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3,703,371

PHOTOCONDUCTIVE ELEMENTS CONTAINING POLYMERIC BINDERS

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U.S. Cl. 96-1.6

16 Claims

ABSTRACT OF THE DISCLOSURE

Photoconductive elements containing a photoconductor and a binder for the photoconductor comprising a poly-(alkylidenebisaryloxyalkyl-co-alkylene isophthalate) are described. The described elements can be sensitized and charged either negatively or positively and used to prepare images electrophotographically.

This is a continuation of application Ser. No. 772,370, filed Oct. 16, 1968, and now abandoned.

This invention relates to novel electrophotographic elements having coatings of binder-containing photoconductive compositions.

Binder-containing photoconductive compositions have been widely used in the preparation of electrophotographic elements. In electrophotographic reproduction processes, these elements are utilized in the formation of latent electrostatic images. In some applications the photoconductive compositions contain an organic photoconductor and a sensitizer uniformly admixed in an inert resinous binder.

(B)

Many binders are currently used in connection with a wide variety of available organic photoconductor compounds and compositions. Typical binders are ordinary polymeric materials, e.g., phenolic resins, ketone resins, acrylic ester resins, polystyrene, etc. However, these binders usually do not impart any particular improvement in light sensitivity to the system. The light sensitivity as indicated by the electrical speed of these particular systems is ordinarily due wholly to the organic photoconductor and sensitizer. Other binders have been found to contribute significantly to the light sensitivity of the system. However, the selection of these polymers for incorporation into photoconductive compositions to form electrophotographic layers has proceeded on a compound-by-compound basis. Nothing has yet been discovered from the numerous binders tested which permits effective prediction and selection of particular polymers exhibiting the desired properties.

It is, therefore, an object of this invention to provide improved novel binder-containing photoconductive compositions which exhibit high light sensitivities.

It is another object to provide transparent electrophotographic elements having the high speed characteristic of the novel photoconductive compositions of this invention.

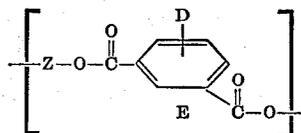
These and other objects of this invention are accomplished by a photoconductive composition which contains a photoconductor admixed with a binder thereof which is a poly-(alkylidenebisaryloxyalkyl-co-alkylene isophthalate) (wherein the alkylene moiety contains 2 to 10 carbon atoms. It has been discovered that such compositions exhibit increased light sensitivities as evidenced by greater

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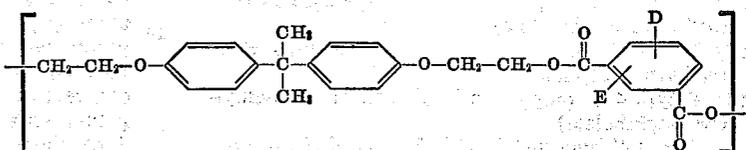
speeds. In particular, substantial increases in speeds are obtained as compared to speeds attainable with many other polymeric binder compositions. These increases in speed are observed when the coating accepts a suitable potential (e.g., 500-600 volts) and the relative speed of the coating is determined on the basis of the reciprocal of the exposure required to reduce the potential of the surface charge by 100 volts (shoulder speed) or to 100 volts (toe speed). The reduction of the surface potential to 100 volts or below is significant in that it represents a requirement for suitable broad area development of a latent image. The relative speed at 100 volts is a measure of the ability to produce and henceforth to develop or otherwise utilize the latent image. When the photoconductor is absent from the coating and only a conventional binder is used, the surface potential does not drop to or below 100 volts and therefore no speed can be assigned to such a composition. When a photoconductor is part of the coating in many conventional polymeric binders, the surface potentials of such resultant compositions usually drop below 100 volts, and thus, a definite speed can be ascertained. However, these speeds are improved when the binders of this invention are employed.

The poly(alkylidenebisaryloxyalkyl-co-alkylene isophthalate) copolyesters of this invention include those consisting essentially of the following repeating units:

(A)

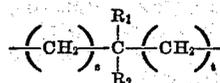


and



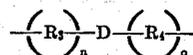
wherein Z is an alkylene group having 2 to 10 carbon atoms including substituted as well as unsubstituted alkylene radicals such as

(a) A straight chain or branched chain alkylene hydrocarbon radical, e.g., those represented by the formula:

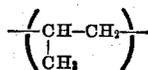


where R₁ and R₂ are either hydrogen or an alkyl radical having 1 to 5 carbon atoms, t is an integer from 1 to 7 and s is an integer from zero to 7, including a trimethylene radical, a tetramethylene radical, a pentamethylene radical, a neopentylene radical, an ethylene radical, a hexamethylene radical, a propylene radical, etc.;

(b) An oxydialkylene radical or a thiadialkylene radical, e.g., those represented by the formula



where each of R₃ and R₄ is an alkylene radical including a branched chain alkylene radical e.g., -(CH₂)₂, -(CH₂-CH₂)₂



etc. where D is an oxygen or sulfur atom and n and o are integers from 2 to 7 including an oxydiethylene radical, a thiadiethylene radical, etc.;

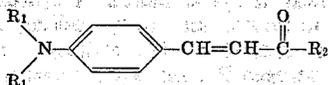
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cyclized to form a fluorene moiety, for example; the amino substituent can be represented by the formula

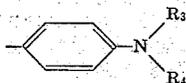


wherein each R can be an alkyl group typically having 1 to 8 carbon atoms, a hydrogen atom, an aryl group, or together the necessary atoms to form a heterocyclic amino group typically having 5 to 6 atoms in the ring such as morpholino, pyridyl, pyrrol, etc.; at least one of D, E and G preferably being a p-dialkylaminophenyl group, when J is an alkyl group, such an alkyl group more generally has 1 to 7 carbon atoms, these materials being more fully described in U.S. Pat. 3,274,000, French Pat. 1,383,461 and in U.S. Ser. No. 627,857 filed Apr. 3, 1967 by Seus and Goldman now U.S. 3,542,544.

(D) Photoconductors comprising 4-diarylamino-substituted chalcones having the formula:

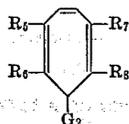
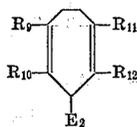


wherein R₁ and R₂ are each phenyl radicals including substituted phenyl radicals and particularly when R₂ is a phenyl radical



wherein R₃ and R₄ are each aryl radicals, aliphatic residues of 1 to 12 carbon atoms such as alkyl radicals preferably having 1 to 4 carbon atoms, or hydrogen; particularly advantageous results being obtained when R₁ is a phenyl radical including a substituted phenyl radical and where R₂ is diphenylaminophenyl, dimethylaminophenyl or phenyl, these materials being more fully described in Fox application U.S. Ser. No. 613,846 now U.S. 3,542,501.

