FOREWORD

This report describes the chemical research performed by the American Cyanamid Company, Organic Chemicals Division, Bound Brook, New Jersey, from 1 April 1961 to 30 April 1962, under Air Force Contract AF 33(616)-8056, Project No. 6301, "Aerospace Systems Personnel Protection," and Task No. 630103, "Vision Enhancement and Protection in Aerospace Environments." Dr. Richard E. Horn, Vision Section, Protection Branch, Life Support Systems Laboratory, 6570th Aerospace Medical Research Laboratories, was contract monitor until November 1961, when Major Paul W. Lappin assumed the responsibility.

The project supervisor at American Cyanamid was Dr. Ralph A. Coleman. The research chemists and authors of this report were Dr. Haig C. Conoian and Dr. Peter V. Susi; they were assisted by Miss Norma A. Bressler, Mr. John M. Dowd, Jr., and Mr. Joseph C. Mollica.

ABSTRACT

With the specific objective of developing a plastic or coating composition having strong near-infrared attenuation and high visible transmittance, a number of organic compounds were prepared and along with many compounds from chemical files were examined spectrophotometrically in solution. Those with near-infrared absorption were incorporated into plastics by one or more of the following methods: solution casting, milling and molding, monomer casting, burnishing, or dyeing. Absorption properties were then measured and light stabilities were determined after accelerated light exposure tests.

PUBLICATION REVIEW

This technical documentary report has been reviewed and is approved.

Wayne H. McCandless
WAYNE H. McCANDLESS
Chief, Life Support Systems Laboratory

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I. INTRODUCTION

In order to give complete protection to the human eye when it is exposed to the high intensity thermal radiation from nuclear weapons, it is necessary to attenuate the infrared portion of the electromagnetic spectrum between 750 and 2000 mμ. Infrared-attenuating coatings or plastics that would be useful for this purpose were investigated. Protective devices, such as goggles, would be opaque to infrared radiation but transparent to visible light during normal usage.

The specific requirements call for a plastic material or coating having at least 85% transmittance of visible light in the range between 400 and 700 mμ and at most 1% transmittance of infrared light between 750 and 2000 mμ. These transmittance values would be obtained with 1/4-inch thickness or less of the plastic. The optical properties of the plastic material should not be affected by thermal shock or changes in temperature and pressure. Also the plastic should possess good storage stability, high optical clarity and scratch resistance. Coatings, in addition to the above properties, must have good adhesive characteristics.

II. METHODS OF APPROACH

Infrared light can be attenuated either by absorption or by reflection. This can take place in the plastic itself or in a coating on the plastic.

The essential requirement, a coating or plastic with a spectrophotometric transmission greater than 85% between 400 and 700 mμ and that also has less than 1% transmittance between 750 and 2000 mμ, could be met by incorporating into a resin adequate quantities of additives that would impart the desired properties, without essentially altering the other physical properties of the coating or plastic. The types of additives that could be used are as follows:

a. Compounds having strong electronic absorption in the near infrared with visible transmission. Such compounds would absorb the infrared portion of the radiant energy.

b. Compounds having strong vibrational absorptions in the 1200 to 2000 mμ range. Such compounds would also absorb the infrared portion of the radiant energy.

c. Colorless pigments having the same refractive index in the visible as the resin in which they are incorporated, but a different refractive index than the resin in the infrared. Such products would attenuate infrared through scattering (Christiansen filter effect).

A second approach is the formation of a mirror-like coating of a strong infrared-absorbing compound on the surface of the base material. This would be done with monomeric or polymeric infrared absorbers. Such a coating would specularly reflect the infrared but transmit the visible light.

In addition to obtaining the desired spectral characteristics, appropriate physical properties must be obtained, such as proper compatibility of the additives in the resins, light, heat and chemical stability.

Most of the compounds evaluated were synthesized during the course of this investigation. In addition, an extensive screening program was conducted on compounds available from the contractor's chemical file. Those that showed no near-infrared absorption are listed in Appendix III.

The general methods of approach, therefore, may be summarized as follows:

   a. Infrared attenuation by absorption, including electronic and vibrational absorption.

   b. Infrared attenuation by reflection, including both specular and diffuse types.

During the course of this investigation major emphasis was placed on method (a).

Infrared-absorbing compounds are known, but they all absorb to some extent in the visible region. A major problem was to find an infrared absorber with a good visible window. Very weak absorptions in the visible might be tolerated, but preferably these should be located near the edges of the visible region, i.e., near 400 or 700 mμ where the eye sensitivity is less. Substantial progress along this line was made during the course of this work. Fig. 2, 3, 4, 5, 12, 17, 19, 22 and 24.

A second problem concerns the broad infrared range (750 to 2000 mμ) in which absorption is needed. Radiation in the 750 to 1400 mμ region should be best absorbed by electronic rather than by vibrational or rotational transitions. However, because of the difficulty of obtaining electronic transitions from 1400 to 2000 mμ, compounds having vibrational transitions between these wavelengths were also considered.

An alternate to the approach of incorporating colorless infrared absorbers in the plastic is to incorporate compounds which under normal conditions are colorless and non-infrared absorbing. Upon exposure to high intensity radiation, these products would become infrared absorbing. Such products would also absorb a high or low percentage of visible light depending on the structure selected and could therefore be of interest provided the change is rapidly reversible. An example of such a compound, which was prepared and evaluated, is 3,6-bis(dimethylamino)-9-phenylfluoren-9-nitrile (XLVIII)*. This compound, however, did not exhibit enough of the expected phototropic property.

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*Roman numerals were assigned compounds in order as cited in the Syntheses section.

The number of compounds known to have strong absorption peaks above 700 mμ is limited. However, some such classes of compounds have been reported in the literature[2,23] or developed in our laboratories[6,7]. The general classes of compounds investigated during this work are:

A. Fluorenol Salts
B. Triarylaminium Free Radical Salts
C. Polymethines
D. Metal Complexes
E. Miscellaneous Compounds

These compounds were evaluated in poly(methyl methacrylate) and other plastics.

A. Fluorenol Salts

A number of color salts of substituted 9-phenylfluoren-9-ols (Compounds XII and XXIV) have been prepared during the course of this investigation. These compounds have interesting spectrophotometric properties (Tables I and II).
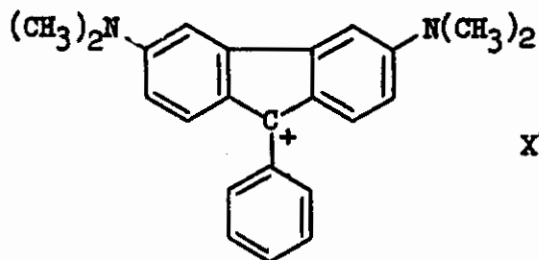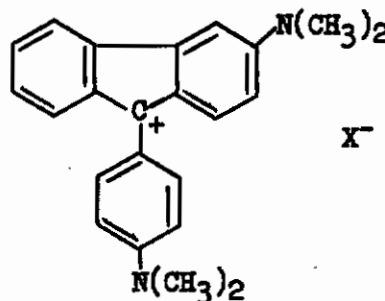
XII   XXIV

Although compound XXIV has a stronger absorptivity in the near infrared, it is more highly colored than compound XII, and does not absorb as far out in the infrared. The high transmittance of compound XII in the visible is marred only by an absorption peak at 480 mμ. These color salts have a greater tendency to hydrolyze to the colorless carbinol than the analogous triphenylmethane dye, Malachite Green. Therefore, the spectral curves were run on glacial acetic acid solutions of the fluorenol salts in order to insure complete formation of the infrared-absorbing cations (XII, XXIV).

The salts of these fluorenols show the same kind of curve in plastics as they do in solution (Fig. 1 to 6). Variations in the anion (X-) moiety of the above compounds has been found to cause a variation in the compatibility in plastics, in hydrolytic stability, and in the degree of light stability (Table III). In particular, the perchlorate salt of XII showed the best light stability. Furthermore burnishing, or rubbing a

finely powdered solid absorber on the surface of the plastic, resulted in a highly attenuating coating with a substantial increase in light stability over plastic samples containing the same absorber incorporated in solution (Table IV). Apparently these compounds are more stable to light when in a crystalline state. Although the burnished samples appeared hazy by transmission, the haze can be diminished somewhat by resin overcoating. Fig. 5 shows the spectral transmittance of the fluorenol salt XII (where X is the tetracyanonickelate anion) on cellulose acetate overcoated with an acrylic resin Acryloid B-72. This was one of the clearest burnished samples obtained.

