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(54) Title: THERMOPLASTIC IMAGING SYSTEM (57) Abstract A thermographic material of the type in which a thermoplastic layer on a photosensitive semiconductor is electrostatically charged and the stack is then heated so that the charge induces structural changes in the thermoplastic layer to provide a hologram or other image. The thermoplastic layer contains or is formed with a layer of magnetic particles, e.g. of $\delta\text{Fe}_2\text{O}_3$ or Fe_3O_4 , which, when a magnetic field is applied simultaneously with heating, greatly improves the results of signal to noise ratio of the imaging system.		

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THERMOPLASTIC IMAGING SYSTEM**SPECIFICATION****CROSS REFERENCE TO RELATED APPLICATION**

This application replaces copending Provisional Application Ser. No. 60/013578 filed 18 March 1996 and claims the benefit thereof under 35 USC 120.

FIELD OF THE INVENTION

Our present invention relates to a thermoplastic imaging system of the type in which a thermoplastic material can be electrostatically shaped by softening of the thermoplastic and rehardening to alter the surface or structure of the thermoplastic in response to a latent image formed by electrostatically charging the system in the presence of a photosensitive semiconductor. The invention, therefore, relates to thermographic materials, i.e. a film which can be provided with an image in the form of changes in the thermoplastic structure, to a method of producing such images and to equipment utilizing such film.

BACKGROUND OF THE INVENTION

The formation of images, for example, holograms and like images in a film having a thermoplastic layer by a process in which the film is charged, the charge distribution is modified to produce a latent image by reason of the presence of a semiconductive layer in the film, and the thermoplastic

layer is heated to alter its structure based upon the charge distribution is known. In this system, in a carrier or support, e.g. of a polyester material such as Mylar, a conductive layer forms an electrode for charging the film and a semiconductive layer, especially an amorphous inorganic semiconductor, is provided to generate a latent image upon appropriate exposure.

The semiconductor layer is disposed adjacent a thermoplastic layer which carries the electric charge when the film is charged. The application of heat to soften the thermoplastic layer causes changes in the thermoplastic structure which may result in deformation on the surface of the film based upon the charge distribution and/or changes in internal refractivity or optical conductivity, by reason of polymerization, chain modifications and the like, or any combination of the two so that, upon cooling, an image is fixed in the film which can be viewed when light is directed onto the thermographic material.

Such systems have been found to be particularly effective in the production of so-called instant holograms, i.e. for a form of holography which allows the holographer to permanently capture a hologram instantly and without a dark-room, developing chemicals, or any of the much more complex development techniques previously required. The film, depending upon construction, transparency and the like can be used for transmission and reflection holograms, and the applications of the film can include interferometry, security applications, verification, authentication, etc. The film may be

used for other optical image reproduction or production techniques as well.

Reference may also be had to frost-imaging technology in which an electrostatic charge forms the basis for a latent image in which a layer is subsequently transformed so as to have regions which are rendered somewhat opaque or frosted. Attention can be directed, in this regard, to the following U.S. Patents: 3,196,011; 5,378,565; 5,344,734; 5,076,990; 5,013,624; 4,732,831; 4,695,528; 4,583,833; 4,548,885; 4,492,747; 4,397,930; 4,282,295; 4,077,803; 4,065,308; 4,047,238; 4,032,338; 4,021,236; 4,018,603; 3,992,090; 3,982,939; 3,982,936; 3,980,476; 3,973,958; 3,973,957; 3,973,954; 3,973,953; 3,961,950; 3,915,700; 3,897,247; 3,892,567; and 3,858,973.

OBJECTS OF THE INVENTION

It is the principal object of the present invention to provide an improved film or imaging material which will advance the principles described above and, in particular, increase the versatility of thermographic techniques for holography and other imaging systems.

It is another object of this invention to provide an improved method of making an image which can significantly increase the resolution over that obtainable with earlier thermographic techniques using thermoplastic layers as imaging materials.

It is still another object of this invention to provide an improved material for thermoplastic imaging.

SUMMARY OF THE INVENTION

These objects and others which will become apparent hereinafter are attained, in accordance with the present invention in a system for thermographic imaging utilizing thermally introduced changes in a thermoplastic layer to develop an image corresponding to a latent image resulting from a charge distribution, that layer in which the thermoplastic layer contains a particulate magnetic material which can be affected by an applied magnetic field.

