutes to between 200° F. and 220° F.

Manganese or iron base accelerators may also be used, however best results were obtained with cobalt-naphthenate. Other organic waxes which are compatible with polyester resins may be used instead of paraffin wax, as well as other heat catalysts in place of benzoyl peroxide.

EXAMPLE NO. IV

Heat Hardenable Composition

	Minimum quantity, g.	Maximum quantity, g.
Basic thixotropic composition including polyester resin.....	1,000	1,000
Solution of 6% cobalt-naphthenate in petroleum spirits.....	2.5	15
Paraffin wax (colorless, to be added in hot, liquid condition).....	0.1	1.0
Methyl-ethyl-ketone peroxide.....	5	20
Silicon resin or silicon fluid (such as is contained in 20% concentration in SF 69 of General Electric Co.).....	1	10
Dye, tinsel or the like.....	(1)	(1)

¹ As required.

EXAMPLE NO. V

Ultraviolet Light Hardenable Composition

	Minimum quantity, g.	Maximum quantity, g.
Basic thixotropic composition including polyester resin.....	1,000	1,000
Solution of 6% cobalt-naphthenate in petroleum spirits.....	2.5	15
Paraffin wax (colorless, to be added in hot, liquid condition).....	0.1	1.0
Benzoin.....	10	50
Dye, tinsel or the like.....	(1)	(1)

¹ As required.

The decorative raised bodies formed of the above composition are to be heated for between 2 and 3 minutes to about 100° F.—120° F. until the composition becomes clear and transparent. Thereafter the objects such as Christmas cards, decorated with raised bodies of the above composition are exposed, preferably on a moving belt to ultraviolet rays emanating from ultraviolet bulbs. It is thus possible to use the same curing equipment for heat hardenable and ultraviolet light hardenable compositions by simply replacing heating bulbs with ultraviolet bulbs. Ultraviolet light hardenable compositions are preferred in cases where the paper, fabric or the like on which the raised bodies are formed should not be exposed to temperature above between 100° F. and 150° F.

In the above preferred embodiment of my invention I have described in detail the polymerizable unsaturated polyester resins which are suitable for use in forming the decorative bodies. It will, however, be apparent to those skilled in the art that any other catalytically cured thermosetting resin may be substituted for the polyester resin there described.

In order to achieve the desired jewel-like appearance, it will be obvious that only clear resins should be used. Examples of classes of resins which may be substituted for the polyester resin are epoxy, amino (urea and melamine) phenols or phenolics, alkyd and silicone resins. Each of these resins is capable of cross-linking in the presence of a catalyst to form a hard, clear gem-like body.

Finally, while unpolymerized resinous compositions may be formed in a wide variety of viscosities, I prefer to use compositions having viscosities in the range of from 500 to 1500 centipoises.

By utilizing a thixotropic agent to increase the apparent viscosity of the resinous composition and by subjecting the resulting thixotropic composition to the previously described time-temperature cure cycle, the decorative bodies may be formed.

It will thus be apparent that in the broadest sense the method which is the subject of this invention contemplates the formation of a thixotropic composition, the destruction of the thixotropic agent by heating at a temperature below the conventional curing temperature of the resin and finally, raising the temperature of the resin to effect final cure.

From the above it will be apparent that by the use of the process thus described, a card having a plurality of separate individually shaped decorative bodies having a wide variety of colors and appearances may be formed and adhered to the surface of a greeting card or the like on a mass production basis without the necessity of individually treating each body. Hand processing is thus, to a large extent, eliminated. In addition, no adhesives are necessary, the body becoming an integral part of the card itself when cured. It will also be apparent from the above description that a wide variety of materials may be used in forming the resin and that the precise curing cycle will be dependent upon the selection of the materials.

Having fully described my invention, it is to be understood that I do not wish to be limited to the precise details of the examples set forth but my invention is of the full scope of the appended claims.