Of the various fluorenol salts XII prepared, the perfluorobutyrate and trifluoroacetate salts were the most compatible in monomer-cast poly-(methyl methacrylate) (Table V) and the ethane sulfonate and diphenyl-4,4'-disulfonate salts were the most compatible in poly(methyl methacrylate) samples prepared by milling and molding, as evidenced by the clarity of the samples obtained. However, in all the samples prepared by milling and molding, the full absorption was not obtained (Table VI). The 0.1% of absorber used was much more than the theoretical concentration required to give 0% transmittance at 960 mμ. This may be due in part to heat instability during the milling process and/or incomplete solubility of the absorber in the resin.

Attempts to incorporate these fluorenol salts in CR-39, poly-(methyl methacrylate), or Laminac 4123 by monomer casting in the presence of peroxide catalyst were unsuccessful. However, this difficulty was partly overcome by using Porophor N (2,2'-azobis[2-methylpropionitrile]) as the catalyst. Another method used to avoid decomposition of the absorber was to dye the compound on the preformed plastic. This was partially successful in the case of CR-39 and poly(methyl methacrylate) samples dyed with the tetrachlorozincate salt of the fluorenol XII (Table VII).

This class of compounds in general shows poor light stability, particularly to ultraviolet radiation. Some degree of improvement in light stability was obtained by use of an ultraviolet-absorbing screen. As can be seen from Table VIII, after three hours of exposure, in the Atlas Fade-Ometer, the protected sample retained 73.5% activity whereas the unprotected sample showed only 23% activity. Considerably better light stability values were obtained in the Atlas Xenon Weather-Ometer (Table IX). These values are more representative of those which would be obtained on exposure to natural sunlight (Fig. 7 and Ref. 18), due to the greater similarity in relative energy distribution in the ultraviolet and visible regions.

B. Triarylaminium Free Radical Salts

This class of compounds showed excellent infrared absorption with very good visible transmission. Since the visible transmission maxima are at about 550 mμ, these compounds possesses a favorable light green color (Fig. 8 to 14). As can be seen from Table X, these compounds all absorb strongly in the near infrared around 960 mμ with somewhat less absorption

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at the lower end of the visible region ($400 \, m\mu$). An unexpected higher molar absorptivity was obtained for the fluoborate salts in each case over the corresponding perchlorate salts. No obvious explanation is evident for this difference.

The best member of this class of compounds is the hexaethyl fluoborate derivative, compound XXX, because of its high absorptivity and good compatibility. In addition, as can be seen from Table XI, this derivative has shown a slight improvement in light stability over other members of this class of compounds, whose light stability in general, is rather low. The light stability can be improved somewhat by the use of an ultraviolet-absorbing screen (Table XI). As was the case with the fluorenol salts, the light stability values of members of this class were higher when a Xenon unit was used as the light source (Table XI).

Difficulties were encountered in attempts to monomer cast these compounds. Their infrared-absorbing properties were destroyed due to apparent instability of the compounds towards the polymerization catalyst used. These compounds were milled and molded into plastics with the fluoborate salts being the most compatible and in particular, the hexaethyl derivative. (Note in Table XII their higher percent transmittance at $550 \, m\mu$.) When the perchlorate salt of the hexamethyl derivative (XXVII) was burnished onto plastic, the expected increase in light stability was not observed (Table XIII), probably due to the high solubility of the compound in the plastic.

Due to limitations in other methods of application of this class of compounds, a greater emphasis was placed on development of dyeing techniques. Samples absorbing very strongly in the near infrared were obtained when the compounds were dyed from 50% aqueous alcohol containing a methyl salicylate emulsion (Table XIV).

As was the case with the fluorenol salts, attempts at vacuum evaporation were unsuccessful resulting only in decomposed products.

C. Polymethines

The main classes of compounds investigated were:

   a.   Substituted tetraphenylvinylcarbonium salts
   b.   Xanthylomethines

These compounds in general possessed high near-infrared absorption with relatively low visible absorption (Fig. 15-25). As can be seen from Tables XV and XVI, an increase in chain length resulted in a bathochromic shift of the absorption curve with an increase in intensity of absorption (a max) in the near infrared. In particular, compound XXXIV (Table XV) and XXXIX (Table XVI) had high absorptivity values, $a = 232$ and 124, respectively. In addition, it was observed that as the chain length was increased in both the above classes of compounds solubility and light stability decreased (Tables XVII and XVIII).

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These compounds were conveniently incorporated by milling and molding (Table XIX) or monomer casting (Table XX) with some difficulty as the chain length was increased due to increased insolubility in the plastic. In particular, compound XXXVIII (Table XVI) could not be incorporated by either milling and molding or monomer casting due to instability to heat and curing catalyst.

These compounds are also known to be easily burnished onto plastics.[22] Attempts at vacuum evaporation were again unsuccessful with decomposition taking place.

Another example of a polymethine that was evaluated was that obtained from Guaiazulene, compound XLI. This derivative showed intense absorption in the infrared, $a = 487$ at 869 mμ, with relatively very little visible absorption (Fig. 26). This compound, however, was rather insoluble and consequently could not be monomer cast and gave a hazy milled and molded sample (Table XIX). In addition, its light stability was rather low as were other members of this general class of compounds (Table XVIII).

D. Metal Complexes

The nickel complex of N-phenyl-N'-(2-methoxyphenyl)-C-phenyl-formazan was prepared and evaluated during the course of this investigation. This compound showed an absorption in the near infrared ($a = 13.2$ at 830 mμ) but also possessed a visible band ($a = 23.5$ at 400 mμ) and consequently was too highly colored to be useful (Fig. 27 and 28). However, its very good light stability was of interest (Table XXI). This derivative was also very compatible in plastics and could be easily milled and molded, monomer cast, or burnished (Tables XXII, XXIII and XIV).

Several other metal complexes that showed near-infrared absorption were uncovered in our screening program. These are shown below:

a) Mn complex of β-nitroso-α-naphthol ($a = 2.63$ at 825 mμ)

b) Dihydroxygermanium phthalocyanine (λmax = 925 mμ) (Fig. 29 and 30). This compound, although too highly colored to be useful, showed very good light stability with 88% of its infrared activity remaining after 500 hours exposure in a Fade-Ometer (Table XXV and Fig. 30).

c) Vanadyl phthalocyanine (λmax = 830 mμ). This compound, which has a visible absorption peak at 680 mμ, has its maximum transmission at 525 mμ with a resulting blue-green color (Fig. 31).

The absorption properties and light stability of the above compounds are compared along with some other organic and inorganic pigments in Tables XXV and XXVI.

As can be seen from Table XXV an advantage of pigmented type absorbers is their increased light stability.

E. **Miscellaneous Compounds**

Several phenothiazine derivatives were prepared and evaluated. As can be seen from Table XXVII and Fig. 32-34, their infrared absorption occurs in the very near infrared along with a relatively strong visible absorption in each case. Because of their relatively poor absorption, no application work was done on this general class of compounds.

In addition to the above classes of compounds, other miscellaneous compounds were prepared and screened. These compounds showed little or no near-infrared absorption. Their syntheses are summarized in the syntheses section of this report.

IV. SYNTHESES

A. Substituted Phenylfluorenol Salts

3,6-Bis(dimethylamino)-9-phenylfluoren-9-ol (V), was initially prepared by a modification of the method of Barker & Barker[2].