It has been found, surprisingly, that the electrostatic charge intensity which is required to effect distortion of the thermoplastic layer can be reduced if, simultaneously with the heating of the thermoplastic layer, a magnetic field is applied to the material. The result is an enhanced resolution over thermoplastic imaging films without such magnetic materials in the plastic layer. Furthermore, a magnetic latent image may be applied for superimposition upon the electrostatic latent image utilizing magnetic recording means if desired and the magnetic material present in the thermoplastic layer may be used to record information in addition to image information in the form of the magnetic latent image mentioned above.

For example, information as to the extent of exposure to light, e.g. to laser beams in the case of a holographic system or information as to degree of heating may be recorded in the film and can be read at the camera or other equipment for processing the film.

The magnetic material which can be used includes any of the particulate materials utilized in the production of

magnetic recording tapes and like magnetic imaging, including ferrites, and other iron oxides, chromium oxides and the like, as well as particulate permanent magnet materials. The magnetic materials can be homogeneously distributed in the thermoplastic layer or can be distributed in a particular pattern therein.

The amount of magnetic material should be the maximum which can be accommodated without detriment to the physical properties of the thermoplastic layer and to the electrostatic recording properties thereof and the particle size of the magnetic material should be the smallest possible to ensure the highest resolution.

One of the surprising advantages of the presence of magnetic materials in the thermoplastic layer is an increase in the photosensitivity of the film so that lesser levels of light or lower intensity can produce the latent image. The presence of the magnetic material and the use of a magnetic field conjointly with heating increases the signals to noise ratio of the registered image and allows lower voltage levels to be employed for charging the film, thereby reducing any adverse effect of corona instability.

The magnetic material can be $\gamma\text{-Fe}_2\text{O}_3$ while some 5 to 25% of the thermoplastic layer may consist of the magnetic powder. The particle size of the magnetic powder should be from 0.005 to about 0.09 μm with best results about 0.03 μm .

The thermoplastic layer can be composed of a copolymer of styrene and butylmethacrylate which consists of 25 to 75% styrene and 75 to 25% butylmethacrylate, with good results being a 50% by weight styrene and 50% by weight

butylmethacrylate composition. The thickness of the thermoplastic layer may range from 0.5 to 2.5 μm and preferably is around 1.5 μm .

The magnetic powder is suspended in the polymer composition dissolved in toluene or some other solvent and the solvent is evaporated.

The photosensitive layer can be an amorphous semiconductor of the arsenic-sulfur, arsenic-selenium or arsenic-sulfur-selenium type. Typical of the amorphous inorganic semiconductors used may be those having a formula in the range $(\text{As}_2\text{S}_3)_{0.3} (\text{As}_2\text{Se}_3)_{0.7}$ to $(\text{As}_2\text{S}_3)_{0.5} (\text{As}_2\text{Se}_3)_{0.5}$.

The presence of the magnetic material distributed in the thermoplastic layer allows the molecular movement of the magnetic particles to be detected during the heating process and permits, according to the invention, control of the heating process during the latter so as to achieve particularly efficient control of the heating and highly reproducible imaging results. The molecular movement in the thermoplastic layer and hence movement of the magnetic particles is a direct reflection of the temperature of the body of thermoplastic and can be measured with particularly high precision by conventional magnetometers. The latter can be used to regulate the heating or to cut it off when the desired temperature or degree of heating has been obtained.

While the aforescribed magnetic materials are highly effective, best results are obtained when the magnetic material is Fe_3O_4 and the Fe_3O_4 powder is present in an amount of 7% by weight of the thermoplastic layer. When the powder is incorporated into that layer, 93% by weight of the layer

may be composed of polyepoxypropylcarbazole admixed with about 30% by weight of dioctylphthalate.

As will be described below, the magnetic powder need not be incorporated into the thermoplastic layer exclusively and some or all of the magnetic powder may form a magnetic powder layer on the thermoplastic layer. In that case, the thermo- plastic layer can include 100% of an intimate blend of the

polyepoxypropylcarbazole with the 30% by weight of dioctylphthalate and the magnetic particle layer may be used in a homogeneous and uniform thickness layer in an amount of 7% by weight of the thermoplastic layer. In practice the magnetic particle layer can be of a thickness equal to the order of the particle size, namely 0.09 μm to 0.005 μm

In the case of the polyepoxypropylcarbazole/
/dioctylphthalate thermoplastic layer a preferred thickness is about 2.0 μm .

