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K. G. SVENSSON
HEAT-SENSITIVE TRANSFER SHEET FOR PRODUCING
A THERMOGRAPHIC FACSIMILE COPY
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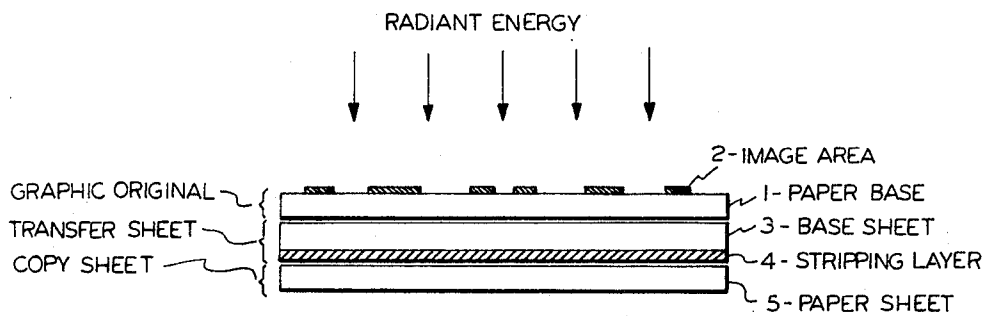


FIG. 1

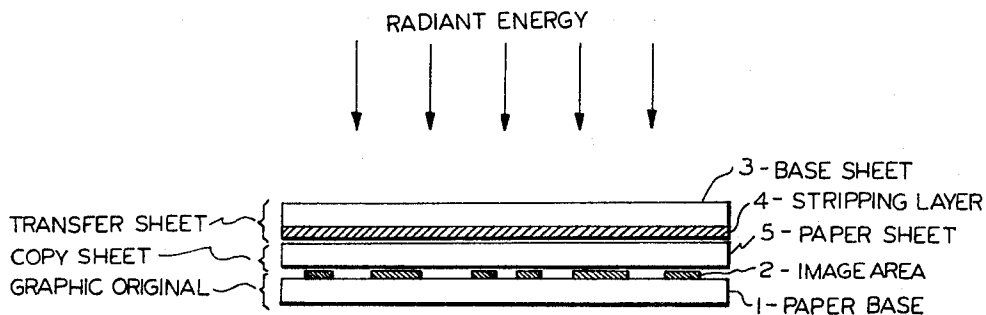


FIG. 2

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**HEAT-SENSITIVE TRANSFER SHEET FOR
PRODUCING A THERMOGRAPHIC FAC-
SIMILE COPY**Karl Gunnar Svensson, Bergslagstorget 2D,
Filipstad, SwedenContinuation-in-part of application Ser. No. 352,250,
Mar. 16, 1964. This application Apr. 18, 1967, Ser.
No. 631,830Claims priority, application Sweden, Nov. 2, 1961,
10,928/61; Mar. 16, 1962, 2,947/62; May 17, 1962,
5,540/62; Italy, Mar. 15, 1963, 32,970/63
5 Claims. (Cl. 117—36.1)**ABSTRACT OF THE DISCLOSURE**

A heat-sensitive copying sheet for producing a thermographic facsimile copy, comprising a base sheet, a heat-sensitive coating layer on one side of said base sheet, said coating layer being substantially non-pressure-sensitive at room temperature and comprising wax as a major component, a dye component in a total amount of 3 to 11% by weight of said layer, said dye component containing at least one dye retaining substantial color strength upon prolonged influence of light and containing in its molecule at least one molecularly bound atom selected from the group consisting of selenium, tellurium, iodine, chromium, manganese, molybdenum, tungsten, iron, nickel, cobalt and copper, and a solvent agent being a solvent for said wax and said dye component and being present in said layer in an amount sufficient to hold all of said dye component in dissolved state in said layer and containing at least one member of the group consisting of benzyl alcohol and compounds containing at least two hydroxyl groups and at most 8 carbon atoms in their molecule, and said coating layer when heated to a temperature within the range of 50–150° C. having a greater ability to anchor itself on a paper surface pressed against it than of being anchored to said base sheet.

This is a continuation-in-part of my continuation-in-part application Ser. No. 352,250, filed Mar. 16, 1964, of my parent application Ser. No. 233,680, filed Oct. 29, 1962, both now abandoned.

For thermographic copying by means of the usual thermographic copying machines a great many different types of copying sheets have previously been proposed. The present invention relates to a different type of heat-sensitive copying sheets for producing a thermographic facsimile copy and comprises a base sheet and a heat-sensitive and substantially non-pressure-sensitive coating layer on one surface of the base sheet, said coating layer having, when heated to a temperature within the range of 50–150° C., a higher ability to anchor itself on a paper surface pressed against it than of being anchored to said base sheet, and said coating layer comprising wax as a major component and at least one dye.