$(CH_3)_2N$ — ⬡ — CH_2 — ⬡ — $N(CH_3)_2$ $\xrightarrow{\text{Nitration}}$

I

II

III

IV

V
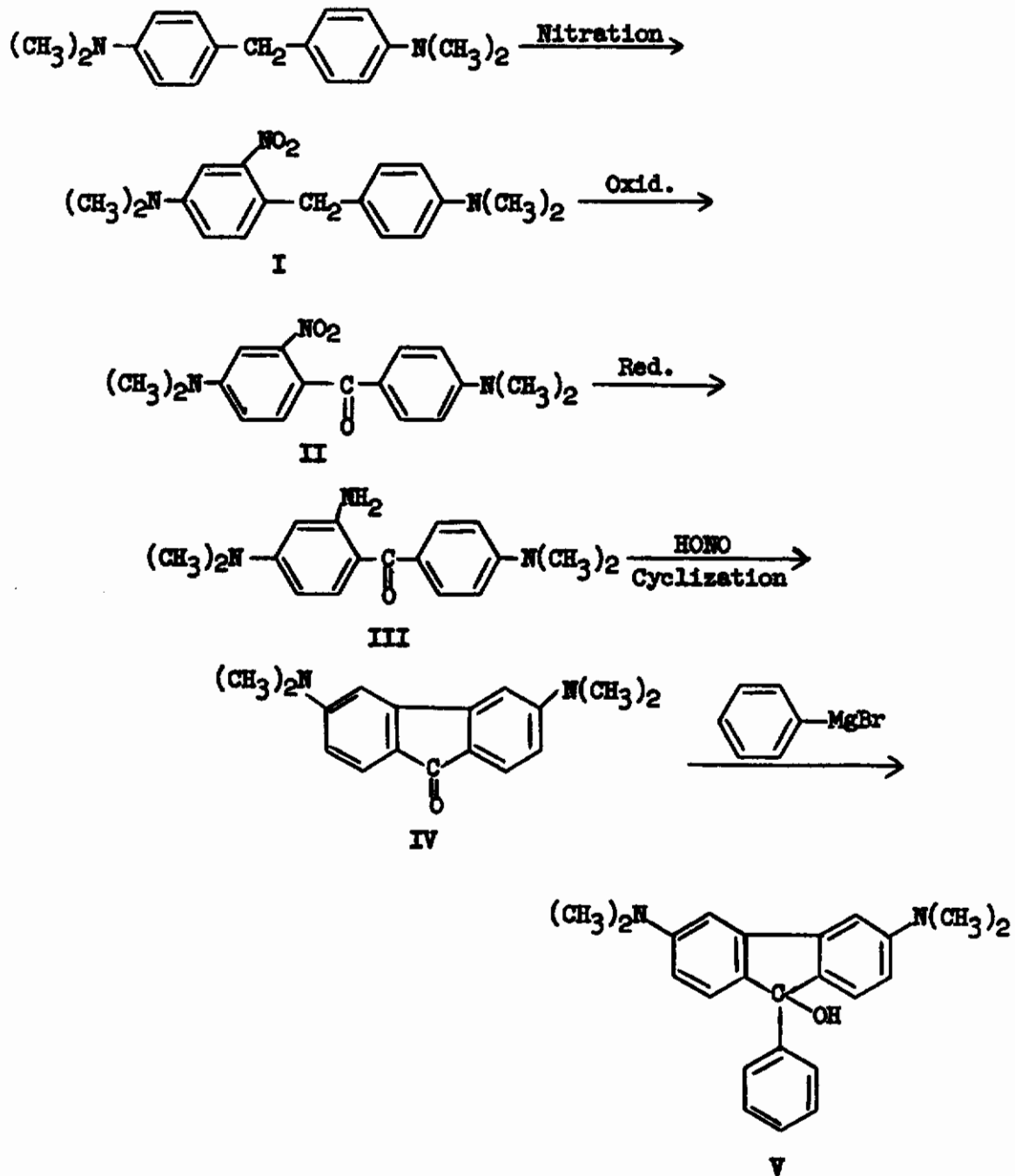
The poorest yield in the above sequence occurs in the cyclization step (diazotization with nitrosylsulfuric acid and thermal cyclization in 88% sulfuric acid). The maximum yield of IV obatined was 10-12% and reproducibility was difficult.

During the course of this investigation a new and more convenient method was found for the preparation of the intermediate fluorenone (IV).
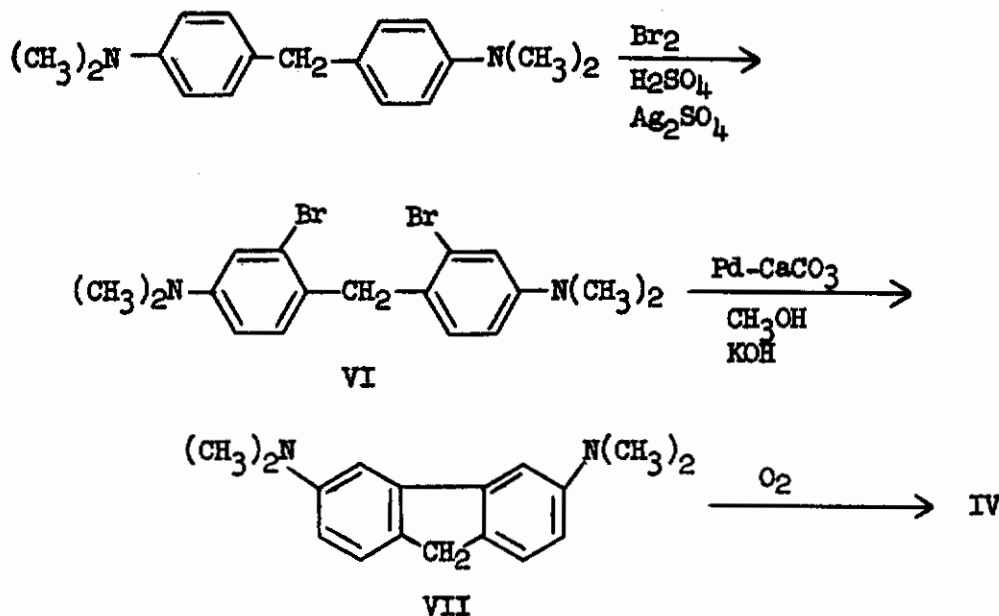
The dibromo compound was obtained in average yields of 60% by bromination in concentrated H_2SO_4 in the presence of Ag_2SO_4. Ring closure was effected in average yields of 15-20% by refluxing in methanolic KOH in the presence of 1% Pd on $CaCO_3$. This use of methanol as both the solvent and reducing agent was successful although Barker & Barker[2] reported negative results with Pd on $CaCO_3$, hydrazine and KOH. Air oxidation to the fluorenone (IV) was conveniently carried out in pyridine in the presence of base (Triton-B) in average yields of 85%.

Using the same sequence of reactions shown above, 3,6-bis(diethyl-amino)-9-phenylfluoren-9-ol was obtained in a crude state. Its spectra in acetic acid showed the same absorption peaks as the dimethylamino analog.

3-Dimethylamino-9-(p-dimethylaminophenyl)fluoren-9-ol (XI) was obtained as shown on the following page:

VIII

IX

X

XI
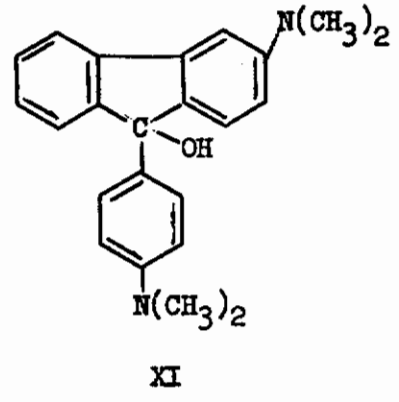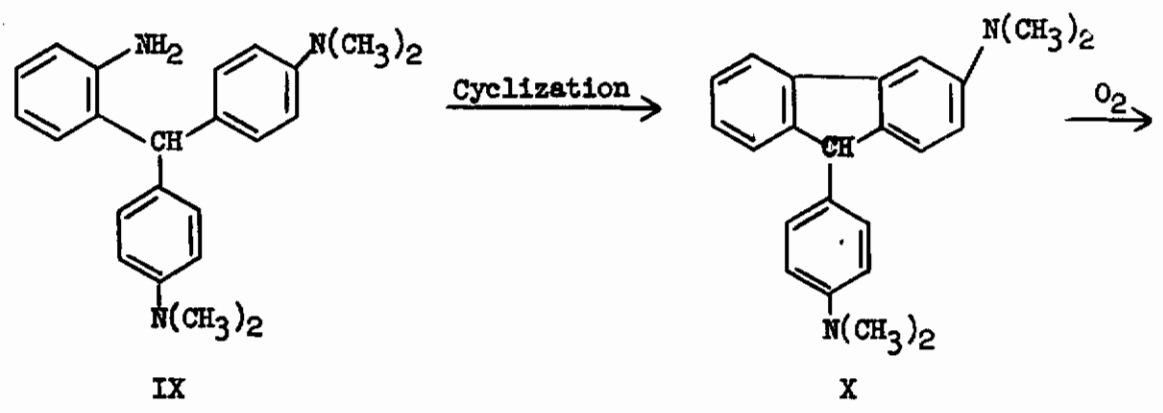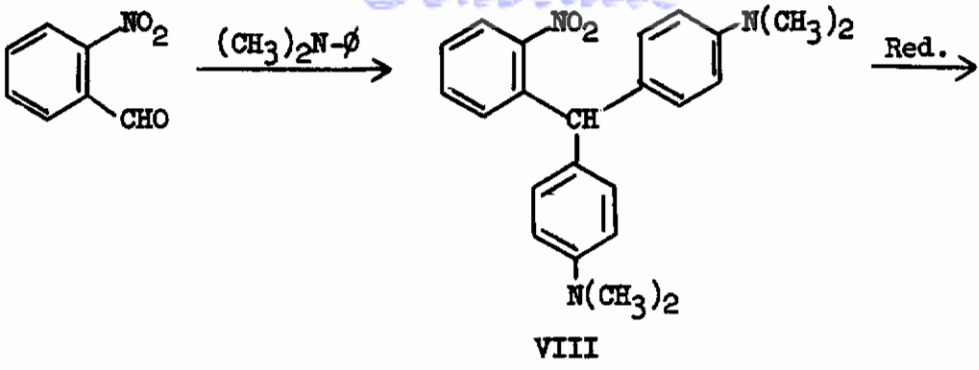
The desired fluorenol salts were then obtained, in most cases, by treatment with the corresponding acid.