When reference is made here to association of the magnetic powder with the thermoplastic layer, it will be understood that that association can involve:

- (a) a uniform dispersion of the magnetic powder in the thermoplastic layer;
- (b) a graded distribution of the magnetic powder in the thermoplastic layer, i.e. a variation in the concentration of the magnetic powder from one side of the thermoplastic layer to the other;
- (c) A layer of the magnetic powder along one or both of the surfaces of the thermoplastic layer;

(d) a combination of a layer of the magnetic powder along one or both of the surfaces of the thermoplastic layer with the incorporation of particles in the thermoplastic mass as in (a) or (b); or

(e) a sandwich construction in which a layer of the magnetic material is sandwiched between layers of the thermoplastic material, possibly in combination with incorporation of magnetic particles in one or the other of the sandwiching layers.

The choice which will be made will depend, of course, on the influence which is desired for the magnetic field contribution to the image to be formed.

A stack or laminate according to the invention can also include a very thin layer of ferromagnetic material of a surface-active substance.

We have found that the principles described above are applicable where the latent image may be generated by other means, for example, electrostatically applied to the layer of thermoplastic material by other than the use of a photoconductor. In that case, the imaging material may include an electrode to assist in the application of the electrostatic charge but even may be free from an electrically conductive layer when the latent image is generated, for example, by other electrostatic means or by magnetic means. The application of a magnetic field to enhance the thermographic effect remains and the latent image can be developed by heating to effect the structural change in the thermoplastic material. The method of the invention can then comprise the steps of:

(a) providing an imaging material comprised at least of a layer of a thermoplastic material and magnetic particles associated with the layer;

(b) forming a latent image in the material;

(c) applying a magnetic field to the material; and

(d) heating the material to effect a structural change in the layer to transform the latent image into a sensible image.

BRIEF DESCRIPTION OF THE DRAWING

The above and other objects, features, and advantages will become more readily apparent from the following description, reference being made to the accompanying drawing in which:

FIG. 1 is a diagrammatic section through an imaging material in accordance with the invention;

FIG. 2 is a diagram of method steps of the invention;

FIG. 3 illustrates a surface pattern created utilizing the material of the invention in a cross sectional view drawn to a larger scale than FIG. 1;

FIG. 4 is a diagram illustrating another aspect of the invention;

FIG. 5 is a cross sectional view similar to FIG. 1 showing another imaging material in accordance with the invention;

FIG. 6 is a cross sectional view of a thermographic imaging material which does not utilize a photoconductor

within the material according to another feature of this invention; and

FIG. 7 is a cross sectional view through a thermographic material representing a modification of the material of FIG. 6.

SPECIFIC DESCRIPTION

The imaging material shown in FIG. 1 has been depicted with exaggerated thicknesses of the various layers and has been illustrated in a particular stack configuration although it should be understood that the layers may be switched in location or additional layers may be produced as long as the conductive layer enables charging of the stack, the layers are disposed so that the light, e.g. laser illumination, can reach the photo-sensitive semiconductive layer, the heating can be applied to the thermoplastic layer and the entire stack is supported so that, when the thermoplastic layer has its structure altered, the alterations can affect light which is transmitted through the film or reflected from it so that the image can be displayed.

More particularly, the material of FIG. 1 can comprise a support 10, preferably of a polyester but also, if desired, of any other transparent or translucent material with a melting point well above the softening temperature of the thermoplastic layer, the ability to receive an adherent coating and optical properties which do not interfere with exposure of the photosensitive layer or the transmission or

reflection reproduction of the resulting image. The support should be a good heat conductor as well to ensure that, if the heating of the thermoplastic layer is effected by contact with a heated surface applied to the polyester support, the latter will not prevent raising the temperature of the thermoplastic layer to the softening point.

The layer 10 is preferably composed of Mylar, i.e. a polyethylene terephthalate.

