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COLOR COUPLERS CONTAINING TERPHENYL RADICALS

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12 Claims. (Cl. 95—6)

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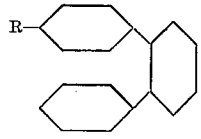
This invention relates to color photography and particularly to color-forming compounds for color photography.

The use of color-forming compounds which react with the development product of primary aromatic amino developing agent to form colored images on photographic development is well known and has been the subject of numerous prior patents. The dyes formed in this way are insoluble in water and in the ordinary photographic developing and fixing baths, although the silver images formed simultaneously with them during the photographic development may be removed from the photographic layer to leave pure dye images in the layer. The coupler compounds employed in this way may be added to the developing solution or in certain cases may be incorporated in the photographic layer prior to exposure. When the coupler compounds are incorporated in the developer solution, it is desired that they be sufficiently soluble in the developer solution so that amounts may be used which produce sufficiently high dye densities in the resulting photographic image. One of the difficulties encountered when color-forming compounds are incorporated in a photographic emulsion is the tendency of the coupler to crystallize in the emulsion. This produces excessive graininess.

It is therefore an object of the present invention to provide novel photographic coupler compounds having desirable color-forming properties. A further object is to provide coupler compounds which may be incorporated in a photographic emulsion which is part of a multi-layer coating and which do not diffuse from the emulsion in which they are incorporated. Other objects will appear from the following description of my invention.

These objects are accomplished by incorporating in the usual coupler molecule a terphenyl radical, especially an o-terphenyl radical. The preparation of o-terphenyl and some of its derivatives have been described by Allen and Pingert in Journal of the American Chemical Society, vol. 64, pages 1365 and 2639.

The couplers which we propose to use may have the following structural formula:



where R represents a nitrogen or carbon atom connected to, or part of, a coupler molecule containing a reactive methylene or a reactive phenolic methenyl group. The reactive methylene groups ordinarily encountered in color couplers are those of the structure

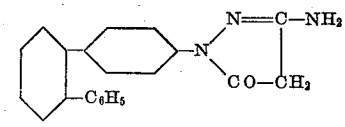
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$$—CO—CH_2—CO— \text{ or } —CO—CH_2—CN$$

The phenolic methenyl groups usually encountered in coupler molecules are those of the phenolic or naphtholic hydroxyl type having a reactive position para to the hydroxyl group, that is, a position which is unsubstituted or which is substituted with a replaceable group. In the general formula above, the terphenyl radical may contain other substituents. The general formula illustrates the structure of o-terphenyl but I may also employ other terphenyl radicals such as m-terphenyl and p-terphenyl as illustrated in the specific examples below.

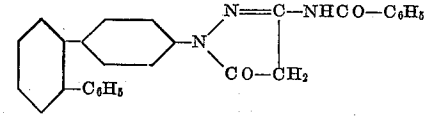
The following examples illustrate compounds which may be used according to my invention and the method of preparing them.

1. 1-[4'-(o-terphenylyl]-3-amino - 5 - pyrazolone



One part each of hydrazino-o-terphenyl and ethyl β-ethoxy-β-iminopropionate are added to 10 parts of ethyl alcohol, the mixture is heated to boiling, and ⅓ part of glacial acetic acid is added; the mixture is refluxed for 2 minutes, and cooled to room temperature. A freshly prepared solution of sodium ethoxide (⅕ part of sodium dissolved in 6⅔ parts of absolute ethyl alcohol) is added to the solution. The alkaline solution is refluxed for 30 minutes; it is then cooled to room temperature and acidified with 2⅔ parts of glacial acetic acid. If no precipitate separates in 30 minutes it is seeded with a previously prepared sample. The solid is filtered off after 2 hours, and washed with alcohol and water. The yield of the aminopyrazolone. M. P. 215–220°, is ⅗ part. For purification one part of the crude pyrazolone is crystallized from 45 parts of alcohol to give 0.5 part of lustrous, tan-colored plates, M. P. 223–225°.

