

RESERVE COPY

PATENT SPECIFICATION

DRAWINGS ATTACHED

1,143,702

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Int. Cl.:—G 03 c 5/34

COMPLETE SPECIFICATION

A process for Developing Diazotype Materials

We, ADDRESSOGRAPH-MULTIGRAPH CORPORATION, a Corporation organized under the laws of the State of Delaware, United States of America, of 1800 West Central Road, Mount Prospect, Illinois, United States of America, do hereby declare the invention for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to methods for developing diazotype materials and more particularly relates to the development of two-component diazotype materials by the application of controlled amounts of a concentrated alkaline liquid.

Diazotype reproduction processes based on light sensitive diazonium compounds and their ability to form azo dyes with a coupling component under proper pH conditions are generally well-known. The various techniques for carrying out the development include the moist or semi-moist process, the dry or ammonia process, and more recently the thermal process. The copying materials adapted for the moist process are referred to as one-component materials since they contain only the light sensitive compounds and the coupling component is provided in a separate buffered developing solution and applied at the time of development. Diazotype materials developed in an atmosphere of water vapor and ammonia gas are generally referred to as two-component materials since they contain both the diazonium salt and the coupling component stabilized against premature coupling in an acidic medium. In the thermal systems heat is employed to release the alkaline agents which are contained along with the diazonium salt and the coupler on the copy material. Each of these systems provides a successful copying process but not without certain disadvantages that limit their utility which would otherwise permit diazo copying to enjoy more widespread application as a reproduction medium.

[Price 4s. 6d.]

Two-component materials have found wide use in the reproduction of engineering drawings. The process of developing with ammonia gas and water vapor is fast and essentially dry. Ammonia vapor readily penetrates into the exposed copy sheet surface and permeates the light sensitive coating containing the acid stabilizers, shifting the pH of the coating to the alkaline side. Any excess of ammonia enhances the rate of the reaction and volatilizes from the copy sheet without leaving a residue. One of the major objections to the use of ammonia in developing diazotype materials is the strong odor of ammonia in the copying area as well as the tendency for the odor to linger on the copies themselves. Installation of ammonia reproduction machines requires venting of the equipment to the outdoors to remove the toxic vapors. This limits the installation of the equipment to sites where the venting may be conveniently accomplished. The equipment is further complicated by the need for a gas developing chamber to contain the ammonia gas and water vapor and the other appurtenances required for feeding the developing ingredients to the chamber. The equipment, understandably, is not well suited for office installations.

The one-component systems have effectively eliminated the odor problem by applying liquid developers directly to the copy sheet. Reproduction equipment for use with the one-component type papers requires the use of heating elements in order to dry the copy sheet after development.

Thermal systems, while offering the advantages of dry copying without the presence of undesirable odors, suffer in that the copying materials themselves may lack stability. The machines are objectionable because of the amount of heat given off into the surrounding work area.

Attempts have been made to simplify the processing of diazotype materials, particularly the two-component type, by using alkaline

liquids applied directly to the latent image bearing surface. These prior attempts to eliminate the complexities of gaseous development have been unsuccessful for the following reasons:

1. In general, the use of dilute alkaline solutions produced prints of rather low image density when compared to development with ammonia.

2. The application of large quantities of dilute alkaline solution did not appreciably improve image density and rendered the sheet moist so that it required a drying step before it could be used.

3. Alkaline developer solutions formulated with non-volatile inorganic salts, such as disclosed in British patent specification No. 295,993, required an inordinately long time to develop into a print having suitable image density.

4. The dye shade of the azo dye images obtained with conventional two-component diazotype materials developed with inorganic salts deviated from the hues obtained with ammonia. The off-shade images in some instances were weaker, giving prints of poor contrast.

5. Developer solutions containing high concentrations of inorganic salts resulted in crystals of these salts forming on the various rollers causing streaked, uneven development. Such concentrated solutions also tended to leave a residue of the salt on the copy causing a more rapid discolouration of the print background.