On the polyester support is a metallic layer 11 which constitutes an electrode, e.g. of aluminum or chromium. The thickness of this layer is not significant and it may be a monolayer. Its purpose is to enable the charging of the stack electrostatically. It may be connected to a conductive surface of a frame in the case of a slide or may otherwise be exposed so that it can be connected to one pole of an electrode charging source or grounded when a pole of that source is grounded.

On the metallic layer 11 is a layer of an arsenic-sulfur, arsenic-selenium or arsenic-sulfur-selenium amorphous semiconductor of a formula in the range given above. With the semiconductor is photosensitive material which, when exposed to light, will alter the uniformity of an electrostatic charge applied to the stack to produce a latent image.

On the photosensitive semiconductor layer 12 is a layer 13 of thermoplastic, preferably the styrene-butylmethacrylate copolymer (50% styrene, 50% butylmethacrylate) which can contain 13% by weight of magnetic powder as represented at 14, the magnetic powder being, for example, $\delta\text{-Fe}_2\text{O}_3$

of a particle size of about $0.03\ \mu\text{m}$. The thickness of the thermoplastic layer is about $1.5\ \mu\text{m}$.

As has been explained, the stack 11-14 and represented at 15 in FIG. 2, is charged by being exposed to a corona discharge electrode connected to a high voltage dc source 17 whose other pole is grounded, the layer 11 being grounded as being represented at 18. The result is a uniform electrostatic charge on the thermoplastic layer 13. When the stack 15 is exposed to light, e.g. the laser beams in the creation of a hologram as shown at 19 and 20, in regions of such exposure, the photosensitive semiconductor is activated to alter the electrostatic charge distribution, thereby creating a latent image in the material based upon the electrostatic charge distribution. Heat can then be applied to a surface of the stack 15, e.g. by a resistively heated plate 21 so that electrostatic forces act on the softened thermoplastic material and cause changes in the surface shape as represented by the peaks 22 and the valleys 23 on the surface of the thermoplastic layer 13 as shown in FIG. 3 or changes are made in the polymer structure itself.

At the same time, a magnetic field is applied transversely to the layer, e.g. by a solenoid 24 (FIG. 2), the magnetic field being represented at H.

By way of example, when the external magnetic field is $9400\ \text{A/m}$ simultaneously with heating for a material as described which has been charged with a corona voltage of 7kV , using an expose time of 0.3 seconds and an illumination intensity of

5×10^{-2} lux, the image resolution can be increased to 1500 mm⁻¹, the diffraction efficiency can be not less than 15% and the signal to noise ratio can be increased to three times that of a system without the application of the magnetic field and magnetic particles.

An induction field of 9567 A/m² can induce deformation in the thermoplastic when heated to its softening point without the optically produced latent image to generate a pattern on the thermoplastic surface which corresponds to the distribution of the particles in the thermoplastic and hence a magnetic latent image therein.

Thus the system can be used, by controlled distribution of the magnetic particles in the thermoplastic layer, to create a latent image therein which is developed by the application of the magnetic field simultaneously with heating and, when an optical latent image is provided, can superimpose the magnetic image on the optical image. The magnetic material also forms an ideal recording medium and in that case a magnetic head can record at 30 on the slide, whatever information may be of interest where the hologram is part of a document security item or for other purposes, the material being read at 31, e.g. by a reading head in the camera with the information being displayed from the reading head or used to control the exposure, heating and magnetic field application in the camera as represented at 32.

In FIG. 2, the magnetometer or other measuring device 33 for the magnetic field in the vicinity of the film, is coupled to the heater to control the latter in response to

measurements of the magnetic field resulting from the presence of the magnetic particles.

In FIG. 5, the imaging material which can be used in the manner already described and with the apparatus which has been described, comprises a polyester support 10, the conductive layer 11 and the layer 12 of amorphous semiconductor, all as has been described. Here the thermoplastic layer 13, however, is composed of the polyepoxypropylcarbazole containing 30% by weight of dioctylphthalate and the magnetic particles are provided in a magnetic powder layer 14' on the thermoplastic layer and consists of Fe_3O_4 particles in a particle size of about $0.3\ \mu\text{m}$ and a thickness of about $0.1\ \mu\text{m}$. A further protective film 13' of the polyepoxypropylcarbazole and dioctylphthalate blend is provided on the magnetic powder layer.