Previously, the dye has been disposed in the coating layer in an undissolved state or possibly a part of the dye has been present in a dissolved state. In the latter case two modifications are known. In one of them most of the dye is present in an undissolved state and a small part of the dye has been chemically reacted and the reaction product has been emulsified or dissolved in the wax component of the coating layer. In the other modification the dye is directly dissolved in the wax component of the coating layer.

When at least part of the dye is present in the coating layer as pigment, i.e. in an undissolved state, the copies

may show great color strength but they will show poor sharpness and strong staining of the background. This seems at least in part to be due to the fact that the pigment is heated more by the radiation heat used at the printing than a dye solution would be, whereby the background areas of the copy are more easily stained when using pigments and also whereby the dye solution is transferred to the copy surface more readily and in larger amount than pigment when heated by heat conduction from the image portions of the original which are heated by radiation heat during the copying process.

When the dye is completely and directly dissolved in the wax component sharp copies without appreciable staining of the background are obtained but the copies show poor contrast because the dyes which can be used cannot be dissolved in the wax component in a sufficiently large amount (the practical maximum being in the order of 2% by weight of the coating layer though the solubility may vary from dye to dye and from vehicle to vehicle, and due to the fact that it is necessary that the coating layer is not pressure-sensitive at normal room temperature, the major component of the coating layer must have a high content of hard wax, so that techniques used in making pressure-sensitive carbon papers cannot be resorted to. Furthermore, there are few dyes which can be directly dissolved in the wax component and the usable ones have poor light fastness.

The primary object of the present invention is to eliminate the above-mentioned shortcomings of the prior art transfer sheets of the kind mentioned and to provide a heat-sensitive and substantially non-pressure sensitive transfer sheet for producing a thermographic facsimile copy showing image areas with improved sharpness and color strength and clean background areas.

Another object of the invention is to provide a transfer sheet of the kind mentioned for producing a facsimile copy showing image areas which retain substantial color strength upon prolonged influence of light.

Another object of the invention is to gain an extended exposure latitude when a rotary thermographic copying machine is used.

A further object of the invention is to extend the range of dyes which can be used in a completely dissolved state in the wax component of the coating layer of a transfer sheet of the kind mentioned.

Thus, the invention provides a heat-sensitive copying sheet for producing a thermographic facsimile copy, comprising a base sheet, a heat-sensitive coating layer on one side of said base sheet, said coating layer being substantially non-pressure-sensitive at room temperature and comprising wax as a major component, a dye component in a total amount of 3 to 11% by weight of said layer, said dye component containing at least one dye retaining substantial color strength upon prolonged influence of light and containing in its molecule at least one molecularly bound atom selected from the group consisting of selenium, tellurium, iodine, chromium, manganese, molybdenum, tungsten, iron, nickel, cobalt and copper, and a solvent agent being a solvent for said wax and said dye component and being present in said layer in an amount sufficient to hold all of said dye component in dissolved state in said layer and containing at least one member of the group consisting of benzyl alcohol and compounds containing at least two hydroxyl groups and at most 8 carbon atoms in their molecule, and said coating layer when heated to a temperature within the range of 50–150° C. having a greater ability to anchor itself on a paper surface pressed against it than of being anchored to said base sheet.

In the accompanying drawing, FIGS. 1 and 2 diagrammatically show two ways of using a heat-sensitive copying

sheet for producing a thermographic facsimile copy of a graphic original, the set of sheets used being shown from the edge on a heavily exaggerated thickness scale.

A graphic original comprising a base 1, usually paper, insignificantly absorbing radiant energy and image surface portions 2 arranged on one face, the front, of said base by means of ink or other material which upon heating heavily absorbs radiant energy, is shown in FIG. 1 placed with its back in contact with the base 3 of a copying sheet, described in the following and having a heat-sensitive but substantially non-pressure-sensitive coating layer 4, which faces away from the graphic original and in contact with which an ordinary paper sheet or other copy receiving sheet 5 is placed. This receiving sheet is to be provided with a facsimile copy of the image surface portions 2 of the graphic original by thermography. In FIG. 1 the sheet set is placed in face-to-face relationship for producing so-called through-copying. The intense radiant energy is indicated by arrows in FIG. 1. At the exposure of the sheet set in a thermographic copying apparatus of the customary type the radiant energy is thrown toward the front of the graphic original. Radiant energy is heavily absorbed by the image surface portions 2 which are thereby heated, but between said image surface portions radiant energy can penetrate through the layers 1, 3, 4, 5 without any appreciable absorption and associated heating, and as a consequence these layers 1, 3, 4, 5 are not appreciably heated by absorption of radiant energy or at least insufficiently heated for producing a ghost image. If the copy sheet 5 is white, as is the case ordinarily, the coating layer 4 should be colored or dark, it should be noted, however, that the coating layer 4 still must not be appreciably more heatable by absorption of radiant energy than are the layers 1, 3 and 5, and in any case said coating layer should be considerably less heatable by absorption of radiant energy than are the image surface portions 2. At the exposure, therefore, substantially only the image surface portions 2 are heated by absorption of radiant energy. The heated image surface portions 2 heat the coating layer 4 by conduction through the layers 1 and 3 to a temperature of the order of 50–150° C. at which elevated temperature the coating layer 4 has a greater ability to anchor itself on the copy sheet 5 than of being anchored to the base sheet 3. When the sheet set is separated after exposure, the copying portions of the coating layer 4 which were situated opposite the image surface portions 2 will consequently locally strip off from the areas to be copied and remain anchored to the copy sheet 5 to form on said sheet a reproduction of the image surface portions 2.