Attempts to control the fluid application by adapting the techniques well-known for developing the one-component diazotype materials such as disclosed in British patent specification No. 544,882 to the development of two-component diazotype papers using alkaline liquids resulted in applying too much fluid. These prior techniques rely, in the main, on first applying an excess amount of developer solution to the copy surface, which excess is then squeezed off. The use of grooved roller systems generally required some form of drying of the copy after development.

The invention provides a method for making a reproduction of an original on a two-component diazotype copying material characterized by (a) exposing the copying material to an actinic radiation pattern corresponding to the light and dark areas on said original producing a latent image thereon, (b) feeding a controlled amount of an alkaline liquid medium by means of a carrier body to a copy developing region, (c) passing said copy material through said region transferring an amount of said liquid not exceeding 3 grams per square meter to said latent image bearing surface, said amount of liquid being evenly and uniformly spread over said area, and forming an azo dye image on said surface.

In summation, it can be said that prior

attempts to simplify development of diazotype materials, and in particular the development of two-component copy papers, were unable to match the dry conditions of ammonia and the speed with which the latent image is developed.

The processes were by no means dry in the sense that the copy sheet still required some form of drying and, finally, the quality of the reproduction was lacking in uniformity and image density by comparison to the quality obtained with ammonia development.

Reference is now made to the accompanying drawings in which:

Figure 1 is a flow diagram of the process comprising the invention,

Figure 2 is a diagrammatic elevational view of the developing apparatus employed in carrying out the purpose of this invention.

To secure the advantages of the known techniques of development of diazotype materials, particularly the two-component type, but to avoid the limitations thereof, a process has been developed which employs liquid developers and yet produces a dry copy directly from the developing step. This is accomplished by supplying controlled amounts of a concentrated developer solution, comprising an aliphatic amine, to a developing region and then passing an exposed diazotype paper through the developing region. At the developing region the controlled small amount of liquid is applied directly to the latent diazo image bearing surface to effectively develop the azo dye image and the prints emerge dry and odor free, ready for use.

When referring to a controlled amount of liquid hereinafter it will be understood to mean amounts not in excess of 3.0 grams of developer per square meter of the image bearing surface and preferably from about 0.5 grams to 2.0 grams per square meter. Surprisingly, small quantities of developer liquid applied over the entire area uniformly as the copy passes through the developing region immediately develop the latent image into a readable azo dye image.

Within the developing region the limited quantity of liquid is applied under pressure being spread out uniformly over its entire surface in a layer having a thickness of about 1 micron, which is sufficient to cause rapid image development. The azo dye image is found to develop up evenly and uniformly everywhere across the copy area indicating the effectiveness of the method. A visible, readable image begins to form instantaneously as the copy emerges from the developing region and increases in density attaining its maximum image density well within one minute and usually within 15 seconds. Understandably, the rate at which maximum density is reached will depend in part on the couplers and diazonium compounds comprising sensitizing formulation used in making the copy sheets.

The condition of the sheet described as

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being dry may be defined as being dry to the touch as soon as it emerges from the developing region. No further drying is required and the copy may be used immediately. The copies are substantially free of objectionable odors. They are flat lying, free of curl, cockles or other irregularities that are normally attendant upon paper surfaces to which liquids have been applied. Further, the copies are not limp due to the presence of excess moisture that would prevent them from being filed, folded or handled by the automatic stacking devices of copying equipment. The emerging copies may be written upon with pen or pencil and otherwise handled.

Giving consideration to the subject of developing two-component diazotype materials in general, it is understood that the reaction is a two-step process, namely:

1. Increasing the pH of the environment in which the diazonium salt and coupling component are found to the level where the coupling action can take place, and

2. The coupling reaction between the coupling component and the diazonium salt. It is the first reaction to which this invention is directed. The second reaction or coupling proceeds at a rate determined solely by the particular reactants once the proper pH has been provided.

In any practical developing process the developer must penetrate the surface of the sheet and neutralize the acid uniformly and evenly. It will be appreciated that incomplete neutralization in certain areas will leave the diazonium salt uncoupled and result in an irregularly developed copy. Accordingly, it is critical that the alkaline liquid developer be applied over the entire surface uniformly so as to achieve an even and uniform treatment. The liquid must react rapidly with the acid component; it should not discolor the print background; and it should not be absorbed into the body of copy material but remain only at the surface thereof. This presents the problem of applying just the right amount of fluid to accomplish development without excessively wetting or saturating the sheet. The composition of the developer liquid will be considered further in greater detail hereinafter.