(E) Non-ionic cycloheptenyl compounds which may be substituted with substituents such as (a) an aryl radical including substituted as well as unsubstituted aryl radicals, (b) a hydroxy radical, (c) an azido radical, (d) a heterocyclic radical having 5 to 6 atoms in the heterocyclic nucleus and at least one heteronitrogen atom, and including substituted and unsubstituted heterocyclic radicals, and (e) an oxygen linked cycloheptenyl moiety. The substitution on the cycloheptenyl nucleus occurs at an unsaturated carbon atom when the cycloheptenyl moiety is a conjugated triene with no aromatic structure fused thereto. However, if there is at least one aromatic structure fused to the cycloheptenyl moiety, then the substituents are attached to a saturated carbon atom. Additional photoconductors within this class are included in one of the following formulae:



and

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where E and G can be either:

- (a) A phenyl radical,
- (b) A naphthyl radical,
- (c) A heterocyclic radical having 5 to 6 atoms in the heterocyclic nucleus and at least one hetero nitrogen atom,
- (d) A hydroxyl radical, or
- (e) An oxygen containing radical having a structure such that the resultant cycloheptenyl compound is a symmetrical ether;

D can be any of the substituents defined for E and G above and is attached to a carbon atom in the cycloheptenyl nucleus having a double bond; (R₉ and R₁₀), (R₁₁ and R₁₂), (R₅ and R₆), and R₇ and R₈) are together the necessary atoms to complete a benzene ring fused to the cycloheptenyl nucleus; these compounds being more fully described in U.S. Ser. No. 654,091 filed July 18, 1967 now U.S. 3,533,786.

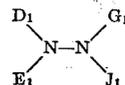
(F) Compounds containing an



nucleus including (1) unsubstituted and substituted N,N-bicarbazyls containing substituents in either or both carbazoyl nuclei such as

- (a) An alkyl radical including a substituted alkyl radical such as a haloalkyl or an alkoxyalkyl radical,
- (b) A phenyl radical including a substituted phenyl radical such as a naphthyl, an aminophenyl or a hydroxyphenyl radical,
- (c) A halogen atom,
- (d) An amino radical including substituted as well as unsubstituted amino radicals such as an alkylamino or a phenylalkylamino radical,
- (e) An alkoxy radical,
- (f) A hydroxyl radical,
- (g) A cyano radical,
- (h) A heterocyclic radical such as a pyrazolyl, a carbazoyl or a pyridyl radical;

(2) tetra-substituted hydrazines containing substituents which are substituted or unsubstituted phenyl radicals, or heterocyclic radicals having 5 to 6 atoms in the hetero nucleus, suitable results being obtained when all four substituents are not unsubstituted phenyl radicals, i.e., if at least one substituent is a substituted phenyl radical or a heterocyclic radical having 5 to 6 atoms in the heteronucleus. Other tetra-substituted hydrazines include those having the following formula:



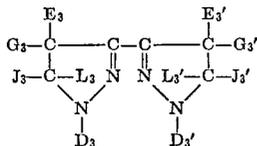
where D₁, E₁, G₁ and J₁ are each either

- (a) A substituted phenyl radical such as a naphthyl radical, an alkylphenyl radical, a halophenyl radical, a hydroxyphenyl radical, a haloalkylphenyl radical or a hydroxyalkylphenyl radical or
- (b) A heterocyclic radical such as an imidazolyl radical, a furyl radical or a pyrazoline radical. In addition, J₁ and E₁ can also be
- (c) An unsubstituted phenyl radical. Especially preferred are those tetra-substituted hydrazines wherein both D₁ and G₁ are either substituted phenyl radicals or heterocyclic radicals. These compounds are more fully described in U.S. Ser. No. 673,962 filed Oct. 9, 1967 now U.S. 3,542,546.

(G) Organic compounds having a 3,3'-bis-aryl-2-pyrazoline nucleus which is substituted in either five-member ring with the same or different substituents. The 1 and 5 positions on both pyrazoline rings can be substituted by an aryl moiety including unsubstituted as well as substituted aryl substituents such as alkoxyaryl, alkaryl, alkaminoaryl, carboxyaryl, hydroxyaryl and haloaryl. The 4 position can contain hydrogen or unsubstituted as well as substituted alkyl and aryl radicals such as alkoxyaryl, alkaryl, alkaminoaryl, haloaryl, hydroxy-

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aryl, alkoxyalkyl, aminoalkyl, carboxyaryl, hydroxyalkyl and haloalkyl. Other photoconductors in this class are represented by the following structure:

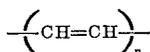


wherein:

D₃, D₃', J₃ and J₃' can be either a phenyl radical including a substituted phenyl radical such as a tolyl radical or a naphthyl radical including a substituted naphthyl radical,

E₃, E₃', G₃, G₃', L₃ and L₃' can be any of the substituents set forth above and in addition can be either a hydrogen atom or an alkyl radical containing 1-8 carbon atoms. These organic photoconductors are more fully described in U.S. Ser. No. 664,642 filed Aug. 31, 1967 now U.S. 3,527,602.

(H) Triarylamines in which at least one of the aryl radicals is substituted by either a vinyl radical or a vinylene radical having at least one active hydrogen-containing group. The phrase "vinylene radical" includes substituted as well as unsubstituted vinylene radicals and also includes those radicals having at least one and as many as three repeating units of vinylene groups such as



wherein *n* is an integer of from one to three. Groups which contain active hydrogen are well known in the art, the definition of this term being set forth in several text books such as "Advanced Organic Chemistry," R. C. Fuson, pp. 154-157, John Wiley & Sons, 1950. The term "active hydrogen-containing group" as used herein includes those compounds encompassed by the discussion in the textbook cited above and in addition includes those compounds which contain groups which are hydrolyzable to active hydrogen-containing groups. Typical active hydrogen-containing groups substituted on the vinylene radical of the triarylamines include:

- (a) Carboxy radicals,
- (b) Hydroxy radicals,
- (c) Ethynyl radicals,
- (d) Ester radicals (e.g.,



wherein R₁₃ is alkyl or aryl) including cyclic ester radicals (e.g.,



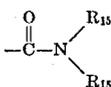
wherein R₁₄ is a cyclic alkylene radical connected to a vinylene combination such as is found in coumarin derivatives),

- (e) Carboxylic acid anhydride radicals,
- (f) Semicarbazono radicals,
- (g) Cyano radicals,
- (h) Acyl halide radicals (e.g.,



etc.), and

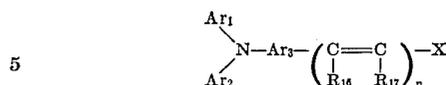
- (i) Amido radicals (e.g.,



wherein R₁₅ is a hydrogen atom, an alkyl group or an aryl group). Other active hydrogen-containing groups include substituted and unsubstituted alkylidene oximido