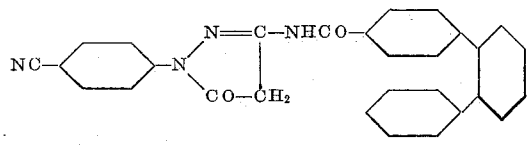
2. 1-[4'-(o-terphenylyl)]-3-benzamido-5 - pyrazolone



A suspension of 1 part of 1-[4'-(o-terphenylyl)]-3-amino-5-pyrazolone in 10 parts of anhydrous pyridine is treated with 1 part of benzoyl chloride. The mixture is heated for 30 minutes on the steam bath, it is then poured into 50 parts of 5% hydrochloric acid. The aqueous solution

is decanted from the gum which separates. The gum is rinsed with water and then dissolved in 20 parts of 5% alcoholic-potassium hydroxide, after 5 minutes the alkaline solution is acidified to litmus with acetic acid; the tan colored solid which separates is washed with alcohol, water and again with alcohol, and dried at 100°. The yield is 1 part of the benzamidopyrazolone, M. P. 228–230° The crude material can be crystallized from ethanol or from acetic acid to give a product with melting point of 234–235°.

3. 1-(p-cyanophenyl)-3-(o-terphenyl-4 - carbamido)-5-pyrazolone

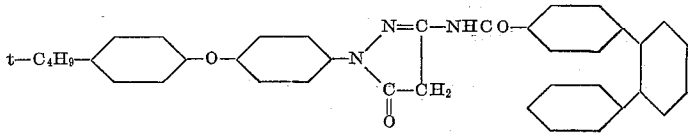


Four parts of 1-(p-cyanophenyl)-3-amino-5-pyrazolone is suspended in 100 parts of dry dioxane and at room temperature with stirring 11.68 parts of o-terphenyl-4-carboxylic acid chloride is added in about 5 minutes. When the initial rise in temperature has subsided, 5.16 parts of quinoline are added and the reaction mixture is stirred for 2 hours. The temperature, which at first reaches about 35° C., falls to about that of the room.

Two hundred parts of cold water is added to the flask and stirring continued until the oily material first formed has solidified. The solid is filtered, washed several times with cold water and dried.

The dry material is suspended in 200 parts of alcohol and 2.9 parts of 40% sodium hydroxide solution is added to the suspension, and the reaction stirred for 2 hours at room temperature. Twenty-five parts of cold water is added to the solution, the whole filtered if necessary, and neutralized with 2.7 parts of acetic acid. The solid formed is filtered, washed with water and dried. After crystallization from 25 parts of alcohol, the product melts at 255–257° C.

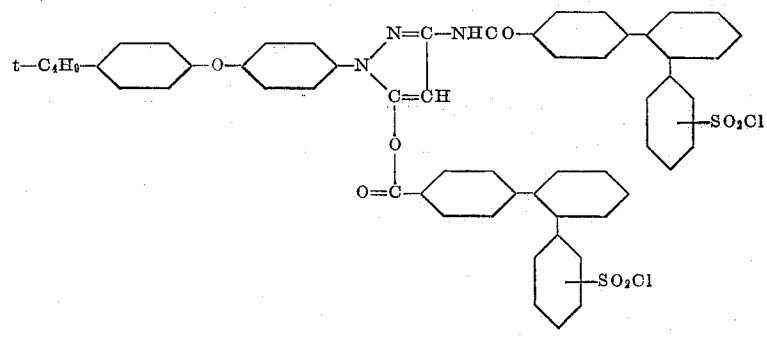
4. 1 - [p - (p' - tert. - butylphenoxy) - phenyl]--3-(4'-o-terphenamido)-5-pyrazolone



This coupler was prepared by heating a mixture of 1.6 parts of 1-p-(p'-tert.-butylphenoxy)-phenyl-3-amino-5-pyrazolone and 3 parts of o-terphenyl-4-carboxy chloride in 15 parts of xylene under a reflux condenser for 0.5 hour.

The diacyl compound which resulted was partially saponified at room temperature with alcoholic potassium hydroxide. The monoacyl compound was precipitated by acetic acid. It recrystallized from acetic acid, forming a crystalline powder, M. P. 180–181°.