Giving pause to consider the nature of the surface of the copy sheet coated with a solution containing a light sensitive diazonium salt, a coupler, and an acid component, it has been found that the chemicals in the sensitizing solution are deposited on the individual random cellulose fiber structure at the paper surface and, in the case of films, absorbed into the treated or prepared portion of the surface. For the liquid developer to be effective, it must penetrate and completely wet out the random fiber structure or the treated film surface.

Referring to Figure 1, there is shown the flow diagram setting forth the steps embody-

ing this invention. A two-component diazotype copy sheet is first exposed to actinic radiation through a suitable translucent original in the conventional manner. The developer liquid is continuously fed to a carrier; thereafter the amount of said liquid is metered to provide the optimum quantity necessary for development; the metered amount of developer is conveyed to the developing region; in the developing region the metered liquid is transferred under pressure to the exposed copy sheet as the latter passes through the developing region from whence it emerges dry and ready for handling.

Referring to Figure 2, there is shown one form of apparatus, identified by the general reference character 20, capable of carrying out the processing steps of the instant invention as shown on the flow diagram in Figure 1. Such apparatus may be modeled after a conventional offset duplicator such as described in U.S. patent 2,165,235 which utilizes multiple roller in conjunction with transfer drums or cylinders to achieve the application of very small quantities of a liquid spread uniformly and evenly over large areas.

A pair of rubber covered rollers 22, 24 in conjunction with the rotatable larger diameter drums or cylinders 26, 28 are disposed with their axes in parallel alignment so that their respective surfaces may be brought into rolling contact. Roller 22 is partially immersed along its length in a trough 30 containing a supply of developer liquid 32. Means (not shown) are provided for driving the cylinder 26 and by suitable gearing (not shown), in timed relation with rollers 22, 24 and cylinder 28. Roller 24 oscillates making contact alternately with the surface of rollers 22 and 26 providing a controlled quantity of the developer 32 to the liquid receptive surface of the carrier cylinder 26, which is further controlled at the nip between cylinders 26 and 28. Cylinder 28 is covered with a rubber blanket 34 capable of accepting the controlled amount of liquid transferred from the carrier 26 under pressure of contact. The rubber blanket 34 now carries on its surface a thin layer of solution, i.e., the controlled amount of developer necessary to develop the image. Underlying the drum 28 and in contact therewith is a metal pressure roller 36. The area of contact between the blanket 34 and the impression roller 36 constitutes a developing region 40 in which the latent image bearing surface of the exposed copy sheet 38 receives the controlled amount of developer fed to the region from said blanket 34.

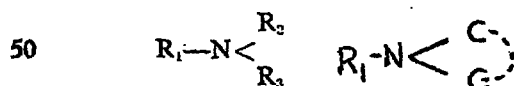
To develop the exposed copy sheet 38, it is passed through the developing region 40 where the controlled amount of developer is applied to the image bearing surface spreading the liquid evenly and uniformly over its entire surface in the range of 0.5—3.0 grams per square meter.

Other means may be employed to carry out the process such as a combination of rollers adapted to deliver an amount of fluid to a carrier and thereafter metering the fluid to the desired quantity and employing means to spread the amount over the area to be developed. It should be pointed out that the success of this process depends on feeding the desired amount of fluid to a developing region without having to remove any excess from the image surface after the initial application to the copy surface and to spread this controlled amount of liquid evenly and uniformly over the entire area of the latent image bearing surface.

Success of the practice of the invention depends in large measure on the developer composition that is employed. The developer composition must be such that it can rapidly react with the acid component in the diazo coating shifting the pH to the level where the coupling reaction can proceed. The developer should remain stable during use in the apparatus and not undergo degradation due to oxidation caused by atmospheric exposure. The background area of developed prints should not appreciably discolor upon aging so as to reduce print contrast. The image dye lines should correspond to the hues obtainable with ammonia and not be off shade. The concentration of the various ingredients in the composition should not vary due to evaporation at ordinary temperatures or the absorption of water from the atmosphere. The developer should, of course, not have an objectionable odor, be non-toxic, and simple to handle and use.