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radicals. Photoconductors included in this class can be represented by the following structure:



wherein:

(a) Ar₁ and Ar₂ are each a phenyl radical including a substituted phenyl radical such as a halophenyl radical, an alkyl phenyl radical or an aminophenyl radical;

(b) Ar₃ is an arylene radical including a substituted arylene radical such as a phenylene radical or a naphthylene radical,

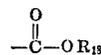
(c) R₁₆ and R₁₇ are each hydrogen, a phenyl radical including a substituted phenyl radical or a lower alkyl radical preferably having 1-8 carbon atoms;

(d) X is either (1) an active hydrogen-containing group such as a carboxy radical, an acyl halide radical, an amido radical, a carboxylic acid anhydride radical, an ester radical, a cyano radical, a hydroxy radical, a semicarbazono radical, an ethynyl radical, or a methylidyne oximido radical, or (2) hydrogen, provided that when X is hydrogen R₁₆ and R₁₇ are also hydrogen; and (e) *n* is an integer of one to three.

The arylene nucleus can be substituted in any position by the vinyl or vinylene moiety. However, when Ar₃ is phenylene, particularly good results are obtained if the substitution occurs in the para position. These materials are more fully described in U.S. Ser. No. 706,800 filed Feb. 20, 1968 now U.S. 3,567,450.

(I) Triarylamines in which at least one of the aryl radicals is substituted by an active hydrogen-containing group. The term "active hydrogen-containing group" has the same meaning as set forth above and again includes those compounds encompassed by the discussion in the textbook and additionally includes those compounds which contain groups which are hydrolyzable to active hydrogen-containing groups. Typical active hydrogen-containing groups which are substituted on an aryl radical of the triarylamines include:

- (a) Carboxy radicals;
- (b) Hydroxy radicals;
- (c) Ethynyl radicals;
- (d) Ester radicals (e.g.,



wherein R₁₈ is an alkyl or an aryl group);

(e) Lower alkylene hydroxy radicals (e.g., having 1-8 carbon atoms);

(f) Carboxylic acid anhydride radicals;

(g) Lower alkylene carboxy radicals (e.g., having 2-8 carbon atoms);

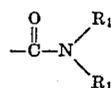
(h) Cyano radicals;

(i) Acyl halide radicals (e.g.,



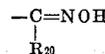
etc.);

(j) Amido radicals (e.g.,



wherein R₁₉ is a hydrogen atom, an alkyl group or an aryl group);

(k) Lower alkylidene oximido radicals having 1-8 carbon atoms including substituted alkylidene oximido radicals (e.g.,

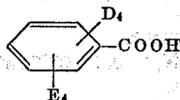


wherein R₂₀ is hydrogen or a lower alkyl radical);

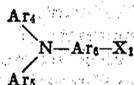
- (1) Semicarbazono radicals; and

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(m) Arylene carboxy radicals including substituted arylene carboxy radicals (e.g.,



wherein D_4 and E_4 are phenyl or lower alkyl radicals. Photoconductors included in this class can be represented by the following structure:



wherein:

(a) Ar_4 and Ar_5 are each a phenyl radical including a substituted phenyl radical such as a halophenyl radical, an alkyl phenyl radical or an amino phenyl radical;

(b) Ar_6 is an arylene radical including a substituted arylene radical such as a phenylene radical or a naphthylene radical; and

(c) X_1 is an active hydrogen-containing group such as a carboxy radical, an acyl halide radical, an amido radical, a carboxylic acid anhydride radical, an ester radical, a cyano radical, a semicarbazono radical, a hydroxy radical, an ethynyl radical, a methylidyne oximido radical or a phenylene carboxy radical. These materials are more fully described in U.S. Ser. No. 706,780 filed Feb. 20, 1968.

(J) Organo-metallic compounds having at least one amino-aryl substituent attached to a Group IVa or Group Va metal atom. The metallic substituents of this class of organic photoconductors are Group IVa or Group Va metals in accordance with the Periodic Table of the Elements (Handbook of Chemistry and Physics, 38th edition, pp. 394-95) and include silicon, germanium, tin and lead from Group IVa and phosphorus, arsenic, antimony and bismuth from Group Va. These materials can be substituted in the metallo nucleus with a wide variety of substituents but at least one of the substituents must be an amino-aryl radical. The amino radical can be positioned anywhere on the aromatic nucleus, but best results are obtained if the aryl moiety is a phenyl radical having the amino group in the 4 or para position. Typical substituents attached to the metal nucleus include the following:

(a) A hydrogen, sulfur or oxygen atom,

(b) An alkyl radical,

(c) An aryl radical including unsubstituted as well as substituted aryl radicals such as aminoaryl, alkylaryl and haloaryl,

(d) An oxygen-containing radical such as an alkoxy or aryloxy radical,

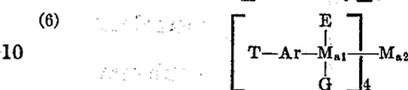
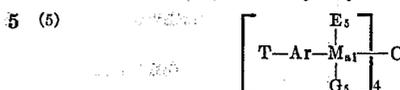
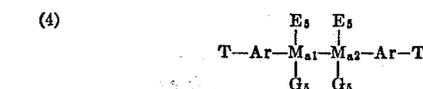
(e) An amino radical including unsubstituted and substituted amino radicals such as mono- and diarylamino and mono- and dialkylamino radicals,

(f) A heterocyclic radical and

(g) A Group IVa or Va organo metallic radical. Photoconductors included in this class can be represented by the following structures:



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where E_5 , G_5 , L_5 and Q_5 can be

(a) A hydrogen atom,

(b) An aryl radical including unsubstituted as well as substituted aryl radicals such as a phenyl radical, a naphthyl radical, a dialkylaminophenyl radical, or a diarylaminophenyl radical,

(c) An alkyl radical having 1 to 8 carbon atoms,

(d) An alkoxy radical having 1 to 8 carbon atoms,

(e) An aryloxy radical such as a phenoxy radical,

(f) An amino radical having the formula



wherein R_{21} and R_{22} can be hydrogen atoms or alkyl radicals having 1 to 8 carbon atoms, or

(g) A heterocyclic radical having 5 to 6 atoms in the hetero nucleus including at least one nitrogen atom such as a triazolyl, a pyridyl radical, etc.;

T is an amino radical such as an alkylamino radical having 1 to 8 carbon atoms or an arylamino radical such as a phenylamino radical;

Ar is an aromatic radical such as phenyl or naphthyl;

M_{a1} and M_{a2} are the same or different Group IVa metals;

M_b is a Group Va metal;

D_5 can be any of the substituents set forth above for E_5 , G_5 , L_5 and Q_5 and in addition can be a Group IVa organo-metallic radical or when taken with E, an oxygen atom or a sulfur atom;

J_5 can be any of the substituents set forth above for E_5 , G_5 , L_5 and Q_5 and in addition can be when taken with E, an oxygen atom or a sulfur atom. These materials are described in U.S. Ser. No. 650,664 filed July 13, 1967.