Copying in accordance with FIG. 1 is usually preferred with graphic originals having image surface portions 2 only on one face. However, with thick originals or originals having image surface portions on both faces reflex copying is usually preferred. The placing of the sheet set in face-to-face relationship at reflux copying will appear from FIG. 2. Here, during exposure, the radiant energy indicated by arrows passes through the layers 3–5 to the graphic original 1, 2 without appreciably heating the layers 3–5. The radiant energy further passes between the image surface portions 2 through the base sheet 1 of the graphic original without appreciably heating said base sheet, but within the image surface portions 2 the radiant energy is absorbed and heats said image surface portions. By conduction the image surface portions 2 then heat the coating layer 4 by the intermediary of the copy sheet 5 so that opposite the image surface portions 2 said copy sheet receives material from the coating layer 4 in the manner earlier described to provide a facsimile reproduction of the image surface portions of the graphic original.

The base 3 of the copying sheet may be a film such as a cellulose, cellulose derivative, polyester or polyamide film, or a non-porous paper sheet. The coating layer 4 may be coated on the base sheet 3 on conventional coating

machines of various kinds. When heated to a temperature within the range of 50 to 150° C., the coating layer 4 has a greater ability of anchoring itself on a paper surface pressed against it than of keeping itself anchored to the base sheet 3, but the coating layer must be substantially non-pressure-sensitive at normal room temperature. The coating layer comprises (1) wax as a major component, (2) a dye component and (3) a solvent agent, being a solvent for said wax and all of said dye component. The coating layer may contain other materials in addition to these three mentioned, if desired.

As the coating layer must be substantially non-pressure-sensitive at room temperature, the wax used as a major component of the coating layer must be a normally hard wax such as carnauba wax or montan wax.

In order to obtain facsimile copies having good stability to light so that their legibility is not impaired upon prolonged influence of light, the dye or dyes used in the coating layer must be of a special kind. Upon prolonged influence of light the dye or dyes may or may not change their hue but in any case they must retain a substantial color strength. The dyes of said kind are characterized by containing in their molecule at least one molecularly bound atom selected from the group consisting of selenium, tellurium, iodine, chromium, manganese, molybdenum, tungsten, iron, nickel, cobalt, and copper. Examples of such dyes are rhodamines modified with iodine and/or chromium. Rhodamine the molecule of which contains a chlorine atom and a carboxylic group can thus be treated in a manner obvious to the chemist to substitute a chromium atom for the hydrogen of the carboxyl group, or one amino group or both amino groups in the rhodamine molecule can be caused to form a complex chromium compound simultaneously as iodine is substituted for the above-mentioned chlorine atom. Further examples of such dyes are Luxol Fast Yellow TN, and Luxol Fast Brown K, sold by E. I. Du Pont De Nemours and Co., U.S.A., and Zaponechtviolett RR, sold by Badische Anilin- und Sodafabrik AG, Ludwigshafen, Germany. Particularly good results are obtained by means of soluble copper phthalocyanines.

Dyes of the above-mentioned kind are substantially not or at least insufficiently directly soluble in the wax of the coating layer but by using a special solvent agent which is a solvent for said wax and said dyes, it is possible to satisfy the requirement of having the dye or dyes in dissolved state in the coating layer and this to such an extent that the total dye concentration in the coating layer amounts to 3 to 11% by weight of the layer, whereby a very good color strength of the facsimile copies is obtained. For this result the solvent agent used contains a member of the group consisting of benzyl alcohol and compounds containing at least two hydroxyl groups and at most 8 carbon atoms in their molecule, such as glycerols, glycols, and glycol ethers. As examples of suitable compounds may be mentioned pentanediol, pentanetriol, pentanetetrol, hexanediol, -triol, -tetrol, -pentol, heptanediol, -triol, -tetrol, -pentol, -hexol, octanediol, -triol, -tetrol, -pentol, -hexol, and -heptol. Further examples are ethylene glycol, propanediol, butanediol and -triol, diethylene glycol, triethylene glycol, tetraethylene glycol, dipropylene glycol, and dibutylene glycol. For obtaining good results the solvent agent preferably should also contain a member of the group consisting of oleyl alcohol, oleic acid, capric acid, homologs of capric acid, lauric acid, and homologs of lauric acid.