That film operates as has been described for the film of FIG. 1.

SPECIFIC EXAMPLE

The imaging material consists of a polyesterterephthalate support having a thickness of $100\ \mu\text{m}$ and onto which a conductive chromium layer is vapor-deposited to a thickness of $0.01\ \mu\text{m}$. On this conductive layer, an amorphous photoconductive layer of an arsenic-sulfur-selenium composition is applied. This composition consists 30% by weight of $\text{As}_2\text{S}_{1.5}\text{Se}_{1.5}$. The amorphous semiconductor layer has a thickness of $2\ \mu\text{m}$. On the amorphous semiconductive layer a $2\ \mu\text{m}$ thick thermoplastic layer of polyepoxypropylcarbazole containing 30%

by weight dioctylphthalate and 7% by weight of Fe_3O_4 magnetic powder of a particle size of $0.03\ \mu\text{m}$, is applied.

The resulting film is placed in a recording device as has been described and is heated for 5 seconds to a recording temperature of 80°C . The corona electrode potential is 5.5 kV. A magnetic field with an intensity of 600 Oersted was applied

perpendicular to the surface of the thermoplastic medium over a period of 60 seconds. At the 51st second following the application of the magnetic field, the recording stack was exposed to the corona discharge of 5.5 kV and after a further second was exposed to light from the image for a period of one second.

A photographic sensitivity was not less than 20 $(\text{lux}\cdot\text{sec})^{-1}$ and the resolution was 1.5 times higher than that of a photothermoplastic medium without the magnetic particles and exposure to the magnetic film.

In FIG. 6, we have shown a thermographic material which omits the photoconductive layer and thus, on a polyester film base 10, can have the conductive layer 11 and a thermoplastic layer 13 of the type described in which magnetic particles 14 are dispersed. Utilizing the conductive layer 11, an electrostatic charge can be applied to the thermoplastic layer 13 and a latent image can be formed electrostatically in the material by, for example, differential discharge of the electrostatic charge or by controlling how the electrostatic charge is originally applied. If the conductive layer 11 is omitted, the material can be electrostatically charged by plates flanking the material.

In another alternative, the latent image can be formed magnetically by providing different magnetic fields to various locations of the film. The film can then be subjected as has been described to heating to develop the latent image and form a sensible image, i.e. an image which can be sensed by the passage of light through the film or by reflecting light from the film. When the image is in a latent electrostatic image, the resolution can be enhanced by the application of a magnetic field in the manner already described.

In FIG. 7, we have shown a material similar to FIG. 6 but wherein the magnetic particles form a layer 14' which is protected by an additional layer 13' of the thermoplastic. The film of FIG. 7 is used in a similar manner to the film of FIG. 6.

IT IS CLAIMED:

1. A thermographic material comprising:

a stack of at least three layers including a thermoplastic layer heatable to a softening temperature to alter a structure of said thermoplastic layer in accordance with an electrostatic charge distribution thereof, a photosensitive semiconductive layer adapted to be exposed to light to establish said charge distribution on said thermoplastic layer in the form of a latent image, and a conductive layer enabling electrostatic charging of said stack; and

particles of a magnetic material associated with said thermoplastic layer and promoting structural change in said thermoplastic layer upon application of a magnetic field to said stack simultaneously with heating thereof to said softening temperature.

2. The thermographic material defined in claim 1 wherein said magnetic material is Fe_3O_4 , a ferrite, chromium oxide or metal particles.

3. The thermographic material defined in claim 2 wherein said magnetic material is present in an amount of 5 to 25% in said thermoplastic layer.

4. The thermographic material defined in claim 3 wherein said magnetic material is a powder present in a particle size of 0.005 to 0.09 μm .

5. The thermographic material defined in claim 1 wherein said stack is provided on a polyester support.

6. The thermographic material defined in claim 5 wherein said semiconductive layer is composed of an arsenic-sulfur, arsenic-selenium or arsenic-sulfur-selenium amorphous layer.

7. The thermographic material defined in claim 1 wherein said particles of magnetic material are distributed in said thermoplastic layer.

8. The thermographic material defined in claim 1 wherein said particles of magnetic material form a layer on said thermoplastic material.