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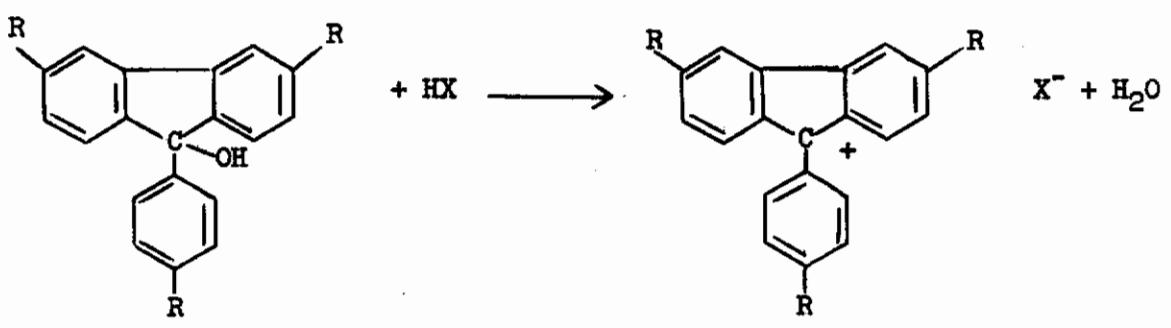
The acetate salts were not isolated but were prepared in situ by dissolving the fluorenol in acetic acid.

The results of the above preparations are summarized in Tables I and II. The observed absorptivity (a) values at 960 mμ (Table I) are compared with those calculated for the pure salt.

Experimental

2-Amino-4,4-bis(dimethylamino)benzophenone (III). - A supply of this compound had been prepared in these laboratories* prior to the start of this investigation by nitration of 4,4'-bis(dimethylamino)diphenylmethane, oxidation with chloranil to the corresponding benzophenone, and reduction with $SnCl_2$ to the amino compound, m.p. 206-208°C.

4,4'-Bis(dimethylamino)-2,2'-dibromodiphenylmethane (VI). - 4,4'-Bis-(dimethylamino)diphenylmethane (25.4 gm, 0.1 mole), silver sulfate (31.2 gm, 0.1 mole), and 300 cc of concentrated sulfuric acid were combined in a 500 cc flask equipped with a stirrer and dropping funnel. Bromine (32.0 gm, 0.2 mole) was added slowly keeping the temperature below 50°C. The mixture was stirred at room temperature for 3 hours and then filtered from the precipitated silver bromide. The filtrate was carefully poured onto ice water containing 2-3 gm of sodium sulfite. The solution was then neutralized carefully with concentrated ammonium hydroxide. The tan product which separated was filtered off, washed with water, and recrystallized from 500 cc of ethanol to give 25.9 gm of product, m.p. 102-104°C., reported[14] 103-104°C.

3,6-Bis(dimethylamino)fluorene (VII). - 4,4'-Bis(dimethylamino)-2,2'-dibromodiphenylmethane (103 gm, 0.25 mole) was added to one liter of methanol in a two-liter three-necked round bottom flask fitted with a reflux condenser and a mechanical stirrer. To the above was added 100 cc of 10 N KOH solution plus 30 gm of 1% Pd on $CaCO_3$ catalyst. The mixture was stirred and refluxed for 6 hours.

The hot reaction mixture was filtered and the filtrate cooled in an ice bath. The solid that separated was dissolved in 600 cc of boiling ethanol, filtered and the filtrate allowed to cool slowly to room temperature. The product was obtained as white needles weighing 12.4 gm, m.p. 151-153°C., reported[2] 149°C.

The 1% Pd on $CaCO_3$ used above was prepared by adding a hot aqueous solution of $CaCl_2$ (111 gm, 1 mole) to a hot aqueous solution of Na_2CO_3 (106 gm, 1 mole) with stirring. The precipitated $CaCO_3$ was filtered off, washed well with water and then suspended in about one liter of water.

Meanwhile, 2 gm of $PdCl_2$ was mixed with 5 cc of concentrated HCl and 25 cc of water and heated on a steam bath until solution was complete. The solution was diluted with 100 cc of water, made almost neutral with 5 N NaOH, and then added with stirring to the above suspension of $CaCO_3$. The mixture was warmed and stirred until all the Pd was precipitated as the

*Susi, P. V., American Cyanamid Company, unpublished results (1959).

hydroxide (oxide) on the CaCO$_3$ (the supernatant liquid was colorless). The solid was filtered off, washed well with distilled water, until the washings were chloride free, then with acetone and dried. The weight of product obtained was 97 gm.

3,6-Bis(dimethylamino)fluorenone (IV).

1. From the Aminobenzophenone (III). - In general, 8.72 gm (0.031 mole) of 2-amino-4,4'-bis(dimethylamino)benzophenone was dissolved in a mixture of 51 cc of concentrated H$_2$SO$_4$ and 18 cc of water. The solution was cooled and diazotized at 0-5°C., with nitrosylsulfuric acid prepared from 2.23 gm (0.0314 mole) of 97% pure NaNO$_2$ and 25 cc of concentrated H$_2$SO$_4$. The mixture was then heated at 70-80°C. for 2-3 hours until the evolution of gas ceased.

The reaction mixture was diluted with twice its volume of water and made basic by the careful addition of concentrated NH$_4$OH solution. The solid that separated was filtered off and dried. The reaction product was then dissolved in 60 cc of boiling ethanol containing 1 gm of KOH per 15 cc of alcohol. The orange solid that separated upon cooling was filtered off.

The crude products from several experiments were combined and recrystallized from alcohol, m.p. 248-250°C., reported[2] 249-250°C. (Average yield 11%.)

2. From the Fluorene (VII). - 3,6-Bis(dimethylamino)fluorene (20 gm, 0.08 mole) was dissolved in one liter of dry pyridine. Eight cc of Triton B (35% benzyltrimethylammonium hydroxide in methanol) was added. Air was then bubbled through the resulting solution for several hours.

The reaction solution was then heated to boiling, water was added until precipitation just started, and the solution was cooled. The product was filtered off and dried. Brick red platelets were obtained weighing 18.7 gm and melting at 255-256°C.

3,6-Bis(dimethylamino)-9-phenylfluoren-9-ol (V).
- 3,6-Bis(dimethylamino)fluorenone (10 gm, 0.0376 mole) was added to a refluxing solution of phenyl magnesium bromide, prepared from 33 gm (0.210 mole) of bromobenzene and 4.9 gm (0.200 mole) of Mg turnings, in 400 cc of ether and 400 cc of benzene. The mixture was then stirred and refluxed for 3 hours.