In the present invention it is critical that small amounts of the developer having the proper amine concentration be capable of being spread uniformly and evenly over the entire copy area.

Developer compositions which meet the aforescribed requirements comprise from 20%—100% by weight of active alkaline components such as an aliphatic amine, the preferred range being from 40% to 80% by weight. The alkaline component may be a compound having the general formula:



where R_1 , R_2 and R_3 are alkyl, cycloalkyl, alkylol, alkoxy or hydrogen, and

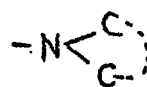
represents a heterocyclic ring for example morpholine and piperidine. The alkaline component may be diluted with a compound selected from a polyhydric alcohol, a glycol-ether, a mono-functional alcohol or water or admixtures thereof. The diluent may be present in amounts ranging from 0—80% by weight of the formulation, the preferred range being 40%—60%. The operable pH range of the developer solutions prepared in accordance with this invention is within 10 to 14.

The following amines have been eminently successful in the practice of the method: monoethanolamine; isopropanolamine; 2-(2-aminoethoxy) ethanol; triethanolamine; diethanolamine; 2-amino-2-methyl-1-propanol; N-methylethanolamine; dimethylethanolamine; hexylamine; cyclohexylamine. Other amines have demonstrated the ability to develop the diazotype print such as beta-hydroxyethyl trimethyl ammonium carbonate; N-aminoethyl-ethanolamine; alkyl substituted morpholine and piperidine; but are somewhat less desirable because they have a detectable odor or they tend to adversely affect the dye shade.

While a developer comprising 100% amine can be successfully employed in the practice of the aforescribed method, it has been found advantageous to employ a diluent to bring the concentration of amine within the preferred limits. The diluent may be a polyhydric alcohol, glycol ethers, or a mono-functional alcohol.

In general, most of the commercially available two-component papers can be used successfully with the process and developers of this invention. The quality of the final print and the rapidity with which the print develops will depend in part on the coupling component and diazonium salt that is used.

The following examples of developer solutions which may be employed in accordance with this invention are given for the purposes of illustrating preferred embodiments of the invention. It will be understood, however, that this invention is not limited to these illustrative embodiments of developer solutions useful in practicing this invention. All percentages are shown as percent by weight unless otherwise specified.



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EXAMPLE I.

A diazotype paper was prepared with the following two-component sensitizing solution:

water	600 c.c.
diethylene glycol	40 c.c.
citric acid	20 grams
zinc chloride	50 grams
isopropyl alcohol	10 c.c.
2,3-dihydroxy naphthalene, 6-sulfonic acid (sodium salt)	40 grams
p-diazo-N,N-diethylaniline zinc chloride	15 grams
thiourea	30 grams
water to make	1000 c.c.

The formulation was coated on a suitable paper such as a direct process sulphite stock, 20 lb. basis weight (17" x 22"—500), in an amount of 15 grams per amount of 15 grams per square meter.

The sensitized paper was exposed to an actinic radiation pattern such as by contact exposure through a transparent or translucent original.

A developer composition was prepared by combining the following ingredients expressed in percent by weight in a mixing vessel at room temperature and stirring until thoroughly mixed:

monoethanolamine	60%	
hexylene glycol	20%	
water	20%	20

The mixture had a pH of 12.7. It was charged into the trough 30 and the above exposed diazo paper was passed through the developing zone 40 at about 9.1 meters/min, which resulted in applying 1.1 grams of the developer solution per square meter of the latent image bearing surface.

A study of the density of the azo dye image measured at various time intervals after going through the processing steps of the present invention showed the following:

Density Units						Ammonia Developed
15 sec.	30 sec.	60 sec.	2 min.	5 min.		
1.07	1.11	1.13	1.15	1.15	1.18	

The above values represent an average of several readings taken on the image using a standard reflectance densitometer and the values are reflectance density units with the instrument calibrated against a magnesium oxide block.