9. The thermographic material defined in claim 1 wherein said particles of magnetic material are distributed in said thermoplastic layer and form a layer on said thermoplastic layer.

10. The thermographic material defined in claim 1 wherein said thermoplastic layer is composed of a styrene-

butylmethacrolate copolymer or a polyepoxypropylcarbazole containing dioctylphthalate

11. A method of producing an image, comprising the steps of:

(a) electrostatically charging a stack of at least three layers including a thermoplastic layer associated with magnetic particles and heatable to a softening temperature to alter a structure of said thermoplastic layer in accordance with an electrostatic charge distribution thereof, a photosensitive semiconductive layer adapted to be exposed to light to establish said charge distribution on said thermoplastic layer in the form of a latent image, and a conductive layer enabling electrostatic charging of said stack to produce an electrostatically charged stack;

(b) applying light to said stack to create an electrostatic charge distribution of said thermoplastic layer in the form of said latent image; and

(c) simultaneously

heating said stack to raise a temperature of said thermoplastic layer to a softening temperature and alter said structure of said thermoplastic layer in accordance with said electrostatic charge distribution thereof, and applying a magnetic field to said stack promoting structural change.

12. The method defined in claim 11 wherein said magnetic material is a ferrite, chromium oxide or metal particles.

13. The method defined in claim 12 wherein said magnetic material is present in an amount of 5 to 25% in said thermoplastic layer.

14. The method defined in claim 13 wherein said magnetic material is a powder present in a particle size of 0.005 to 0.09 μm .

15. The method defined in claim 11 wherein said stack is provided on a polyester support.

16. The method defined in claim 15 wherein said semiconductive layer is composed of an arsenic-sulfur, arsenic-selenium or arsenic-sulfur-selenium amorphous layer.

17. The method defined in claim 11, further comprising the step of recording in the magnetic material of said thermoplastic layer information and reading the recorded information.

18. The method defined in claim 11 wherein said magnetic material has a distribution forming a latent image in said thermoplastic layer which is developed by the heating and application of said magnetic field to superimpose the latent

image of said magnetic material on an image developed from the latent image of said charge distribution.

19. The method defined in claim 11 wherein said magnetic particles are distributed in said thermoplastic layer.

20. The method defined in claim 11 wherein said magnetic particles form a layer on said thermoplastic layer.

21. A method of producing an image comprising the steps of:

(a) providing a stack of layers including at least one layer composed of a thermoplastic and containing magnetic particles in a distribution forming a latent image;

(b) heating said stack to a temperature sufficient to soften said thermoplastic layer; and

(c) applying simultaneously with the heating of said thermoplastic layer magnetic field of a field strength sufficient to effect a change in structure of said thermoplastic layer simultaneously with the heating thereof, thereby structurally altering said thermoplastic layer; and

(d) cooling said thermoplastic layer to freeze the change of structure therein.

22. The method defined in claim 21, further comprising the steps of:

detecting a magnetic field resulting from presence of the magnetic particles in said thermoplastic layer; and

controlling said heating of said stack in response to the detected magnetic field.

23. A method of producing a sensible image, comprising the steps of:

(a) providing an imaging material comprised at least of a layer of a thermoplastic material and magnetic particles associated with said layer;

(b) forming a latent image in said material;

(c) applying a magnetic field to said material; and

(d) heating said material to effect a structural change in said layer to transform said latent image into a sensible image.

24. The method defined in claim 23 wherein said magnetic particles are distributed in said layer.

25. The method defined in claim 23 wherein said magnetic particles are formed as a layer on said layer of thermoplastic material.