The reaction mixture was stirred into 3 times its volume of water and then ammonium chloride was added to dissolve the Mg(OH)$_2$ which separated. The benzene-ether layer was removed and the aqueous slurry was extracted with warm benzene. The combined extracts were dried over Na$_2$SO$_4$ and then evaporated down to a volume of about 300 cc. Petroleum ether (600 cc) was added and the mixture was cooled in ice. There was obtained 10.0 gm of a light tan solid melting at 196-197°C. Recrystallization from aqueous acetone raised the melting point to 209-210°C., reported[2] 209-210°C.

Salts of 3,6-bis(dimethylamino)-9-phenylfluoren-9-ol. Oxalate Salt (XIII). - The above fluorenol (2.0 gm, 0.00582 mole) was dissolved in a hot solution of 1.2 gm (0.00953 mole) of oxalic acid in 70 cc of water. To the dark colored hot solution was added 0.12 gm of ammonium oxalate in 2 cc of water. The resulting solution was allowed to cool slowly to room temperature.

The dark green precipitate that separated was filtered off, washed with cold water and then with acetone and dried under vacuum. There was obtained 2.0 gm of product melting at 164-166°C.

Calculated for $C_{52}H_{50}N_4O_{12}$: C, 67.7; H, 5.5; N, 6.1; O, 20.8. Found: C, 67.6; H, 5.7; N, 6.0; O, 20.6.

Tetrachlorozincate Salt (XIV). - The fluorenol (1.0 gm, 0.00291 mole) was dissolved in a mixture of 100 cc of water and 4 cc of concentrated HCl. A solution of $ZnCl_2$ in 25 cc water containing a few drops of concentrated HCl was added, followed by the slow addition of a saturated NaCl solution. After precipitation had started, the mixture was allowed to stand overnight. The dark brown solid was then filtered off. It weighed 1.0 gm.

Phosphomolybdate Salt (XV). - A stock solution of phosphomolybdic acid was prepared by dissolving Na_2HPO_4 (0.852 gm, 0.006 mole) and $Na_2MoO_4 \cdot 2H_2O$ (17.4 gm, 0.072 mole) in 200 cc of water and adding enough concentrated HCl to bring the pH of the solution to about 3.0.

A solution of the fluorenol (1.03 gm, 0.003 mole) in 20 cc of acetic acid and 100 cc of water was warmed to 60-70°C. The stock solution above, 175 cc, was added with stirring. The mixture was then heated to boiling, 25 cc more of the stock solution added, and the mixture allowed to stand overnight. The brown solid which separated was filtered off, washed with water, acetone and dried at 50-60°C. There was obtained 2.6 gm of material that did not melt below 350°C.

Tetracyanonickelate Salt (XVI) - To a solution of 0.29 gm (0.001 mole) of $Ni(NO_3)_2 \cdot 6H_2O$ dissolved in 0.5 cc of water was added a solution of 2.7 gm (0.04 mole) of 96% KCN dissolved in 5 cc of water until the solid that originally precipitated redissolved and a red-yellow solution was obtained.

To the above was added 10 cc of water and 0.344 gm (0.001 mole) of the fluorenol dissolved in 5 cc of water containing 1 cc of 1 N HCl (0.001 mole). The dark green solid that separated was washed well with water and dried.

Fluoborate Salt (XVII) - To a solution of the fluorenol (1.72 gm, 0.005 mole) dissolved in 300 cc of hot ethanol was added 1 gm of 50% aqueous HBF_4 with stirring. The mixture was cooled and the brown solid that separated was collected, washed with ether and dried. It weighed 1.4 gm and melted with decomposition at 295-300°C.

Calcd. for $C_{23}H_{23}N_2BF_4$: C, 66.8; H, 5.6; N, 6.8. Found: C, 66.3; H, 6.0; N, 6.6.

Miscellaneous Salts (XVIII-XXIII) - The fluorenol (4.47 gm, 1.3×10^{-2} mole) was dissolved in 500 cc of benzene. To 50 cc (1.3×10^{-3} mole) of the above color base stock solution was added an equivalent amount of the corresponding acid dissolved in a small volume of benzene or alcohol. After standing overnight, the product was filtered off and dired.

In the case of the ethane sulfonic acid salt (XX), 70 cc of ether was added to induce precipitation. The diphenyl-4,4'-disulfonic acid salt (XXI) was obtained by evaporating the reaction solution to dryness and triturating the residue with ether.

2-Nitro-4'4"-bis(dimethylamino)triphenylmethane (VIII). - A mixture of 90.6 gm (0.60 mole) of o-nitrobenzaldehyde, 218.1 gm (1.80 moles) of dimethylaniline, and 102.3 gm (0.75 mole) of freshly fused $ZnCl_2$ were stirred and heated on a steam bath for 6 hours and then allowed to stand overnight.

The reaction mixture was steam distilled to remove the excess dimethylaniline and then cooled. The solid residue that remained was washed several times with water and then triturated with 900 cc of hot alcohol. After cooling, the solid that separated was filtered off and recrystallized from a mixture of 200 cc of benzene and 800 cc of ethanol to give 143 gm of product, m.p. 158-159°C., reported[12] 159-160°C.

2-Amino-4',4"-bis(dimethylamino)triphenylmethane (IX). - The above nitro compound (143 gm, 0.38 mole) was added portionwise to a mixture of $SnCl_2$ (360 gm, 1.6 moles) and 1100 cc of concentrated HCl at 70-80°C. After addition was completed the mixture was stirred and refluxed for 2 hours and then allowed to stand overnight.

The reaction mixture was filtered and the solid that was collected was dissolved in hot water, made strongly basic with sodium hydroxide and filtered. The crude product was recrystallized from one liter of alcohol to give 90 gm of a pale yellow solid, m.p. 131-132°C., reported[12] 134-135°C.

3-Dimethylamino-9-(p-dimethylaminophenyl)fluorene (X). - The above amino compound (90 gm, 0.26 mole) was dissolved in a mixture of 133 cc of concentrated H_2SO_4 and 272 cc of water. The solution was cooled and diazotized with a solution of 18.3 gm of 98% $NaNO_2$ in 115 cc of water at 0-5°C. The mixture was allowed to gradually warm up to room temperature and then heated on the steam bath until gas evolution ceased. After cooling, the mixture was made basic by the addition of dilute (1:1) NH_4OH solution. The solid that separated was removed, digested with a small amount of hot alcohol, cooled and filtered. The solid was then dissolved in a small amount of hot alcohol using enough concentrated HCl to cause complete solution. The mixture was cooled, made basic by the addition of alcoholic KOH solution, and the product that separated filtered off and recrystallized from benzene-alcohol to give 10.5 gm of pale yellow product, m.p. 155-156°C., reported[15] 158°C.

3-Dimethylamino-9-(p-dimethylaminophenyl)fluoren-9-ol (XI). - The above fluorene compound (10.2 gm, 0.031 mole) was dissolved in 450 cc of pyridine. Five drops of Triton B solution was added and air was bubbled through the mixture for 3 hours. The reaction mixture was then evaporated to dryness to give 10.7 gm of crude product as a dark brown oil that partially solidified on standing.

After repeated crystallizations from alcohol (Darco treatment included) the product was obtained as a white solid, m.p. 148-149°C.

Calc'd. for $C_{23}H_{24}N_2O$: C, 80.4; H, 7.1; N, 7.9. Found: C, 80.0; H, 6.9; N, 8.0.