I claim:

1. The process of decorating a surface with simulated gem-like solid translucent elements attached to the surface, and in which are embedded solid glittering decorative particles, which comprises: registering said surface with a stencil, having openings corresponding to the design to be produced on said surface; covering the surfaces exposed by the stencil with a mixture of gel-like thixotropic material in which is included solid glittering particles, and of sufficient depth to provide raised bodies on the surfaces; said thixotropic material in its normal state being milky and serving to adhere to the surfaces without requiring any other adhesive; separating the stencil from the surface; heating the thixotropic material to a temper-

ature of from 100° F. to 120° F. for a period sufficient for the material to clear; and then continuing the heating at an elevated temperature to complete the hardening of the thixotropic material.

2. In a method of forming raised decorative bodies on a surface to be decorated, the steps comprising: forming a clear liquid polymerizable unsaturated polyester resin; mixing the liquid resin with a thixotropic agent to convert the resin into a substantially stable cloudy gel; applying the stable gel to the surface to be decorated in predetermined shape and form; heating the gel at a temperature and for a time sufficient to permit the thixotropic agent to lose its efficacy and the gel to clear; increasing the temperature of the resulting clear gel to effect catalytic curing of the resin whereby a raised decorative body is formed.

3. In a method of forming raised decorative bodies on a surface to be decorated, the steps comprising: reacting an unsaturated polycarboxylic acid with a polyhydric alcohol to form a polymerizable unsaturated polyester; admixing the polyester with a monomeric compound containing a polymerizable $\text{CH}_2=\text{C}<$ group and a peroxide catalyst to form a clear liquid polymerizable unsaturated polyester resin; mixing the liquid resin with a thixotropic agent to convert the resin into a substantially stable cloudy gel; applying the stable gel to the surface to be decorated in predetermined shape and form; heating the gel at a temperature and for a time sufficient to permit the thixotropic agent to lose its efficacy and the gel to clear; increasing the temperature of the resulting clear gel to effect catalytic curing of the resin whereby a raised decorative body is formed.

4. In a method of forming raised decorative bodies on a surface to be decorated, the steps comprising: reacting an unsaturated polycarboxylic acid with a polyhydric alcohol to form a polymerizable unsaturated polyester; admixing the polyester with a monomeric compound containing a polymerizable $\text{CH}_2=\text{C}<$ group and a peroxide catalyst to form a clear liquid polymerizable unsaturated polyester resin; mixing the liquid resin with a thixotropic agent to convert the resin into a substantially stable cloudy gel; introducing into the substantially stable cloudy gel a soluble colored dye and a plurality of discrete particles of a solid decorative material to form a homogeneous dyed gel having the plurality of discrete particles uniformly distributed therethrough; applying the stable gel to the surface to be decorated in predetermined shape and form; heating the gel at a temperature and for a time sufficient to permit the thixotropic agent to lose its efficacy, the gel to clear and the solid material to settle to form a uniform layer adjacent the surface to be decorated; increasing the temperature of the resulting gel to effect catalytic curing of the resin whereby a raised decorative body is formed.

5. In a method of forming raised decorative bodies on a surface to be decorated, the steps comprising: reacting an unsaturated polycarboxylic acid with a polyhydric alcohol to form a polymerizable unsaturated polyester; admixing the polyester with a monomeric compound containing a polymerizable $\text{CH}_2=\text{C}<$ group and a peroxide catalyst to form a clear liquid polymerizable unsaturated polyester resin; mixing the liquid resin with a thixotropic agent to convert the resin into a substantially stable cloudy gel; introducing into the substantially stable cloudy gel a soluble colored dye and a plurality of discrete particles of a solid decorative material to form a homogeneous dyed gel having the plurality of discrete particles uniformly distributed therethrough; applying the stable gel to the surface to be decorated in predetermined shape and form; subjecting the gel to a temperature in the range of from 110° F. to 120° F. for a period of from 2 to 6 minutes to permit the thixotropic agent to lose its efficacy, the gel to clear and the solid material to settle to a uniform layer adjacent the surface to be decorated; increasing the temperature to a maximum of

250° F. for a period of not less than 3 minutes to effect catalytic curing of the resin whereby a raised decorative body is formed.