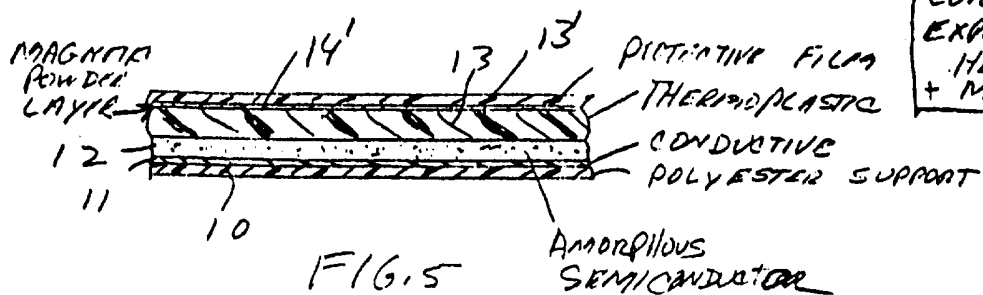
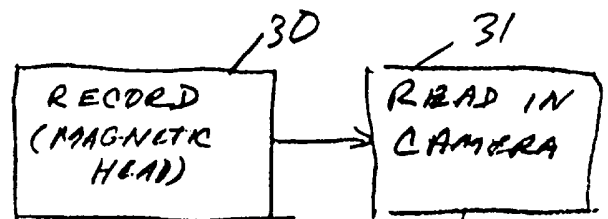
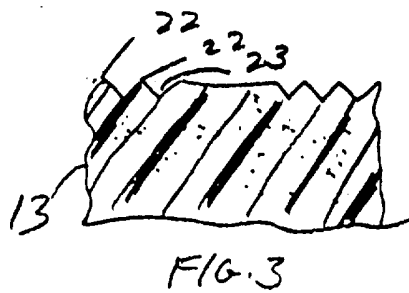
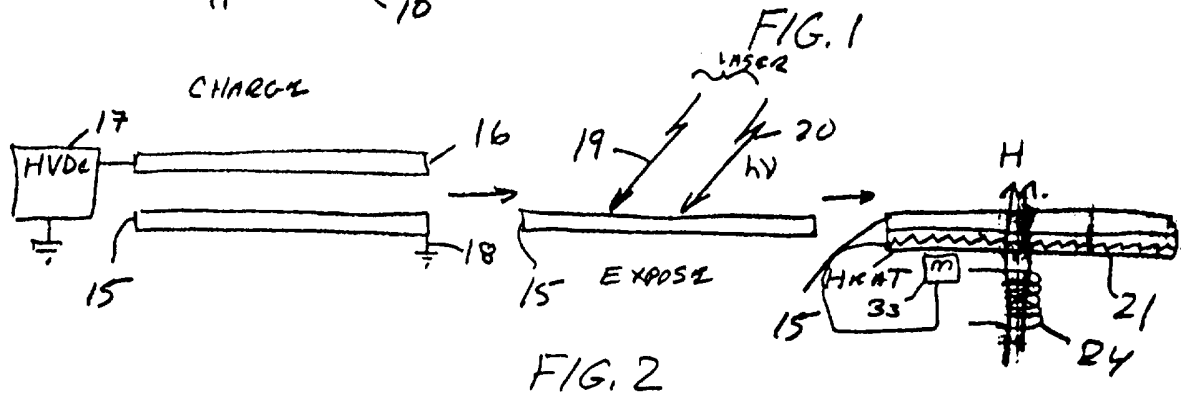
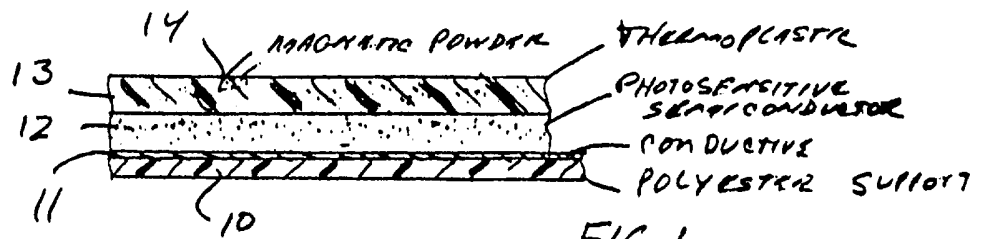
26. The method defined in claim 23 wherein said latent image is formed at least in part by applying an electrostatic field to said material.

27. The method defined in claim 23 wherein said latent image is formed at least in part by application of a magnetic field to said material.

28. A thermographic imaging material comprising a thermoplastic layer and magnetic particles associated with said layer whereby a latent image can be formed in said material and developed by heating of said material to effect a structural change in said layer to transform said latent image into a sensible image.

29. The thermographic material defined in claim 28 wherein said particles are distributed in said layer.

30. The thermographic material defined in claim 28 wherein said particles are in the form of a magnetic layer on said thermoplastic layer.



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