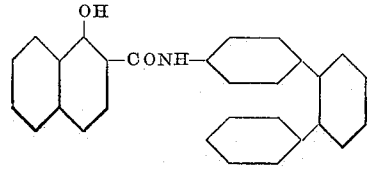
5. 1 - p - (p' - tert. - butylphenoxy) - phenyl-3 - (x' - chlorosulfonyl - o - terphenamido)-5-(x'-chlorosulfonyl-o-terphenoxy)-pyrazole



This coupler was prepared by condensing one mole of 1-p-(p'-tert.-butylphenoxy)-phenyl-3-amino-5-pyrazolone and two moles of 4'-carboxy-x-sulfo-o-terphenyl dichloride in boiling anhydrous ethyl acetate. The product was obtained as a light tan powder on removal of the solvent.

	Calc. for $C_{57}H_{43}Cl_2N_3O_8S_2$	Found
C	66.3	65.3
H	4.2	4.7
Cl	6.9	7.2

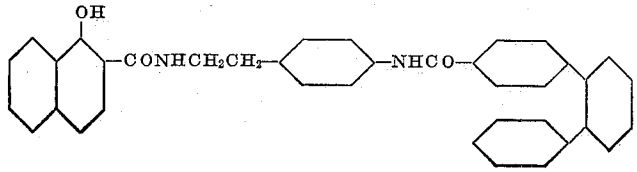
6. 1 - hydroxy - 2 - {[4' - (2'' - phenyl)-phenyl]-naphthanilide}



This coupler was prepared by heating a mixture of equimolecular amounts of phenyl 1-hydroxy-2-naphthoate and 4'-amino-o-terphenyl at 150–160° for 10 minutes. The product recrystallized from alcohol in rods; M. P. 180–181°.

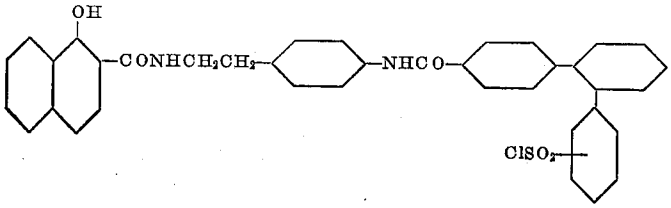
	Calc. for $C_{29}H_{21}NO_2$	Found
C	83.85	82.61
H	5.06	5.23

7. 1 - hydroxy - 2 - {β - [4' - (4'' - o - terphenamido)]-phenethyl}-naphthamide



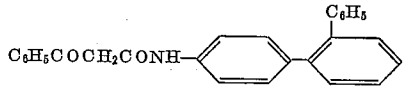
This coupler was prepared by the condensation below 50° C. of 1-hydroxy-2-β-(4'-aminophenyl)-ethylnaphthamide hydrochloride and o-terphenyl-4'-carboxy chloride in dioxane in the presence of quinoline. The product crystallized from glacial acetic acid in plates, M. P. 227–228°.

8. 1-hydroxy-2-{β-[4'-(x''-chlorosulfonyl-4''-o-terphenamido)]-phenethyl}-naphthamide



This coupler was prepared by condensing at room temperature for 1 hour a mixture of equimolecular parts of 1-hydroxy-2-β-(4'-aminophenyl)-ethylnaphthamide and 4'-carboxy-x-sulfo-o-terphenyl dichloride in dioxane and quinoline. The solid was isolated by drowning the reaction mixture in dilute hydrochloric acid and filtering. It was dried at 50° C. and had a melting point of 80–130°.

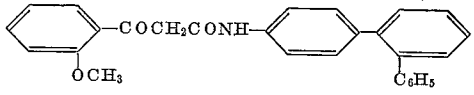
9. 4-benzoylacetamido-o-terphenyl



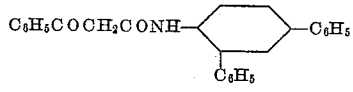
A solution of 2.5 parts of ethyl benzoylacetate and 10 parts of xylene in an open vessel was heated by means of an oil bath maintained at 150–160°. Then 2.45 parts (0.01 mole) of 4-amino-o-terphenyl was added and the mixture was heated to gentle boiling for one hour. The solution was cooled to 70°, 4 volumes of technical "hexane" was added and the mixture chilled. The crystalline product was filtered, washed with hexane and recrystallized from absolute alcohol (50 to 1). The yield of product, melting at 163–164° C., was 2.55 parts (65%).