(K) Any other organic compound which exhibits photoconductive properties such as those set forth in Australian Pat. 248,402.

Representative organic photoconductors useful in this invention include the compounds listed below:

TABLE I

	diphenylamine
55	dinaphthylamine
	N,N'-diphenylbenzidine
	N-phenyl-1-naphthylamine
	N-phenyl-2-naphthylamine
	N,N'-diphenyl-p-phenylenediamine
60	2-carboxy-5-chloro-4'-methoxydiphenylamine
	p-anilinophenol
	N,N'-di-2-naphthyl-p-phenylenediamine
	4,4'-benzylidene-bis-(N,N-dimethyl-m-toluidine)
	triphenylamine
65	N,N,N',N'-tetraphenyl-m-phenylenediamine
	4-acetyltriphenylamine
	4-hexanoyltriphenylamine
	4-lauroyltriphenylamine
70	4-hexyltriphenylamine
	4-dodecyltriphenylamine
	4,4'-bis(diphenylamino)benzil
	4,4'-bis(diphenylamino)benzophenone
	poly[N,4''-(N,N',N'-triphenylbenzidine)]
	polyadipyltriphenylamine
75	polysebacyltriphenylamine

polydecamethylenetriphenylamine
 poly-N-(4-vinylphenyl)diphenylamine
 poly-N-(vinylphenyl)- α,α' -dinaphthylamine
 4,4'-benzylidene-bis(N,N-diethyl-m-toluidine)
 4',4''-diamino-4-dimethylamino-2',2''-dimethyltri-
 phenylmethane
 4',4''-bis(diethylamino)-2,6-dichloro-2',2''-dimethyl-
 triphenylmethane
 4',4''-bis(diethylamino)-2',2''-dimethyldiphenylnaph-
 thylmethane
 2',2''-dimethyl-4,4',4''-tris(dimethylamino)triphenyl-
 methane
 4',4''-bis(diethylamino)-4-dimethylamino-2',2''-
 dimethyltriphenylmethane
 4',4''-bis(diethylamino)-2-chloro-2',2''-dimethyl-
 4-dimethylaminotriphenylmethane
 4',4''-bis(diethylamino)-4-dimethylamino-2,2',2''-
 trimethyltriphenylmethane
 4',4''-bis(dimethylamino)-2-chloro-2',2''-dimethyl-
 triphenylmethane
 4',4''-bis(dimethylamino)-2',2''-dimethyl-4-methoxy-
 triphenylmethane
 bis(4-diethylamino)-1,1,1-triphenylethane
 bis(4-diethylamino)tetraphenylmethane
 4',4''-bis(benzylethylamino)-2',2''-dimethyltriphenyl-
 methane
 4',4''-bis(diethylamino)-2,2''-diethoxytriphenylmethane
 4,4'-bis(dimethylamino)-1,1,1-triphenylethane
 1-(4-N,N-dimethylaminophenyl)-1,1-diphenylethane
 4-dimethylaminotetraphenylmethane
 4-diethylaminotetraphenylmethane
 4,4'-bis(diphenylamino)chalcone
 4-diphenylamino-4'-dimethylaminochalcone
 4-dimethylamino-4'-diphenylaminochalcone
 4,4'-bis(dimethylamino)chalcone
 4,4'-bis(diethylamino)chalcone
 4-diethylamine-4'-diphenylaminochalcone
 4,4'-bis(n-amyloxy)chalcone
 4,4'-bis(nitro)chalcone
 4-diphenylaminochalcone
 4-dimethylaminochalcone
 4'-diphenylaminochalcone
 4'-dimethylaminochalcone
 bis-[5-(5H-dibenzo[a,d]cycloheptenyl)]ether
 5-hydroxy-5H-dibenzo[a,d]cycloheptene
 1-{5-(5H-dibenzo[a,d]cycloheptenyl)}-4,5-dicarbo-
 methoxy-1,2,3-triazole
 1-{5-(5H-dibenzo[a,d]cycloheptenyl)}-4,5-dibenzoyl-
 1,2,3-triazole
 5-azido-5H-dibenzo[a,d]cycloheptene
 1-{5-(10,11-dihydro-5H-dibenzo[a,d]cycloheptenyl)}-
 4,5-dicarbomethoxy-1,2,3-triazole
 1-{5-(10,11-dihydro-5H-dibenzo[a,d]cycloheptenyl)}-
 4,5-dibenzoyl-1,2,3-triazole
 4-[5-(5H-dibenzo[a,d]cycloheptenyl)]-N,N-dimethyl
 aniline
 N,N-diethyl-3-methyl-4-[5-(5H-dibenzo[a,d]cyclo-
 heptenyl)]aniline
 4-[5-(5H-dibenzo[a,d]cycloheptenyl)]-1-dimethyl-
 aminonaphthalene
 N,N-diethyl-3-methyl-4-[5-(10,11-dihydro-5H-dibenzo
 [a,d]cycloheptenyl)]aniline
 3-(4-dimethylaminophenyl)-1,3,5-cycloheptatriene
 3-(4-diethylamino-2-methylphenyl)-1,3,5-cyclo-
 heptatriene
 3-(4-dimethylaminonaphthyl)-1,3,5-cycloheptatriene
 N,N-diethyl-3-methyl-4-[5-(5H-dibenzo[a,d]cyclo-
 heptenyl)]aniline
 tetra- α -naphthylhydrazine
 tetra(3-methyl-4-hydroxyphenyl)hydrazine
 tetra(m-hydroxyethylphenyl)hydrazine
 tetra(2-methyl-5-chloroethylphenyl)hydrazine
 tetra(2-methyl-5-hydroxyphenyl)hydrazine
 tetra(1-imidazolyl)hydrazine