B. Triarylaminium Free Radical Salts

The tris(p-dialkylaminophenyl)aminium salts were prepared by methods similar to those reported in the literature[23].
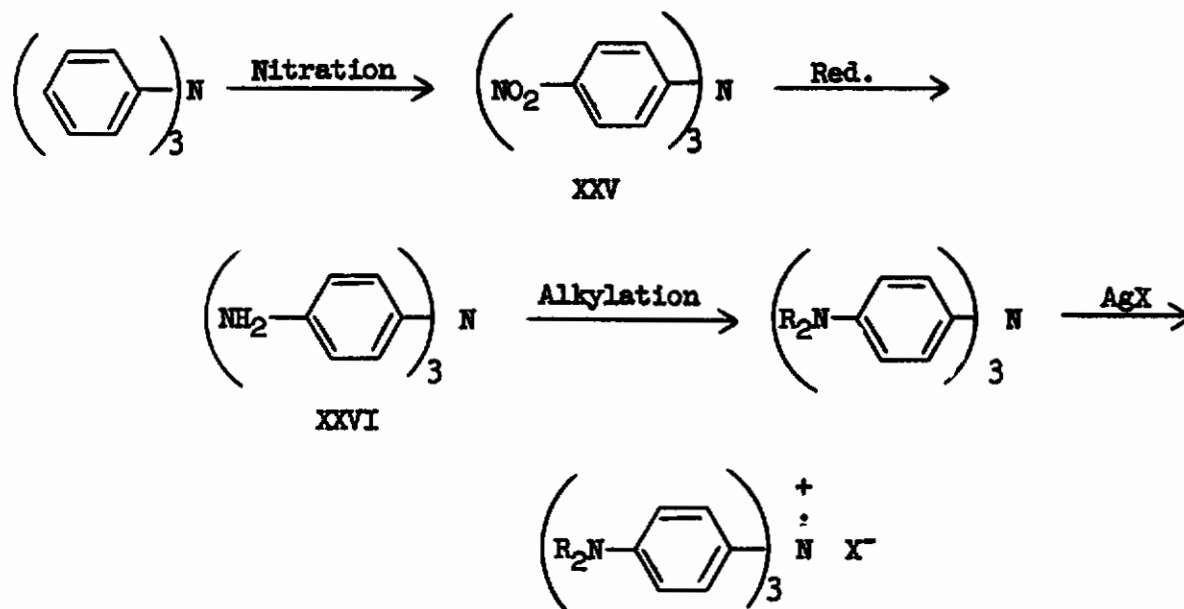
The lowest yields in the above sequence occurred in the alkylation step. In the case of the dimethyl or diethyl derivative, an intermediate quaternary salt was obtained from which the amine was liberated by refluxing with strong base.

The dibutyl derivative could not be obtained in a pure form. As a result the crude compound was oxidized to the aminium salt and evaluated as such.

The results of the above series of preparations are summarized in Table X. Although the aminium salts are relatively stable (shelf life), decomposition does occur on standing as can be seen from the absorption data obtained on the dimethyl derivative (XXVIII) after one month standing.

15

Tris(p-nitrophenyl)amine (XXV). - Triphenylamine (150 gm, 0.612 mole) was added to 800 cc of glacial acetic acid in a two-liter, three-necked round bottom flask fitted with a condenser and a stirrer. The mixture was stirred and heated to 60°C., and 90% fuming nitric acid (129 gm, 1.84 moles) was added dropwise so as to maintain the reaction temperature between 60-65°C. (During this addition, the reaction mixture turned from a green to a brown color.) After the addition was completed, the reaction mixture was stirred and heated at 65°C. for one hour, and then filtered hot. The crude solid was triturated several times with boiling acetic acid followed by boiling alcohol and then dried. There was obtained 150 gm of yellow solid melting at 260-265°C. The literature reports the melting point of this compound at 280°C.[16]; 349-350°C.[23]; 383°C.[29].

Tris(p-aminophenyl)amine (XXVI) - Tris(p-nitrophenyl)amine (148 gm, 0.39 mole) was added portionwise to a stirred solution of stannous chloride (1070 gm, 4.83 moles) in 1800 cc of concentrated hydrochloric acid at 60°C. After addition was completed, the mixture was refluxed overnight. (During the course of the reaction, the color of the reaction mixture changed from dark yellow to light yellow to green.)

The reaction mixture was then cooled and filtered. The light green solid that was collected was dissolved in hot water, filtered and then made strongly basic with sodium hydroxide. The solid that separated was filtered off, washed with dilute sodium hydroxide, then with water and dried. There was obtained 90 gm of a purple powder melting at 225°C., reported[16] 230°C.

Tris(p-dimethylaminophenyl)amine. - A mixture of 11.6 gm (0.04 mole) of tris(p-aminophenyl)amine, 51.1 gm (0.36 mole) of methyl iodide and 38.6 gm (0.28 mole) of potassium carbonate was stirred and refluxed overnight in 200 cc of 80% aqueous acetone. The mixture was then cooled and filtered. The solid that was collected was washed with 100 cc of hot 80% aqueous acetone, dried, and then added to 150 cc of sodium ethoxide solution. The resulting mixture was refluxed for 48 hours and then evaporated to dryness.

The residue was triturated with water and filtered. The dark purple solid that remained was recrystallized from heptane to give 4.2 gm of purple plates, m.p. 157-159°C., reported[24] 157-159°C.

Tris(p-diethylaminophenyl)amine. - By using ethyl iodide in the above procedure, the corresponding diethylamino derivative was obtained as a brown oil. The crude product was dissolved in 5 N HCl and the solution evaporated to dryness. The hydrochloride obtained was recrystallized repeatedly from alcohol-heptane mixture until it melted at 221-222°C. The salt was then dissolved in water, made basic with sodium hydroxide and extracted with heptane. Evaporation of the heptane solution left an oil that was dissolved in alcohol and cooled in dry-ice-acetone. The yellow solid that separated was recrystallized from alcohol, m.p. 105°C.

Calc'd. for $C_{30}H_{42}N_4$: C, 78.6; H, 9.2; N, 12.2. Found: C, 78.3; H, 9.2; N, 12.3.

<u>Tris(p-dibutylaminophenyl)amine</u>. - A mixture of 17.4 gm (0.06 mole) of tris(p-aminophenyl)amine, 49.3 gm (0.36 mole) of butyl bromide and 49.7 gm (0.36 mole) of potassium carbonate was stirred and refluxed in 300 cc of 80% aqueous acetone for eight days.

The acetone was distilled from the reaction mixture and the aqueous layer extracted with hot heptane. Evaporation of the heptane solution left 31 gm of a dark brown oil.

The crude oil was dissolved in 5 N HCl, treated with Darco, and then evaporated to dryness. The salt remaining was dissolved in hot heptane-alcohol, treated with Darco, and then evaporated to dryness.

The solid residue was dissolved in water, filtered from some insoluble material and made basic. The oil that separated was extracted with heptane, the extracts dried over Na_2SO_4, and then evaporated down. There was obtained 1.5 gm of brown oil.

<u>Tris(p-dimethylaminophenyl)aminium Perchlorate (XXVII)</u>. - To 3.74 gm (0.01 mole) of tris(p-dimethylaminophenyl)amine in 250 cc of dry acetone was added 45 cc (0.009 mole) of 0.2 N silver perchlorate solution in acetone. The mixture was stirred for 1/2 hour and then filtered from the precipitated silver. To the green colored filtrate there was added an excess of ether and the mixture was cooled. The product that separated as a green solid was collected and dried. It weighed 3.75 gm and decomposed by 165°C.

Calc'd. for $C_{24}H_{30}N_4ClO_4$: N, 11.8. Found N, 12.1.

<u>Tris(p-dimethylaminophenyl)aminium Fluoborate (XXVIII)</u>. - Tris(p-dimethylaminophenyl)amine (2.24 gm, 0.006 mole) was dissolved in 150 cc of acetone and 27 cc (0.0054 mole) of 0.2 N silver fluoborate solution in acetone was added. After stirring for 1/2 hour, the mixture was filtered and the filtrate diluted with 700 cc of ether. There was obtained 1.73 gm of green solid melting with decomposition at 155-156°C.

Calc'd. for $C_{24}H_{30}N_4BF_4$: C, 62.5; H, 6.6; N, 12.2. Found: C, 62.6; H, 6.7; N, 12.3.

<u>Tris(p-diethylaminophenyl)aminium Fluoborate (XXX)</u>. - To 0.6 gm (0.0013 mole) of tris(p-diethylaminophenyl)amine in 30 cc of acetene was added 6 cc (0.0012 mole) of 0.2 N silver fluoborate solution in acetone. After standing overnight, the mixture was filtered, and the filtrate evaporated to dryness. The product remained as a green solid which decomposed by 140°C.

Calc'd. for $C_{30}H_{42}N_4BF_4$: N, 10.3. Found: N, 10.5.

<u>Tris(p-diethylaminophenyl)aminium Perchlorate (XXIX)</u>. - Prepared by the same procedure as the fluoborate salt above using 6 cc of 0.2 N silver perchlorate solution in acetone. The product was obtained as a glassy green solid.

by the same method as the other derivatives described above. There was
obtained from 1.4 gm of the amino compound, 1.5 gm of the product as a
dark green semisolid.