Alternatively the procedure described in "Organic Syntheses," vol. 25, p. 7, for benzoylacetanilide may be employed.

10. By a similar procedure, 4-(o-methoxybenzoylacetamido)-o-terphenyl was obtained, using ethyl o-methoxybenzoylacetate. It melts at 149–150° C.

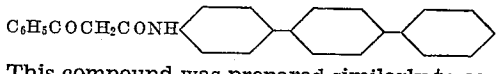


11. 4-(benzoylacetamido-m-terphenyl)



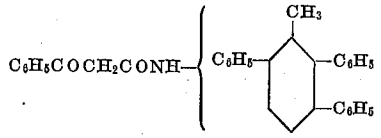
This compound was prepared similarly to compound 9 from ethylbenzoylacetate and 4-amino-m-terphenyl. It had a melting point of 154–155° C. 4-amino-m terphenyl is described in Journal of the American Chemical Society, vol. 64, page 2485 (1942).

12. 4-(benzoylacetamido-p-terphenyl)



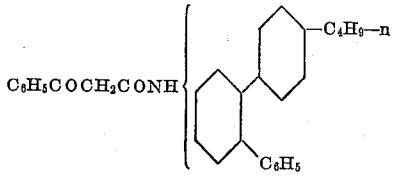
This compound was prepared similarly to compound 9 (from ethylbenzoylacetate and 4-amino-p-terphenyl). It had a melting point of 262–263° C. 4-amino-p-terphenyl is described in Journal of the American Chemical Society (1938), page 1364.

13. x-(Benzoylacetamido)-2,3,6-triphenyl toluene



This compound was prepared similarly to compound 9 (from ethyl benzoylacetate and x-amino-2,3,6-triphenyl toluene). It had a melting point of 193–194° C.

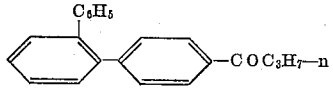
14. x-(Benzoylacetamido)-4-n-butyl-o-terphenyl



A mixture of 3 parts of ethyl benzoylacetate and 10 parts of xylene was heated to reflux and 2 parts of x-amino-4-n-butyl-o-terphenyl was added. The solution was heated at reflux for 1 hr. It was then concentrated in vacuo to give a yellow oil which coupled with 2-amino-5-diethylaminotoluene hydrochloride, in the presence of an oxidizing agent, to give a yellow dye.

The intermediates used in the preparation of the couplers described above were prepared as follows:

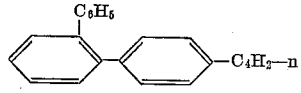
4-n-butyryl-o-terphenyl



A solution of 138 parts of o-terphenyl, 67 parts of n-butyryl chloride and 150 parts of ethylene dichloride was cooled to −10° C., and 87 parts of resublimed aluminum chloride added in portions with stirring during one hour. The mixture was stirred for four hours at −5 to 0° and decomposed with iced hydrochloric acid. The organic layer was removed, washed with dilute acid, mixed with water and made almost neutral with sodium carbonate solution. The solvent was removed by steam distillation, and the residual oil extracted with 700 parts of ether. The ether solution was washed and evaporated. Distillation of the crude, fluorescent oil yielded 120 parts of the ketone; B. P. 187–190° C. (1 mm.). It crystallized from methanol in hard, transparent prisms; M. P. 78° C.

Calcd. for $C_{22}H_{20}O$: C, 87.9; H, 6.7. Found: C, 87.6; H, 6.7.