N,N-di- α -naphthyl-N',N'-di(3-methyl-4-hydroxy-
 phenyl)hydrazine
 N-3-furyl-N-(2-methyl-4-hydroxyphenyl)-N',N'-di- β -
 naphthylhydrazine
 5 tetra- β -naphthylhydrazine
 N,N'-di- β -naphthyl-N,N'-diphenylhydrazine
 tetra-4-tolylhydrazine
 N,N'-diphenyl-N,N'-di(3-methyl-4-hydroxyphenyl)
 hydrazine
 10 N,N'-diphenyl-N,N'-di-p-chlorophenyl hydrazine
 phenyltri-(2-methyl-5-hydroxyphenyl)hydrazine
 N,N'-bicarbazylyl
 cyclo-tetrakis(3,9-carbazolylene)
 6-(3-carbazolyl)-cyclo-tetrakis(3,9-carbazolylene)
 15 6-(9-carbazolyl)-cyclo-tetrakis(3,9-carbazolylene)
 3,3'-bis(3-carbazolyl)-9,9'-bicarbazolyl
 3-(3-carbazolyl)-9-(9-carbazolyl)carbazole
 3-(9-carbazolyl)-9,9'-bicarbazolyl
 3,3'-diethyl-9,9'-bicarbazolyl
 20 3,3'-diphenyl-9,9'-bicarbazolyl
 3,3'-dichloro-9,9'-bicarbazolyl
 4,4'-bis(diethylamino)-9,9'-bicarbazolyl
 3,3'-diethoxy-9,9'-bicarbazolyl
 1,1'-dihydroxy-9,9'-bicarbazolyl
 25 2,2'-dicyano-9,9'-bicarbazolyl
 tetra(p-diethylaminophenyl)hydrazine
 3,3'-bis(1,5-diphenyl-2-pyrazoline)
 3,3'-bis(1-p-tolyl-5-phenyl-2-pyrazoline)
 3,3'-bis(1,5-[1-naphthyl]-2-pyrazoline)
 30 1,5-diphenyl-3-[3'-(1'-p-tolyl-5'-phenyl)-2'-pyrazolyl]-
 2-pyrazoline
 3,3'-bis(1,5-diphenyl-4,5-dimethyl-2-pyrazoline)
 3,3'-bis(1,4,5-triphenyl-2-pyrazoline)
 3,3'-bis(1,5-di-p-tolyl-4-methoxy-2-pyrazoline)
 35 3,3'-bis(1,5-diphenyl-4-dimethylamino-2-pyrazoline)
 3,3'-bis[1,5-diphenyl-4-(p-chlorophenyl)-2-
 pyrazoline]
 3,3'-bis[1,5-diphenyl-4,5-di-(p-diethylaminophenyl)-2-
 pyrazoline]
 40 3,3'-bis[1,5-diphenyl-4-(p-methoxyphenyl)-5-
 ethyl-2-pyrazoline]
 3,3'-bis(1,5-diphenyl-4-chloromethyl-2-pyrazoline)
 1,5-diphenyl-4,5-dimethyl-3-[3'-(1'-p-tolyl-4'-diethyl-
 5',5'-methylphenyl)-2'-pyrazolyl]-2-pyrazoline
 45 4-(p-diphenylaminophenyl)-3-buten-1-yne
 p-diphenylaminostyrene
 ethyl p-diphenylaminocinnamate
 methyl p-diphenylaminocinnamate
 p-diphenylaminocinnamoyl chloride
 50 p-diphenylaminocinnamic acid N,N-diphenylamide
 p-diphenylaminocinnamic acid anhydride
 3-(p-diphenylaminophenyl)-2-butenic acid
 bis(p-diphenylaminobenzal) succinic acid
 4-N,N-bis(p-bromophenyl)aminocinnamic acid
 55 1-(4-diphenylamino)naphthacrylic acid
 p-diphenylaminocinnamic acid
 p-diphenylaminocinnamionitrile
 7-diphenylamino coumarin
 60 p-diphenylaminophenylvinylacrylic acid
 p-diphenylaminobenzyl p'-diphenylaminocinnamate
 7-(p-diphenylaminostyryl) coumarin
 p-diphenylaminocinnamyl alcohol
 4-diphenylaminocinnamaldehyde semicarbazone
 65 O-p-diphenylaminocinnamoyl p'-diphenylaminobenzalde-
 hyde oxime
 p-diphenylaminocinnamaldehyde oxime
 1,3-bis(p-diphenylaminophenyl)-2-propen-1-ol
 methyl p-diphenylaminobenzoate
 70 N,N-diphenylanthranilic acid
 3-p-diphenylaminophenyl-1-propanol
 4-acetyltriphenylamine semicarbazone
 ethyl 2,6-diphenyl-4-(p-diphenylaminophenyl)benzoate
 1-(p-diphenylaminophenyl)-1-hydroxy-3-butene
 75 4-hydroxymethyltriphenylamine

1-(p-diphenylaminophenyl)ethanol
 4-hydroxytriphenylamine
 2-hydroxytriphenylamine
 4-formyltriphenylamine oxime
 4-acetyltriphenylamine oxime
 1-(p-diphenylaminophenyl)hexanol
 1-(p-diphenylaminophenyl)dodecanol
 p-diphenylaminobenzoic acid anhydride
 4-cyanotriphenylamine
 p-diphenylaminobenzoic acid N,N-diphenylamide
 p-diphenylaminobenzoic acid
 p-diphenylaminobenzoyl chloride
 3-p-diphenylaminophenylpropionic acid
 4-formyltriphenylamine semicarbazone
 triphenyl-p-diethylaminophenylsilane
 triphenyl-p-diethylaminophenylsilane
 triphenyl-p-diethylaminophenylgermane
 triphenyl-p-dimethylaminophenylstannane
 triphenyl-p-diethylaminophenylstannane
 diphenyl-di-(p-diethylaminophenyl)stannane
 triphenyl-p-diethylaminophenylplumbane
 tetra-p-diethylaminophenylplumbane
 phenyl-di-(p-diethylaminophenyl)phosphine
 bis-(p-diethylaminophenyl)phosphine oxide
 tri-p-dimethylaminophenylarsine
 tri-p-diethylaminophenylarsine
 2-methyl-4-dimethylaminophenylarsine oxide
 tri-p-diethylaminophenylbismuthine
 methyl-di-(p-diethylaminophenyl)arsine
 methyl-di-(p-diethylaminophenyl)phosphine
 phenyl-tri-(p-diethylaminophenyl)stannane
 methyl-tri-(p-diethylaminophenyl)stannane
 tetra-p-diethylaminophenylgermane
 diphenyl-p-diethylaminophenylsilane
 p-diethylaminophenylarsine
 tetrakis-[diphenyl-(p-diethylaminophenyl)plumbyl]
 methane
 tetrakis-[diphenyl-(p-diethylaminophenyl)stannyl]
 stannane
 bis-[phenyl-(p-diethylaminophenyl)]dibismuthine
 tri-(p-diethylaminophenyl)phosphine sulfide
 di-(p-diethylaminophenyl)thioxotin

The photoconductive layers of the invention can also be sensitized by the addition of effective amounts of sensitizing compounds to exhibit improved electrophotosensitivity. Sensitizing compounds useful with the photoconductive compounds of the present invention can be selected from a wide variety of materials, including such materials as pyrylium including thiapyrylium and selenapyrylium dye salts disclosed in Van Allan et al., U.S. Pat. 3,250,615; fluorenes, such as

7,12-dioxo-13-dibenzo(a,h)fluorene,
 5,10-odioxo-4a,11-diazabenzob(b)fluorene,
 3,13-dioxo-7-oxadibenzo(b,g)fluorene,

and the like; aromatic nitro compounds of the kinds described in U.S. Pat. 2,610,120; anthrones like those disclosed in U.S. Pat. 2,670,284; quinones, U.S. Pat. 2,670,286; benzophenones U.S. Pat. 2,670,287; thiazoles U.S. Pat. 2,732,301; mineral acids; carboxylic acids, such as maleic acid; dichloroacetic acid, and salicylic acid; sulfonic and phosphoric acids; and various dyes, such as cyanine (including carbocyanine), merocyanine, diarylmethane, thiazine, azine, oxazine, xanthene, phthalein, acridine, azo, anthraquinone dyes and the like and mixtures thereof. The sensitizers preferred for use with the compounds of this invention are selected from pyrylium including selenapyrylium and thiapyrylium salts, and cyanine including carbocyanine dyes.