The copy was immediately readable upon emerging from the last processing step. Moderate amounts of heat applied at the time of development such as by warming the developer solution or heating the emerging copy will accelerate the azo dye image formation. In the case of black line prints this may be desir-

able. However, it should be pointed out that use of heat is optional and not necessary to the successful operation of the process. The copy was dry to the touch, measuring less than 10% in moisture content; it could be handled, written upon with ink or pencil, folded or filed, as required. An examination of the copy after it was aged for a period of several weeks showed no undue discoloration of the background, that is, it was comparable in all respects to diazo prints processed by known conventional developing techniques.

EXAMPLE II.

A developer of the following formulation was prepared:

5	monoethanolamine	60%
	water	40%

An exposed two-component diazotype material, such as described in Example I, was processed in accordance with the flow diagram of Figure 1. A measurement of the amount of developer applied to the image revealed that from 1.1 to 1.2 grams of developer were applied per square meter of latent image bearing surface. A visible readable azo dye image was immediately observed as the copy sheet emerged from the last processing step which, after 15 seconds, had a density of about 1.13 units measured on a standard reflectance densitometer.

The copy was dry and ready for use.

EXAMPLE III.

The developer in this example comprised 100% monoethanolamine. A two-component diazotype copy paper of the type described in Example I was exposed. Development was accomplished by the method depicted in Figure 1. The print developed in about 8 to 10 seconds showing a pickup of about 0.70 to 0.86 grams per square meter of imaged surface. It had a density of 1.0.

The following examples represent various developing formulations used in the process of this invention in addition to those described in the previous examples. In all cases the developer formulations produced diazo prints of the quality described in the previous examples.

EXAMPLE IV.

40	2-(2-aminoethoxy) ethanol	40%
	hexylene glycol	25%
	diethyleneglycol	10%
	water	25%
	pH 12.3	

EXAMPLE V.

45	2-(2-aminoethoxy)ethanol	50%
	water	50%
	pH 12.5	

EXAMPLE VI.

	2-(2-aminoethoxy)ethanol	100%
	pH 13.2	

EXAMPLE VII.

50	diethanolamine	40%
	diethyleneglycol	30%
	4-methoxy-4 methyl-pentanol-2	20%
	water	10%
	pH 11.7	

EXAMPLE VIII.

	morpholine	5%
	monoethanolamine	10%
	dimethyl ethanolamine	5%
	Pentoxol solvent	20%
	Cellosolve (a Registered Trade Mark)	10%
	hexylene glycol	25%
	water	25%
	pH 11.9	

EXAMPLE IX.

	monoethanolamine	50%
	N-methylethanolamine	10%
	triethanolamine	10%
	methyl Cellosolve	10%
	water	20%
	pH 12.2	

EXAMPLE X.

	monoethanolamine	40%
	ethylene glycol	30%
	glycerine	10%
	water	20%
	pH 12.4	

EXAMPLE XI.

	monoethanolamine	40%
	tetraethylene glycol	30%
	water	30%
	pH 12.8	

EXAMPLE XII.

	diethylene triamine	50%
	hexylene glycol	25%
	water	25%
	pH 12.9	

EXAMPLE XIII.

	hexylamine	50%
	water	50%
	pH 12.6	

EXAMPLE XIV.

	cyclohexylamine	50%
	diethylene glycol	25%
	water	25%
	pH 13.1	

In all the foregoing examples the developers were extremely stable. While the developers did experience some evaporation under low humidity conditions, it was found that their consistency remained essentially unchanged. No appreciable change was noted in their effectiveness as alkaline compounds when exposed to air for long periods of time under conditions of use as depicted in Figure 2 of the drawings. The materials were found safe to handle and could be stored in containers for long periods of time without any adverse effect.

(4)

In the foregoing discussion there have been presented in detail the method useful in developing two-component diazotype materials which are intended to be exemplary without limiting the method contemplated by the present invention as defined in the claims.

In our copending application No. 1076/68 (Serial No. 1,143,703) which has been divided from this present application there are described and claimed developer compositions for use in the method described herein.