4-n-butyl-o-terphenyl

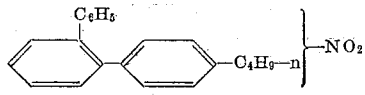


Amalgamated zinc was prepared by treating 250 parts of mossy zinc with 300 parts of water, 25 parts of mercuric chloride and 12 parts of concentrated hydrochloric acid. To the zinc amal-

gam thus obtained was added 35 parts of concentrated hydrochloric acid, 10 parts of acetic acid, and a solution of 50 parts of 4-n-butyryl-o-terphenyl in 100 parts of toluene. The mixture was heated under reflux for 24 hours; hydrogen chloride was passed in slowly during the first six hours. The toluene solution was removed, washed and distilled. If necessary the distillate may again be subjected to the above treatment, using a fresh portion of amalgamated zinc. The product (34 parts) distilled at 165–167° C. (1 mm.), and crystallized from methanol in large, transparent prisms; M. P. 44° C.

Calcd. for $C_{22}H_{22}$: C, 92.3; H, 7.7. Found: C, 92.3; H, 7.7.

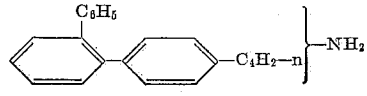
4-n-butyl-x-nitro-o-terphenyl



The nitration of 4-n-butyl-o-terphenyl (8.6 parts) was carried out in the manner described for 4-nitro-o-terphenyl distillation of the crude product yielded 6.7 parts of an intractible, yellow oil; B. P. 196–206° C. (1 mm.).

Calcd. for $C_{22}H_{21}NO_2$: N, 4.2. Found: N, 4.3.

x-Amino-4-n-butyl-o-terphenyl



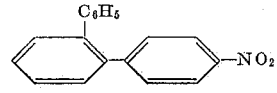
The reduction of 4-n-butyl-x-nitro-o-terphenyl was carried out in the manner described for 4-amino-o-terphenyl. The product was an intractible oil which was purified by preparation of the p-nitrobenzanilide derivative, followed by hydrolysis. The pure amine crystallized from alcohol in fine white needles; M. P. 60° C.

Calcd. for $C_{22}H_{22}N$: N, 4.6. Found: N, 4.9.

The p-nitrobenzanilide derivative recrystallized from alcohol melted at 155.5° C.

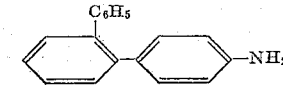
Calcd. for $C_{29}H_{26}N_2O_3$: N, 6.2. Found: N, 6.3.

4-nitro-o-terphenyl



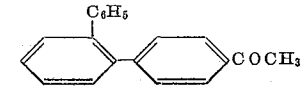
A solution of 115 parts of 95% o-terphenyl in 250 parts of glacial acetic acid was heated to 55–60° C., and a solution of 25 parts of fuming nitric acid (d. 1.49) and 50 parts of red fuming nitric acid (d. 1.59) was added dropwise during one hour. The temperature was kept at 55–60° C. by regulating the rate of addition and by cooling when necessary. The reaction mixture was then heated at the same temperature for 6 hours, cooled, and poured into 1500 parts of water. The yellow, semi-crystalline tar which formed was washed successively, by decantation, with warm water, 5% sodium carbonate solution (400 parts), and again with warm water. It was dissolved in 750 parts of ether and washed again with water, twice with 50 parts of 5% sodium carbonate solution, water and saturated brine. When the ether was evaporated the residue weighed 136 parts (99% crude yield). Crystallization from 1300 parts of 3A alcohol yielded 61 parts of product, melting at 108–113.5° C. Recrystallization from 600 parts of alcohol gave 52.3 parts (38% yield) of 4-nitro-o-terphenyl, melting at 111–114° C.

4-amino-o-terphenyl



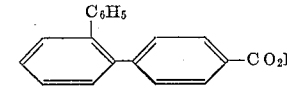
A hot solution of 140 parts of 4-nitro-o-terphenyl (M. P. 111–114°) in 1600 parts of 3A alcohol was placed in a copper, low-pressure bomb of 4500 parts capacity. About 10 parts of Raney nickel and 5 parts of sodium carbonate were added, and hydrogen was introduced to a pressure of 50 lbs. The bomb was shaken and heated externally by steam until the absorption of hydrogen had ceased. It was necessary to introduce more hydrogen as the pressure dropped to 10–20 lbs. The hot reduction mixture was filtered by suction through a pad of Filter-Cel, the filtrate cooled to 0°, and the product removed by filtration. The first crop weighed 75.4 parts; M. P. 117–118°. Concentration of the filtrate to 300 parts gave an additional 41.8 parts (M. P. 116–117°) for a total yield of 117.2 parts (94%).