Where a sensitizing compound is employed with the binder and organic photoconductor to form a sensitized electrophotographic element, it is the normal practice to mix a suitable amount of the sensitizing compound with the coating composition so that, after thorough mixing, the sensitizing compound is uniformly distributed in the

coated element. Other methods of incorporating the sensitizer or the effect of the sensitizer may, however, be employed consistent with the practice of this invention. In preparing the photoconductive layers, no sensitizing compound is required to give photoconductivity in the layers which contain the photoconducting substance therefore, no sensitizer is required in a particular photoconductive layer. However, since relatively minor amounts of sensitizing compound give substantial improvement in speed in such layers, the sensitizer is preferred. The amount of sensitizer that can be added to a photoconductor-incorporating layer to give effective increases in speed can vary widely. The optimum concentration in any given case will vary with the specific photoconductor and sensitizing compound used. In general, substantial speed gains can be obtained where an appropriate sensitizer is added in a concentration range from about 0.0001 to about 30 percent by weight based on the weight of the film-forming coating composition. Normally, a sensitizer is added to the coating composition in an amount by weight from about 0.005 to about 5.0 percent by weight of the total coating composition.

Solvents useful for preparing coating compositions with the binders of the present invention can include a wide variety of organic solvents for the components of the coating composition. For example, benzene; toluene; acetone; 2-butanone; chlorinated hydrocarbons such as methylene chloride; ethylene chloride; and the like; ethers, such as tetrahydrofuran and the like, or mixtures of such solvents can advantageously be employed in the practice of this invention.

In preparing the coating compositions utilizing the binders disclosed herein useful results are obtained where the photoconductive substance is present in an amount equal to at least about 1 weight percent of the coating composition. The upper limit in the amount of photoconductive material present can be widely varied in accordance with usual practice. It is normally required that the photoconductive material be present in an amount ranging from about 1 weight percent of the coating composition to about 99 weight percent of the coating composition. A preferred weight range for the photoconductive material in the coating composition is from about 10 weight percent to about 60 weight percent.

Coating thicknesses of the photoconductive composition on a support can vary widely. Normally, a wet coating thickness in the range of about 0.001 inch to about 0.01 inch is useful in the practice of the invention. A preferred range of coating thickness is from about 0.002 inch to about 0.006 inch before drying although such thicknesses can vary widely depending on the particular application desired for the electrophotographic element.

Suitable supporting materials for coating the photoconductive layers of the present invention can include any of the electrically conducting supports, for example, paper (at a relative humidity above 20 percent); aluminum-paper lamintes; metal foils, such as aluminum foil, zinc foil, etc.; metal plates, such as aluminum, copper, zinc, brass, and galvanized plates; vapor deposited metal layers such as silver, nickel or aluminum on conventional film supports such as cellulose acetate, poly(ethylene terephthalate), polystyrene and the like conducting supports.

An especially useful conducting support can be prepared by coating a film support material such as poly(ethylene terephthalate) with a layer containing a semiconductor dispersed in a resin. A suitable conducting coating can be prepared from the sodium salt of a carboxy-ester lactone of maleic anhydride and a vinyl acetate polymer, cuprous iodide and the like. Such conducting layers and methods for their optimum preparation and use are disclosed in U.S. 3,007,901, 3,245,833 and 3,267,807.

The compositions of the present invention can be employed in photoconductive elements useful in any of

the well known electrophotographic processes which require photoconductive layers. One such process is the xerographic process. In a process of this type, an electrophotographic element held in the dark, is given a blanket electrostatic charge by placing it under a corona discharge to give a uniform charge to the surface of the photoconductive layer. This charge is retained by the layer owing to the substantial dark insulating property of the layer, i.e., the low conductivity of the layer in the dark. The electrostatic charge formed on the surface of the photoconductive layer is then selectively dissipated from the surface of the layer by imagewise exposure to light by means of a conventional exposure operation such as for example, by a contact-printing technique, or by lens projection of an image, or reflex or bireflex techniques and the like, to thereby form a latent electrostatic image in the photoconductive layer. Exposing the surface in this manner forms a pattern of electrostatic charge by virtue of the fact that light energy striking the photoconductor causes the electrostatic charge in the light struck areas to be conducted away from the surface in proportion to the intensity of the illumination in a particular area.

The charge pattern produced by exposure is then developed or transferred to another surface and developed there, i.e., either the charge or uncharged areas rendered visible, by treatment with a medium comprising electrostatically responsive particles having optical density. The developing electrostatically responsive particles can be in the form of a dust, or powder and generally comprise a pigment in a resinous carrier called a toner. A preferred method of applying such a toner to a latent electrostatic image for solid area development is by the use of a magnetic brush. Methods of forming and using a magnetic brush toner applicator are described in the following U.S. patents: 2,786,439; 2,786,440; 2,786,441; 2,811,465; 2,874,063; 2,984,163; 3,040,704; 3,117,884; and reissue Re. 25,779. Liquid development of the latent electrostatic image may also be used. In liquid development the developing particles are carried to the image-bearing surface in an electrically insulating liquid carrier. Methods of development of this type are widely known and have been described in the patent literature, for example, U.S. Pat. 2,297,691 and in Australian Pat. 212,315. In dry developing processes the most widely used method of obtaining a permanent record is achieved by selecting a developing particle which has as one of its components a low-melting resin. Heating the powder image then causes the resin to melt or fuse into or on the element. The powder is, therefore, caused to adhere permanently to the surface of the photoconductive layer. In other cases, a transfer of the charge image or powder image formed on the photoconductive layer can be made to a second support such as paper which would then become the final print after developing and fusing or fusing respectively. Techniques of the type indicated are well known in the art and have been described in a number of U.S. and foreign patents, such as U.S. Pats. 2,297,691 and 2,551,582, and in "RCA Review," vol. 15 (1954), pages 469-484.