In preparing a coating composition for forming the coating layer of the copying sheet the dye or dyes are first dissolved in the first-mentioned component of the solvent agent, for instance a mixture of ethylene glycol and octanediol, in heat. The solution formed is then mixed with the second component of the solvent agent, for instance oleyl alcohol, and this mixture is stirred into the molten wax. The coating composition formed is coated on the base sheet in the ordinary manner. The optimum

thickness of the coating layer is such that the square meter weight of the coating layer is 4 to 10 gr. If the dye material is used in an amount of less than 3% by weight of the coating layer the color strength of the facsimile copies is unsatisfactory and dye material in excess of 11% does not add materially to the color strength of the facsimile copies. Therefore, in practice it is preferred to use 3-5 % dye material in the coating layer.

Some examples of suitable compositions for the coating layer will be given in the following but the invention is not to be considered as limited in any way by these examples.

Example 1

	Parts by weight
Montan wax	865
9-octadecen-e-1-ol,cis-	50
A glycol	20
Benzyl alcohol	20
Copper phthalocyanine	40
Methyl violet	5

The coating layer gives a glue copy image which under the action of light certainly becomes green but still remains fully legible. The benzyl alcohol imparts the best solubility to the copper phthalocyanine. The glycol, for example propylene glycol or another relatively difficultly volatile glycol, has the additional advantage of preventing the paper from curling and also contributes to minimizing possible trouble in the form of static electricity in the copying material. Methyl violet is not lightfast but is used to adjust the original hue of the facsimile copies obtained.

Example 2

	Parts by weight
Montan wax	855
9-octadecen-e-1-ol,cis-	50
A glycol	30
Benzyl alcohol	20
Copper phthalocyanine	45

This coating layer gives a turquoise copy image of very high stability to light and almost unchanged color hue.

Example 3

	Parts by weight
Montan wax	815
9-octadecen-e-1-ol,cis-	50
Tetrapropylene-benzene-sulfonic acid, sodium salt ..	50
A glycol	20
Benzyl alcohol	20
Copper phthalocyanine	35
Methyl violet	10

This composition is also characterized by good dye solution and excellent coating properties, homogeneity and stability to light.

Example 4

Corresponds to Example 2 except that a rhodamine modified with chromium and iodine has been wholly or partly substituted for the copper phthalocyanine. The print can thereby be imparted all intermediate hues from blue-green via blue and violet to red. The stability to light is

very high and the color hue is changed but insignificantly with time. It is also possible to use Luxol Fast Yellow TN or Luxol Fast Brown K or Zaponechtviolett RR instead of part of the copper phthalocyanine in Example 2. The concentration of said dyes can be increased up to totally about 110 parts by weight, if desired.

What I claim and desire to secure by Letters Patent is:

1. A heat-sensitive copying sheet for producing a thermographic facsimile copy, comprising a base sheet, a heat-sensitive coating layer on one side of said base sheet, said coating layer being substantially non-pressure-sensitive at room temperature and comprising wax as a major component, a dye component in a total amount of 3 to 11% by weight of said layer, said dye component containing at least one dye retaining substantial color strength upon prolonged influence of light and containing in its molecule at least one molecularly bound atom selected from the group consisting of selenium, tellurium, iodine, chromium, manganese, molybdenum, tungsten, iron, nickel, cobalt, and copper, and a solvent agent being a solvent for said wax and said dye component and being present in said layer in an amount sufficient to hold all of said dye component in dissolved state in said layer and containing at least one member of the group consisting of benzyl alcohol and compounds containing at least two hydroxyl groups and at most 8 carbon atoms in their molecule, and said coating layer when heated to a temperature within the range of 50-150° C. having a greater ability to anchor itself on a paper surface pressed against it than of being anchored to said base sheet.

2. A copying sheet according to claim 1, in which said component of said solvent agent is a member of the group consisting of glycerols, glycols, and glycol ethers.

3. A copying sheet according to claim 1, in which said solvent agent further contains a member of the group consisting of oleyl alcohol, oleic acid, capric acid, homologs of capric acid, lauric acid, and homologs of lauric acid.

4. A copying sheet according to claim 1, in which said dye is a copper phthalocyanine.

5. A copying sheet according to claim 1, in which said dye is a rhodamine modified to contain in its molecule at least one member of the group consisting of iodine and chromium.

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MURRAY KATZ, *Primary Examiner*.