4-acetyl-o-terphenyl



A solution of 138 parts of 95% o-terphenyl in 150 parts of ethylene dichloride, and 50 parts of acetyl chloride was placed in a suitable vessel, equipped with a stirrer, condenser and thermometer. Then 87 parts of resublimed aluminum chloride was added in portions during one hour keeping the temperature below 0°. When the addition was completed, stirring was continued for 2 hours; the mixture was then decomposed by iced hydrochloric acid, and the solvent removed by steam distillation. The residual oil was taken up in ether and washed successively with water, 5% sodium carbonate, water, saturated brine and then filtered. The product separated on concentration of the filtrate. Yield of 101 parts (62%); M. P. 93.5–94°.

4-carboxy-o-terphenyl



A sodium hypochlorite solution was prepared by passing 111 parts of chlorine into a solution of 150 parts of sodium hydroxide in 225 parts of water and containing 450 parts of crushed ice. The temperature was kept below 0° during the addition of chlorine by an ice-salt bath.

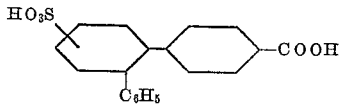
The hypochlorite solution was added in a fine stream to a warm solution of 90 parts of 4-acetyl-o-terphenyl in 1200 parts of methanol contained in a suitable vessel. The solution was stirred during the addition and considerable heat was evolved. Finally the mixture was heated under reflux for 5 hrs., cooled to about 5°, filtered, and washed with water. The solid cake was dissolved in 900 parts of hot water and filtered. The hot filtrate was acidified with concentrated hydrochloric acid (about 30 parts required), cooled, filtered, washed with water and dried. Yield of the crude acid was 75–80 parts.

By concentration of the methanolic filtrate (from the first filtration) additional sodium salt

was obtained, from which 18 parts more of the crude acid was recovered.

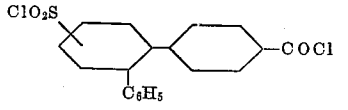
The acid was recrystallized from methanol to give 68.4 parts of pure product. Concentration of the filtrate to 200 parts gave an additional 11 parts for a total yield of 79.4 parts (87.7%); M. P. 202–203°.

4-carboxy-x'-sulfo-o-terphenyl



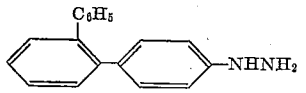
A solution of 27 parts of 4-carboxy-o-terphenyl in 300 parts of tetrachloroethane was cooled to 10° and treated dropwise with 12 parts of redistilled chlorosulfonic acid in 35 parts of tetrachloroethane. The mixture was then stirred overnight at 55–60° C., cooled, and the product filtered off, and rinsed with ether. The yield was 23 parts (66%). It can be crystallized from nitromethane, nitroethane, or chloroform, after which it has a melting point of 243–248°.

4-carboxy-x'-sulfo-o-terphenyl dichloride



An intimate mixture of 19 parts of 4-carboxy-x'-sulfo-o-terphenyl and 22.4 parts of phosphorus pentachloride, and protected from external moisture, was warmed on the steam bath, with occasional shaking, until the reaction had subsided. The liquid, which solidified on cooling, was broken up, decomposed by ice water and the acid chloride extracted with 650 parts of ether. After removal of the solvent under reduced pressure, 13 parts (62% yield) of the dichloride, M. P. 141–149°, remained. No suitable solvent for crystallization was found. It was characterized by conversion to the diamide, M. P. 272–277°, which had the correct analysis.