The compositions of the present invention can be used in electrophotographic elements having many structural variations. For example, the photoconductive composition can be coated in the form of single layers or multiple layers on a suitable opaque or transparent conducting support. Likewise, the layers can be contiguous or spaced having layers of insulating material or other photoconductive material between layers or overcoated or interposed between the photoconductive layer or sensitizing layer and the conducting layer. It is also possible to adjust the position of the support and the conducting layer placing a photoconductor layer over a support and coating the exposed face of the support or the exposed or overcoated face of the photoconductor with a conducting layer. Configurations differing from those contained in the ex-

amples can be useful or even preferred for the same or different application for the electrophotographic element.

The following examples are included for a further understanding of this invention.

Example 1

1.5 grams of poly(4,4'-isopropylidenebisphenoxyethyl-co-tetramethylene isophthalate) binder containing 0.5 gram of 4,4'-benzylidene-bis(N,N-diethyl-m-toluidine) photoconductor and .04 gram of 2,4-(4-ethoxyphenyl)-6-(4-n-amlyoxystyryl) pyrylium fluoroborate sensitizer are dissolved in 15.6 grams of methylene chloride by stirring the solids in the solvent for one hour at room temperature. The resulting solution is hand coated at a wet coating thickness of 0.004 inch on a conducting layer comprising the sodium salt of a carboxyester lactone, such as described in U.S. 3,120,028, which in turn is coated on a cellulose acetate film base. The coating block is maintained at a temperature of 90° F. After drying, the electrophotographic element is charged under positive corona source until the surface potential, as measured by an electrometer probe, reaches about 600 volts. It is then subjected to exposure from behind a stepped density gray scale to a 3000° K. tungsten source. The exposure causes reduction of the surface potential of the element under each step of the gray scale from its initial potential, V_0 , to some lower potential, V , whose exact value depends on the actual amount of exposure in meter-candle-seconds received by the area. The results of the measurements are plotted on a graph of surface potential V vs. log exposure for each step. The shoulder speed is the numerical expression of 10^4 multiplied by the reciprocal of the exposure in meter-candle-seconds required to reduce the 600 volt charged surface potential by 100 volts. The toe speed is the numerical expression of 10^4 multiplied by the reciprocal of the exposure in meter-candle-seconds required to reduce the 600 volt charged surface potential to 100 volts. This coating is found to have a positive 100 v. toe speed of 250. Similar results are obtained when 0.5 gram of bis(4-diethylamino)-1,1,1-triphenylethane or 0.5 gram of bis(4-diethylamino)tetraphenylmethane are used as photoconductors in place of the 4,4'-benzylidene bis(N,N-diethyl-m-toluidine) for both positive and negative charging.

Example 2

Example 1 is repeated except that binder employed is poly(4,4'-isopropylidenebisphenoxyethyl-co-ethylene terephthalate). This binder does not fall within the scope of the invention in that the terephthalate is used instead of the isophthalate and is included for comparison purposes only. The coating has a positive 100 volt toe speed of 128.

The following Examples 3 and 4 are identical to Example 1 except for the binder employed. 1.5 grams of various binders are used in each of the following examples. In each case a significant improvement is noted in the 100 v. toe speeds over binders of the type described in the preceding example.

TABLE II

Example	Binder	Positive 100 v. toe speed
3	Poly(4,4'-isopropylidenebisphenoxyethyl-co-ethylene isophthalate)	200
4	Poly(4,4'-isopropylidenebisphenoxyethyl-co-neopentyl isophthalate)	200

Example 5

The coating compositions of Examples 1, 3 and 4 are again coated in the manner described in Example 1. In a darkened room, the surface of each of the photoconductive layers so prepared is charged to a potential of about +600 volts under a corona charger. The layer is then covered with a transparent sheet bearing a pattern of

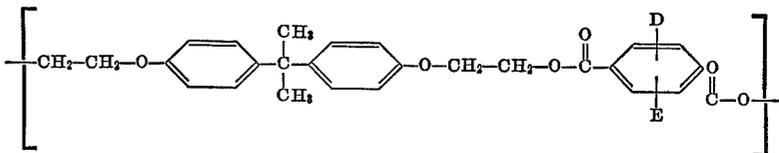
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opaque and light transmitting areas and exposed to the radiation from an incandescent lamp with an illumination intensity of about 75 meter-candles for 12 seconds. The resulting electrostatic latent image is developed in the usual manner by cascading over the surface of the layer a mixture of negatively charged black thermoplastic toner particles and glass beads. A good reproduction of the pattern results in each instance.

Example 6

The copolyesters described herein are made by standard melt condensation techniques. Poly(4,4'-isopropylidenebisphenoxyethyl - co - ethylene isophthalate) is prepared by mixing 0.15 mole of dimethyl isophthalate, 0.075 mole of 4,4' - isopropylidenebisphenoxyethanol, 0.10 mole of ethylene glycol and 0.05 gram of tetrabutyl orthotitanate. Nitrogen is bubbled through the mixture,

(B)

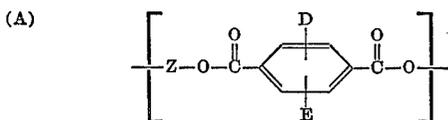


and it is heated at 200° C. for 2 hours to distill off methanol. The temperature is raised to 250° C. and vacuum applied until complete polymerization is completed. The resultant polymer contains 50% of repeating unit A and 50% B. The other polymers are prepared by the same method by replacing ethylene glycol with the appropriate compound. For example, trimethylene glycol would be used to prepare polymer 1, tetramethylene glycol for polymer 2, diethylene glycol for compound 4, etc.

The invention has been described in detail with particular reference to preferred embodiments thereof but it will be understood that variations and modifications can be effected within the spirit and scope of the invention as described hereinabove and as defined in the appended claims.

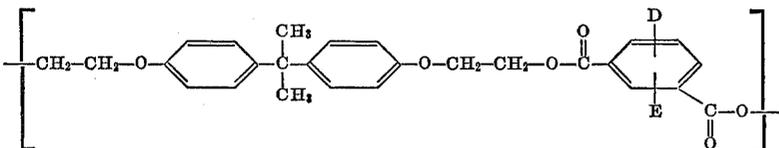
I claim:

1. An electrophotographic element capable of reproducing continuous tones comprising a support having coated thereon a photoconductive composition comprising a photoconductor and a binder for said photoconductor comprising a polyester having 2-80% repeating units of



and 20-98% of

(B)



wherein Z is an alkylene group having 2 to 10 carbon atoms, and D and E are each selected from the group consisting of a hydrogen atom, a halogen atom, an aryl group, an alkyl group, an aryloxy group and an alkoxy group.