WHAT WE CLAIM IS:—

1. A method for making a reproduction of an original on a two-component diazotype copying material characterized by

- a) exposing the copying material to an actinic radiation pattern corresponding to the light and dark areas on said original producing a latent image thereon,

- b) feeding a controlled amount of an alkaline liquid medium by means of a carrier body to a copy developing region,

- c) passing said copy material through said region transferring an amount of said liquid not exceeding 3 grams per square meter to said latent image bearing surface, said amount of liquid being evenly and uniformly spread over said area, and forming an azo dye image on said surface.

2. The method as claimed in claim 1 in which said copy emerges from said developing region in a dry condition ready for use.

3. The method as claimed in claim 1 or 2, in which said liquid consists essentially of an aliphatic amine.

4. The method as claimed in claim 3 in which said liquid comprises from 20% to 100% by weight of an aliphatic amine.

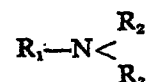
5. The method as claimed in any of the preceding claims wherein the amount of said developing liquid applied to said latent image surface is in the range of from 0.5 grams to 2.0 grams per square meter of said surface.

6. The method as claimed in claim 1 in

which said copy is maintained under pressure while in said developing region.

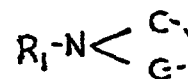
7. The method as claimed in claim 6 wherein said alkaline liquid medium contains an organic amine which is monoethanolamine, diethanolamine, 2-(2-amino ethoxy)ethanol, monoisopropanolamine, methylethanolamine, dimethylethanolamine or triethanolamine.

8. A method as claimed in claim 1 or 2, in which the alkaline liquid medium comprises a component of the general formula:

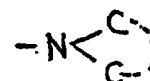


in which R_1 , R_2 and R_3 are selected from alkyl, cycloalkyl, alkylol, alkoxy and hydrogen.

9. A method as claimed in claim 1 or 2 in which the alkaline liquid medium comprises a component of the general formula:



in which R_1 is as herein defined and



is a heterocyclic ring.

10. A method as claimed in claim 9 in which the ring is morpholine or piperidine.

11. A method for making a reproduction substantially as herein described with reference to the accompanying drawing.

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In our copending application No. 1076/68 (Serial No. 1,143,703) which has been divided from this present application there are described and claimed developer compositions for use in the method described herein.

WHAT WE CLAIM IS:—

1. A method for making a reproduction of an original on a two-component diazotype copying material characterized by

a) exposing the copying material to an actinic radiation pattern corresponding to the light and dark areas on said original producing a latent image thereon,

b) feeding a controlled amount of an alkaline liquid medium by means of a carrier body to a copy developing region,

c) passing said copy material through said region transferring an amount of said liquid not exceeding 3 grams per square meter to said latent image bearing surface, said amount of liquid being evenly and uniformly spread over said area, and forming an azo dye image on said surface.

2. The method as claimed in claim 1 in which said copy emerges from said developing region in a dry condition ready for use.

3. The method as claimed in claim 1 or 2, in which said liquid consists essentially of an aliphatic amine.

4. The method as claimed in claim 3 in which said liquid comprises from 20% to 100% by weight of an aliphatic amine.

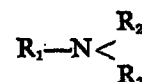
5. The method as claimed in any of the preceding claims wherein the amount of said developing liquid applied to said latent image surface is in the range of from 0.5 grams to 2.0 grams per square meter of said surface.

6. The method as claimed in claim 1 in

which said copy is maintained under pressure while in said developing region.

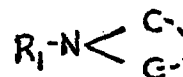
7. The method as claimed in claim 6 wherein said alkaline liquid medium contains an organic amine which is monoethanolamine, diethanolamine, 2-(2-amino ethoxy)ethanol, monoisopropanolamine, methylethanolamine, dimethylethanolamine or triethanolamine.

8. A method as claimed in claim 1 or 2, in which the alkaline liquid medium comprises a component of the general formula:

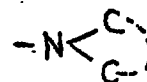


in which R_1 , R_2 and R_3 are selected from alkyl, cycloalkyl, alkylol, alkoxy and hydrogen.

9. A method as claimed in claim 1 or 2 in which the alkaline liquid medium comprises a component of the general formula:



in which R_1 is as herein defined and



is a heterocyclic ring.

10. A method as claimed in claim 9 in which the ring is morpholine or piperidine.

11. A method for making a reproduction substantially as herein described with reference to the accompanying drawing.

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