4-hydrazino-o-terphenyl



The diazonium chloride of 4-amino-o-terphenyl was prepared at 0° from 7.35 parts of the amine, 9 parts of concentrated hydrochloric acid, 40 parts of water and 2.2 parts of sodium nitrite. The resulting solution was added gradually to a cold (−10° C.) solution of 18 parts of stannous chloride dihydrate in 30 parts of concentrated hydrochloric acid. The insoluble complex was removed by filtration and stirred for 15 minutes with 22 parts of 40% sodium hydroxide solution and sufficient crushed ice to keep the temperature below 10° C. The cream-colored solid was filtered, suspended in ether and shaken with 30 parts of 5% sodium hydroxide solution until completely dissolved. The ether solution was washed, dried and evaporated to a small volume, and petroleum ether was added to precipitate 4.1 parts of the crude, cream-colored hydrazine (M. P. 118–124° C.). Recrystallization from alcohol raised the melting point to 123–125°.

Analysis: Calcd. for $C_{18}H_{16}N_2$: C, 83.1; H, 6.2. Found: C, 82.7; H, 6.4.

This compound was unstable and decomposed over a period of a few weeks.

The coupler compounds of my invention may be incorporated in the photographic layer prior to exposure and the colored image formed by development of the layer in a primary aromatic amino developing solution. In the case of multilayer film, the color forming compounds may be incorporated in one or more of the emulsion layers. They may be incorporated in the emulsion as described in Mannes and Godowsky U. S. Patent 2,304,940, granted December 15, 1942, or Jelley and Vittum U. S. Patent 2,322,027, granted June 15, 1943. Certain of the couplers may also be incorporated in the developing solution. When incorporated in the emulsion layer, the exposed emulsion may be developed in the following developer to form a colored image or colored images in the film.

	Grams
p-Amino-diethylaniline sulfate	2.5
Sodium sulfite (anhydrous)	2
Sodium carbonate (anhydrous)	20
Potassium bromide	1
Water to 1 liter	

The aromatic amino developing agents used with the coupler compounds of my invention include the alkyl phenylenediamines and alkyl tolueneediamines. These compounds are usually used in the form of hydrochloride or the sulfate. Suitable developing agents are diethyl-p-phenylende diamine hydrochloride, monomethyl-p-phenylenediamine hydrochloride, dimethyl-p-phenylenediamine hydrochloride and 2-amino-5-diethylamino toluene hydrochloride. The p-aminophenols and their substitution products may also be used where the amino group is unsubstituted. All of these compounds have an unsubstituted amino group which enables the oxidation product of the developer to couple with the color-forming compound to form a dye image.

My development process may be employed for the production of colored photographic images in layers of gelatin or other water-permeable carrier such as collodion organic esters of cellulose or synthetic resins. The carrier may be supported by a transparent material such as glass, a cellulose ester or a non-transparent reflecting material such as paper on an opaque cellulose ester. The emulsion may be coated on the support as a single layer or as a superposed layer on one or both sides of the support. The superposed layers may be differentially sensitized to form a natural color photographic image in the well-known manner.

The couplers of my invention may be easily prepared from readily available materials and are therefore more economical than many couplers previously described in the art. They have a high degree of solubility in solutions used to disperse them in the emulsion and are more readily dispersible according to the methods of Mannes and Godowsky U. S. Patent 2,304,940 and Jelley and Vittum U. S. Patent 2,322,027 than many couplers hitherto available. They have a higher resistance to crystallization than corresponding couplers containing a biphenyl group. All of these advantages make them desirable couplers for use in color photography.

It will be understood that the examples and compounds set forth herein are illustrative only and that my invention is to be taken as limited only by the scope of the appended claims.

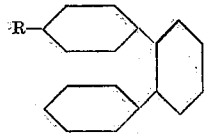
What I claim is:

1. A photographic emulsion for forming colored images, comprising a water-permeable car-

rier containing a sensitive silver halide and a coupler compound containing a reactive group selected from the class consisting of methylene and phenolic methenyl groups which are reactive with the development product of a primary aromatic amino developing agent, said coupler compound having attached thereto in a position other than at said reactive group, a terphenyl radical.

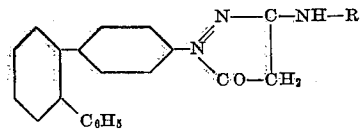
2. A photographic emulsion for forming colored images, comprising a water-permeable carrier containing a sensitive silver halide and a coupler compound containing a reactive group selected from the class consisting of methylene and phenolic methenyl groups which are reactive with the development product of a primary aromatic amino developing agent, said coupler compound having attached thereto in a position other than at said reactive group, an ortho-terphenyl radical.