6. In a method of forming raised decorative bodies on a surface to be decorated, the steps comprising: reacting 76 parts by weight propylene glycol, 49 parts by weight maleic anhydride and 74 parts by weight phthalic anhydride in an inert atmosphere and with constant agitation at a temperature above 100° F. but below 200° F. until approximately 30 parts by weight water has been evolved to form a polymerizable unsaturated polyester; cooling the resulting polyester to 100° F.; adding 80 parts by weight styrene and 0.005 part by weight methyl-ethylketone peroxide to the resulting polyester to form a polymerizable unsaturated polyester resin; increasing the apparent viscosity of the resin by admixing therewith 0.5 part by weight hydrogenated castor oil to form a substantially stable cloudy thixotropic mixture; applying the stable mixture to the surface to be decorated in predetermined shape and form; subjecting the resulting mixture to a temperature of 120° F. for 5 minutes and 6 seconds, increasing the temperature to 145° F. for 3 minutes and 49 seconds, increasing the temperature to 200° F. for 2 minutes 33 seconds and again increasing the temperature to 250° F. for 2 minutes and 33 seconds whereby a plurality of raised decorative bodies is formed.

7. In a method of forming raised decorative bodies on a surface to be decorated, the steps comprising: forming a clear liquid polymerizable resin; mixing the liquid resin with a thixotropic agent to convert the resin into a substantially stable cloudy gel; applying the stable gel to the surface to be decorated in predetermined shape and form; heating the gel at a temperature and for a time sufficient to permit the thixotropic agent to lose its efficacy and the gel to clear; increasing the temperature of the resulting clear gel to effect catalytic curing of the resin whereby a raised decorative body is formed.

8. In a method of forming raised decorative bodies on a surface to be decorated, the steps comprising: forming a clear liquid polymerizable resin having a viscosity in the range of from 500 to 1500 centipoises; mixing the liquid resin with a thixotropic agent to convert the resin into a substantially stable cloudy gel; applying the stable gel to the surface to be decorated in predetermined shape and form; heating the gel at a temperature and for a time sufficient to permit the thixotropic agent to lose its efficacy and the gel to clear; increasing the temperature of the resulting clear gel to effect catalytic curing of the resin whereby a raised decorative body is formed.

9. In a method of forming raised decorative bodies on a surface, the steps comprising: applying a stable gel to said surface in predetermined shape and form, said stable gel consisting essentially of a mixture of a clear, liquid polymerizable resin and a thixotropic agent; heating the gel at a temperature and for a time sufficient to permit the thixotropic agent to lose its efficacy and the gel to clear; increasing the temperature of the resulting clear gel to effect catalytic curing of the resin whereby a raised decorative body is formed.

10. In a method of forming raised decorative bodies on a surface, the steps comprising: adding finely divided tinsel material to a stable gel to form a homogeneous mixture thereof, said gel consisting essentially of a mixture of a clear, liquid polymerizable resin and a thixotropic agent; applying said mixture to said surface in predetermined shape and form; heating the mixture at a temperature and for a time sufficient to permit the thixotropic agent to lose its efficacy, the gel to clear and the tinsel material to settle to a uniform layer adjacent said surface; increasing the temperature of the resulting clear gel to effect catalytic curing of the resin whereby a raised decorative body is formed.

11. In a method of forming raised decorative bodies on a surface, the steps comprising: adding finely divided tinsel material to a stable gel to form a homogeneous mixture thereof, said gel consisting essentially of a mixture of a clear, liquid polymerizable resin and a thixotropic agent; applying said mixture to said surface in predetermined shape and form; subjecting the mixture to a temperature in the range of from 110° F. to 120° F. for a period of from two to six minutes to permit the thixotropic agent to lose its efficacy, the gel to clear and the tinsel material to settle to a uniform layer adjacent said surface; increasing the temperature to a maximum of 250° F. for a period of not less than three minutes to effect catalytic curing of the resin whereby a raised decorative body is formed.

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