C. Polymethines

1. Tetraphenylvinylcarbonium Salts
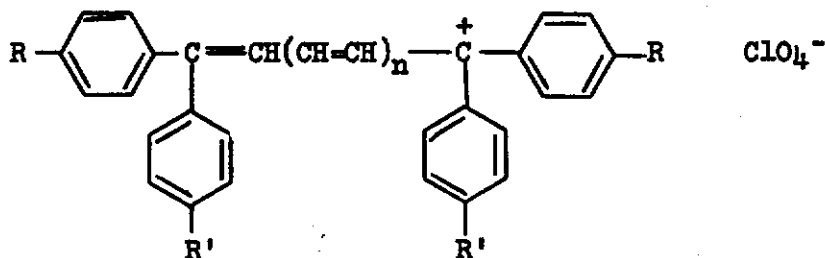
Members of this class of compounds were prepared by known litera-
ture procedures[19], by reaction of a substituted 1,1-diphenylethylene with
the corresponding ketone (n=0), ethyl orthoformate (n=1), or 1,3,3-tri-
methoxypropene (n=2) in the presence of perchloric acid.

In the cases where $R=(CH_3)_2N$ and $R'=(CH_3)_2N$ or H, (XXXI-XXXV)
the reactions proceeded smoothly. Difficulties were encountered in
attempts to prepare the methoxy (CH_3O) analogs of the above. Only where
$R=R'=CH_3O$ and n=2 (XXXVI) was a pure product obtained successfully.

The results obtained are summarized in Table XV.

Experimental

General Procedure. - 1,1-Bis(p-dimethylaminophenyl)ethylene[27]
was obtained by reaction of methyl magnesium iodide with 4,4'-bis(dimethyl-
amino)benzophenone. 1,1-Bis(p-methoxyphenyl)ethylene[10] was obtained by
reaction of methyl magnesium iodide with 4,4'-dimethoxybenzophenone.
1-(p-Dimethylaminophenyl)-1-phenylethylene[11], m.p. 56-57°C., was obtained
by reaction of 4-dimethylaminobenzophenone[13] with methyl magnesium iodide
followed by thermal dehydration of the resulting carbinol.

An illustrative procedure is shown in the preparation of 1,1,5,5-
tetrakis(p-dimethylamino)divinylcarbonium perchlorate (XXXII). To a
mixture of 22.1 gm (0.0831 mole) of 1,1-bis(p-dimethylaminophenyl)ethylene
and 8.3 cc of ethyl orthoformate in 50 cc of acetic anhydride was added
5.6 gm of 72% aqueous perchloric acid, dissolved in 50 cc of acetic
anhydride. The resulting dark blue mixture was stirred a few minutes and
then allowed to stand at room temperature for 2 hours. An additional
8.3 cc of ethyl orthoformate was added, the mixture stirred and then
allowed to stand an additional hour at room temperature. Excess ether was
added and the solid that separated was filtered off, washed well with
acetic acid, then with ether and dried. There was obtained 16 gm of
product melting with decomposition at 224°C., reported[19] 228-230°C.

1,1,7,7-Tetrakis(p-methoxyphenyl)trivinylcarbonium Perchlorate

(XXXVI). - To 2.4 gm (0.01 mole) of 1,1-bis(p-methoxyphenyl)ethylene was added 1 cc of 1,3,3-trimethoxypropene and 5 cc of acetic anhydride. A solution of 0.65 gm of 72% perchloric acid in 10 cc of acetic anhydride was then added and the mixture was stirred for 5 minutes and then cooled. The mixture was filtered and the solid that was collected was washed with ethanol and ether. There was obtained 0.92 gm of brown crystals melting with decomposition at 187-188°C.

Calc'd. for $C_{35}H_{33}ClO_8$: C, 68.1; H, 5.4; Cl, 5.8; O, 20.8. Found: C, 68.0; H, 6.0; Cl, 6.0; O, 20.5.

2. Xanthylomethines
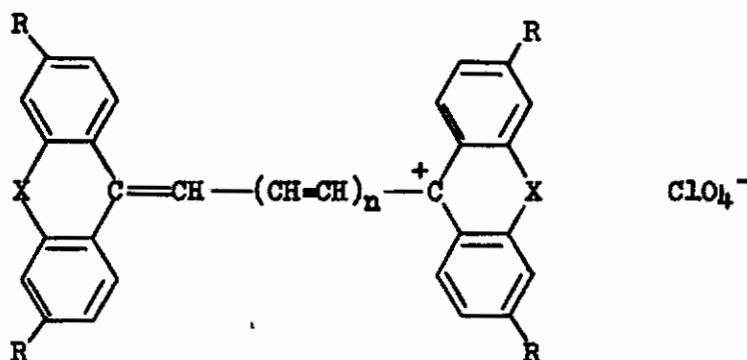
The parent members of this class of compounds (R=H; X=O) were reported by Wizinger and Arni[31]. It was of interest during the course of this investigation to prepare these and other derivatives /R=CH_3O, (CH_3)_2N; X=O,S/. The general route investigated is shown below:
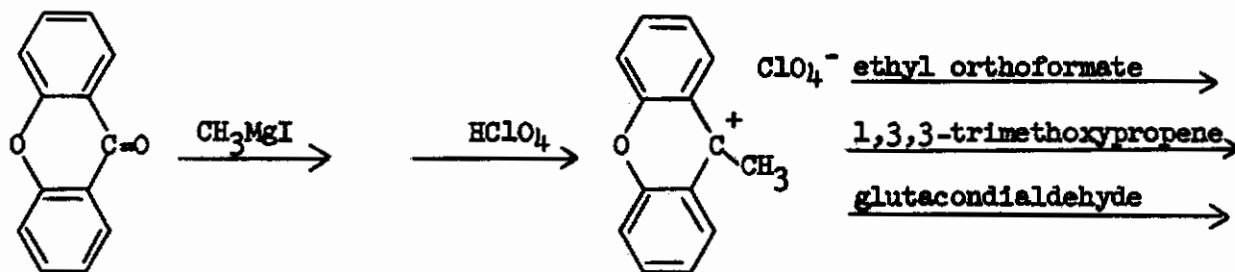
$$\longrightarrow \text{Product (where } n=1)$$
$$\longrightarrow \text{Product (where } n=2)$$
$$\longrightarrow \text{Product (where } n=3)$$

In the cases where R=H, and X=O, the products (n=1, 2 and 3) were easily obtained (XXXVII-XXXIX). Difficulties were encountered where R=(CH_3)_2N and X=S, O or where R=CH_3O, X=O. Only one member of the above derivatives was obtained successfully (XL) (where R=CH_3O, X=O, n=3).

The results of these preparations are shown in Table XVI.

Experimental

Bis(xanthene)trimethine Perchlorate (XXXVII). - Methyl magnesium bromide from 12 gm Mg, 50 gm CH_3Br, and 200 cc of ether was treated with 20 gm of xanthone in 400 cc of dry benzene. The mixture was stirred and refluxed for 4 hours and then allowed to stand overnight. After decomposition with ice and HCl, the organic layer was removed, dried over anhydrous sodium sulfate and then treated with 15 cc of 70% perchloric acid. The solid that separated was filtered off, washed with alcohol to give 19.1 gm of 9-methylxanthylium perchlorate as red crystals, melting with decomposition at 202-206°C., reported[31] 202-204°C.

The above perchlorate (2.95 gm 0.01 mole) was dissolved in 50 cc of hot 2:1 acetic acid - acetic anhydride. After the addition of 1.5 cc of ethyl orthoformate, the mixture was heated on the steam bath for several minutes. It was then cooled and filtered to give 2.6 gm of green crystals melting with decomposition at 219-221°C., reported[31] 233-235°C.

Bis(xanthene)pentamethine Perchlorate (XXXVIII). - 9-Methylxanthylium perchlorate (2.95 gm, 0.01 mole) was dissolved in 50 cc of hot 2:1 acetic acid - acetic anhydride and then allowed to cool to room temperature. There was added 1.3 cc of 1,3,3-trimethoxypropene and the mixture was allowed to stand at room temperature for one hour. The reaction mixture was then filtered to give 1.1 gm of black crystals melting with decomposition at 270-274°C., reported[31] 234-237°C.