3. A photographic emulsion for forming colored images comprising a water-permeable carrier containing a sensitive silver halide and coupler compound having the general formula:



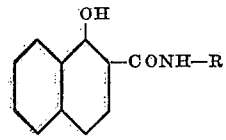
where R is selected from the class consisting of nitrogen and carbon atoms connected to a group containing a reactive group selected from the class consisting of methylene and phenolic methenyl groups which are reactive with the development product of a primary aromatic amino developing agent.

4. A photographic emulsion for forming colored images, comprising a gelatino-silver halide emulsion containing a coupler compound of the following formula:



where R is selected from the class consisting of hydrogen and benzoyl.

5. A photographic emulsion for forming colored images, comprising a gelatino-silver halide emulsion containing a coupler compound of the following formula:



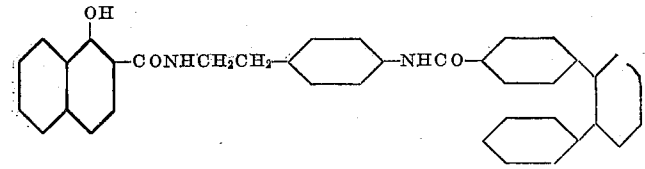
where R is selected from the class consisting of 4-o-terphenyl and β-[4-(4'-o-terphenamido)] phenethyl radicals.

6. A photographic emulsion for forming colored images, comprising a gelatino-silver halide emulsion containing a coupler compound of the following formula:

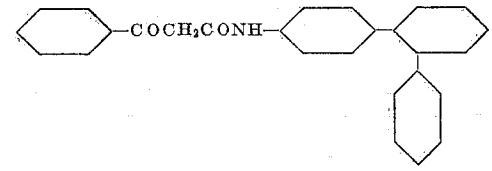
R—COCH$_2$CONH—R'

where R is a mononuclear aryl radical, and R' is a terphenyl radical.

7. A photographic emulsion for forming colored images, comprising a gelatino-silver halide emulsion containing a coupler compound of the following formula:

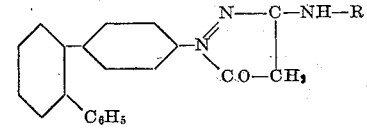


8. A photographic emulsion for forming colored images, comprising a gelatino-silver halide emulsion containing a coupler compound of the following formula:



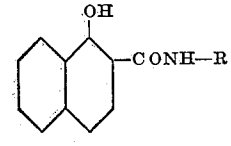
9. The method of producing a colored photographic image in a silver halide emulsion layer, which comprises exposing said layer and developing it with a primary aromatic amino developing agent in the presence of a coupler compound, containing a reactive group selected from the class consisting of methylene and phenolic methenyl groups which are reactive with the development product of a primary aromatic amino developing agent, said coupler compound having attached thereto in a position other than at said reactive group, a terphenyl radical.

10. The method of producing a colored photographic image in a silver halide emulsion layer, which comprises exposing said layer and developing it with a primary aromatic amino developing agent in the presence of a coupler compound having the following formula:



where R is selected from the class consisting of hydrogen and benzoyl.

11. The method of producing a colored photographic image in a silver halide emulsion layer, which comprises exposing said layer and developing it with a primary aromatic amino developing agent in the presence of a coupler compound having the following formula:



where R is selected from the class consisting of 4-o-terphenyl and β-[4-(4'-o-terphenamido)] phenethyl radicals.

12. The method of producing a colored photographic image in a silver halide emulsion layer, which comprises exposing said layer and develop-

ing it with a primary aromatic amino developing agent in the presence of a coupler compound having the following formula:

$$R—COCH_2CONH—R'$$

where R is a mononuclear aryl radical and R' is a terphenyl radical.

CHARLES F. H. ALLEN.

REFERENCES CITED

The following references are of record in the file of this patent:

UNITED STATES PATENTS

Number	Name	Date
2,039,730	Mannes	May 5, 1936
2,179,239	Wilmanns	Nov. 7, 1939