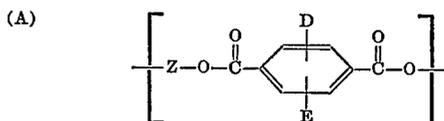
2. The electrophotographic element of claim 1 wherein Z is selected from the group consisting of

- a straight chain alkylene hydrocarbon radical,
- an oxydialkylene radical,
- an alkylene bis(oxyalkylene) radical,
- a cycloalkylene hydrocarbon radical,
- a branched chain alkylene hydrocarbon radical,
- a thiodialkylene radical and
- an alkylene bis(thiaalkylene) radical.

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3. The electrophotographic element of claim 1 wherein the photoconductive composition contains a sensitizer selected from the group consisting of cyanine and pyrylium dye salts.

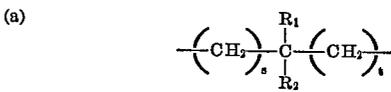
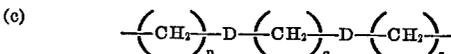
4. An electrophotographic element capable of reproducing continuous tones comprising a support having coated thereon a photoconductive composition comprising an organic photoconductor, a sensitizer and a binder for said photoconductor and sensitizer, comprising a random, linear co-polyester having 2-80% repeating units of



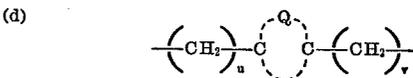
and 20-98% of

wherein Z is an alkylene group having 2 to 10 carbon atoms, and D and E are each selected from the group consisting of a hydrogen atom, a halogen atom, an aryl group, an alkyl group, an aryloxy group and an alkoxy group.

5. The electrophotographic element of claim 4 wherein Z is selected from the group consisting of

(R₁)_nD(R₂)_o

and



wherein:

t is an integer from 1 to 7;

n and o are each integers from 2 to 7;

s is an integer from zero to 7;

u and v are each integers from 0 to 5;

p, q and r are each integers from 2 to 6;

D is selected from the group consisting of an oxygen atom and a sulfur atom;

Q represents the atoms necessary to complete a 3 to 6 carbon atom cycloalkyl radical;

R₁ and R₂ are each selected from the group consisting of hydrogen and an alkyl radical having 1 to 5 carbon atoms; and

R₃ and R₄ are each alkylene radicals.

6. The electrophotographic element of claim 4 wherein the sensitizer is selected from the group consisting of carbocyanine, pyrylium, thiapyrylium and selenapyrylium dye salts.

7. The electrophotographic element of claim 4 wherein the organic photoconductor is 4,4'-benzylidenebis(N,N-diethyl-m-toluidine).

8. The electrophotographic element of claim 4 wherein the binder is poly(4,4'-isopropylidenebisphenoxyethyl-co-trimethylene isophthalate).

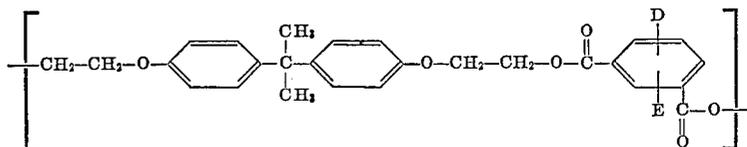
9. The electrophotographic element of claim 4 wherein the binder is poly(4,4'-isopropylidenebisphenoxyethyl-co-tetramethylene isophthalate).

10. The electrophotographic element of claim 4 wherein the binder is poly(4,4'-isopropylidenebisphenoxyethyl-co-neopentyl isophthalate).

11. The electrophotographic element of claim 4 wherein the binder is poly(4,4'-isopropylidenebisphenoxyethyl-co-ethylene isophthalate).

12. An electrophotographic element capable of reproducing continuous tones comprising a support having coated thereon a photoconductive composition comprising 10 to about 60 weight percent of 4,4'-benzylidenebis

(B)



(N,N-diethyl-m-toluidine) as an organic photoconductor, 0.005 to about 5.0 weight percent of a sensitizer for said photoconductor and poly(4,4'-isopropylidenebisphenoxyethyl-co-trimethylene isophthalate) as a binder for said photoconductive composition.

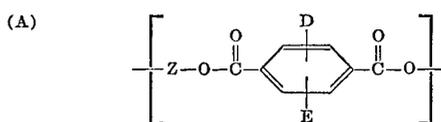
13. An electrophotographic element capable of reproducing continuous tones comprising a support having coated thereon a photoconductive composition comprising 10 to about 60 weight percent of 4,4'-benzylidenebis(N,N-diethyl-m-toluidine) as an organic photoconductor, 0.005 to about 5.0 weight percent of a sensitizer for said photoconductor and poly(4,4'-isopropylidenebisphenoxyethyl-co-tetramethylene isophthalate) as a binder for said photoconductive composition.

14. An electrophotographic element capable of reproducing continuous tones comprising a support having coated thereon a photoconductive composition comprising 10 to about 60 weight percent of 4,4'-benzylidenebis(N,N-diethyl-m-toluidine) as an organic photoconductor, 0.005 to about 5.0 weight percent of a sensitizer for said photoconductor and poly(4,4'-isopropylidenebisphenoxyethyl-co-neopentyl isophthalate) as a binder for said photoconductive composition.

15. An electrophotographic element capable of reproducing continuous tones comprising a support having coated thereon a photoconductive composition comprising 10 to about 60 weight percent of 4,4'-benzylidenebis(N,N-diethyl-m-toluidine) as an organic photoconductor, 0.005 to about 5.0 weight percent of a sensitizer for said photoconductor and poly(4,4'-isopropylidenebisphenoxy-

ethyl-co-ethylene isophthalate) as a binder for said photoconductive composition.

16. In an electrophotographic process wherein an electrostatic charge pattern is formed on an electrophotographic element by applying a uniform charge to the surface of the photoconductive layer and exposing to a light image, the improvement characterized in that said electrophotographic element has a photoconductive layer containing a binder having 2-80% repeating units of



and 20-98% of

wherein Z is an alkylene group having 2 to 10 carbon atoms, and D and E are each selected from the group consisting of a hydrogen atom, a halogen atom, an aryl group, an alkyl group, an aryloxy group and an alkoxy group.

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CHARLES E. VAN HORN, Primary Examiner

U.S. Cl. X.R.

96-1.5, 1.8; 252-501; 260-47 C

UNITED STATES PATENT OFFICE
CERTIFICATE OF CORRECTION

Patent No. 3,703,371 Dated November 21, 1972

Inventor(s) Stewart H. Merrill

It is certified that error appears in the above-identified patent and that said Letters Patent are hereby corrected as shown below:

Column 1, line 7, "16" should read ---31---;
line 23, "16" should read ---31---.

Signed and sealed this 12th day of June 1973.

(SEAL)
Attest:

EDWARD M. FLETCHER, JR.
Attesting Officer

ROBERT GOTTSCHALK
Commissioner of Patents