Bis(xanthene)heptamethine Perchlorate (XXXIX). - 9-Methylxanthylium perchlorate (2.95 gm, 0.01 mole) was dissolved in 60 cc of boiling methanol. The sodium salt of glutacondialdehyde[3,26] (0.6 gm, 0.005 mole) was added followed by 2 cc of 72% perchloric acid. The mixture was stirred and refluxed for several minutes and then allowed to cool to room temperature. The solid that separated was filtered off, recrystallized from 1:1 acetic acid - acetic anhydride to give 0.3 gm of brown solid melting with decomposition 199-200°C., reported[31] 190-193°C.

Bis(3,6-dimethoxyanthene)heptamethine Perchlorate (XL). - 3,6-Dimethoxyxanthone, m.p. 184-186°C., reported[1] 180°C., was obtained by the thermal dehydration of 4,4'-dimethoxy-2,2'-dihydroxybenzophenone. Reaction with methyl magnesium iodide followed by the addition of perchloric acid gave the corresponding 9-methylxanthylium perchlorate.

The above salt (0.74 gm, 0.0021 mole) was dissolved in 30 cc of hot methanol. Sodium glutacondialdehyde (0.13 gm, 0.0011 mole) was added followed by 15 cc of perchloric acid. The mixture was refluxed for 15 minutes, cooled and filtered. There was obtained 0.22 gm of brown solid that, after recrystallization from methanol, melted at 201-202°C.

Calc'd. for: $C_{37}H_{31}ClO_{10}$: C, 66.2; H, 4.6; Cl, 5.3; O, 23.9.
Found: C, 66.0; H, 4.8; Cl, 5.1; O, 23.8.

3. Azulene Derivative

Prepared by the reaction of Guaiazulene (1,4-dimethyl-7-isopropyl-azulene) with glutacondialdehyde in the presence of perchloric acid.

XLI
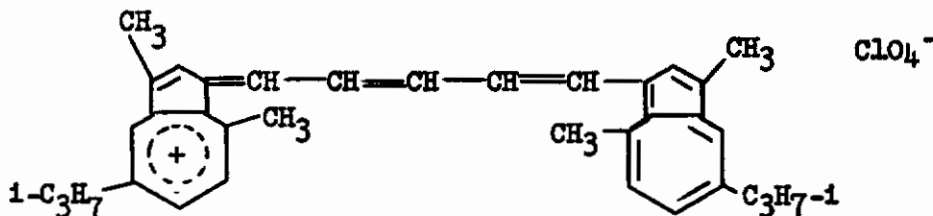
λ_{max} (mμ)   860
a_{max} 487

Experimental

Guaiazulene (5.94 gm, 0.03 mole) and the sodium salt of gluta-condialdehyde (1.8 gm, 0.015 mole) were dissolved in 180 cc of methanol. To the deep blue solution was added 5 cc of 72% HClO$_4$. The resulting purple solution was stirred at room temperature for 10 minutes, boiled for 5 minutes and cooled. The solid that separated was collected, washed with a small quantity of alcohol and dried to give 1.65 gm of brown solid which did not melt below 300°C. The literature[20] reports the compound as a brown solid which does not melt below 280°C. Recrystallization of the above from 2:1 acetonitrile-methanol gave 1.1 gm of a coppery brown solid that melted with decomposition at 245°C. (rapid heating).

D. Metal Complexes

Only one metal complex was synthesized during the course of this work which showed appreciable infrared absorption. The nickel comples of N-phenyl-N^1-(2-methoxyphenyl)-C-phenylformazan was obtained from diazotized o-anisidine and benzaldehyde phenylhydrazone followed by complexing with nickel.
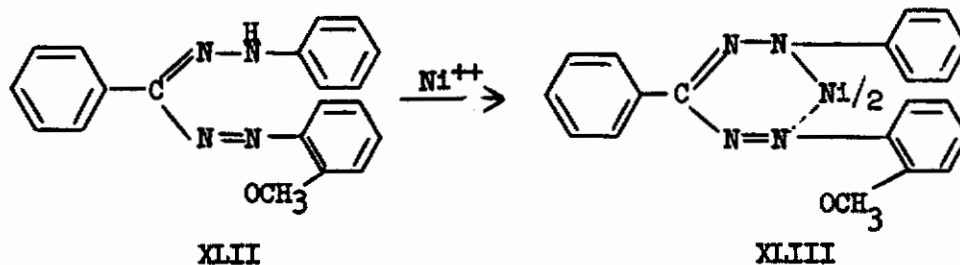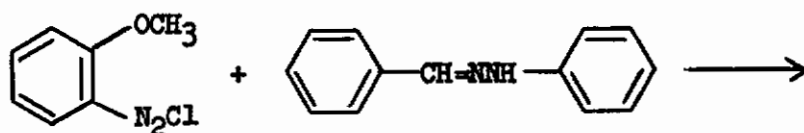
$$\lambda(m\mu) \quad 400 \quad 830 \text{ (max)}$$
$$\alpha \quad\quad 23.5 \quad 13.2$$

An attempt to prepare the corresponding manganese derivative by the above method was unsuccessful.

Experimental

N-Phenyl-N'-(2-methoxyphenyl)-C-phenylformazan (XLII). - o-Anisidine (24.6 gm, 0.2 mole) in 100 cc of 5 N HCl was diazotized at 0-5°C. with 14.1 gm (0.2 mole) of 98% NaNO$_2$ in water. Benzaldehyde phenylhydrazone (39.2 gm, 0.2 mole) was dissolved in 2 liters of alcohol and cooled to 0°C. The diazonium solution and the hydrazone solution were added simultaneously with rapid stirring to 60 gm of KOH in 300 cc of alcohol at 15°C. The mixture was allowed to stand overnight.

The reaction mixture was made acidic with acetic acid and filtered. The crude solid was washed with water (50°C) and then recrystallized from 1250 cc of ethanol to give 13.0 gm of product m.p. 132°C., reported[17] 129°C.

Nickel Complex of N-Phenyl-N'-(2-methoxyphenyl)-C-phenylformazan (XLIII). - To 3.3 gm (0.01 mole) of the above formazan in 100 cc of ethanol was added 2.9 gm (0.01 mole) of nickel nitrate hexahydrate dissolved in a small volume of alcohol and 60 cc of concentrated alcoholic NH$_3$ solution. The mixture was stirred and heated for 2 hours and then filtered hot. The solid collected was washed with warm 2.5% acetic acid and then repeatedly with boiling alcohol until the washings were pale yellow. The product was dried and gave 3.2 gm of black solid melting with decomposition at 262°C., reported[17] 273°C.

E. Miscellaneous Compounds

1. Phenothiazine Derivatives

Members of this class of compounds were prepared by reported procedures[32] by reaction of the corresponding benzophenone (or its dichloride) with phenothiazine in the presence of $ZnCl_2$ and $POCl_3$, and isolation of the products as the fluoborates. Results are shown in Table XXVII.

Experimental

An illustrative procedure[32] is shown in the preparation of compound (XLVI). A mixture of 7.26 gm (0.03 mole) of 4,4'-dimethoxybenzophenone, 6 gm of freshly fused $ZnCl_2$, and 30 cc of $POCl_3$ was heated at 90°C. for one-half hour. Phenothiazine (5.97 gm, 0.03 mole) was then added and the solution was refluxed gently for 2 hours. The dark reaction mixture was cooled to about 60°C., and carefully poured into an excess of water (250 cc) at 50-60°C. with stirring and external cooling as necessary. The mixture was then cooled and the water decanted from the dark hard mass that remained. The crude solid was then taken up in boiling alcohol and 10 cc. of 50% HBF_4 solution was added. The mixture was cooled and the solid that separated was collected, washed with ether and dried to give 8.0 gm of dark blue material.

2. 3,6-Bis(dimethylamino)-9-phenylfluoren-9-nitrile (XLVIII).
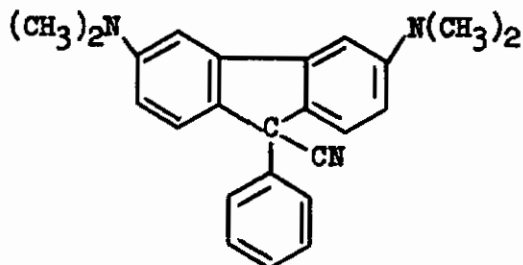
Three grams of the tetrachlorozincate salt (XIV) of the corresponding fluorenol was dissolved in warm water and excess KCN solution was added. The solid that separated was washed with dilute KCN solution, then with water and recrystallized from alcohol. There was obtained 1.5 gm of light tan solid, m.p. 240-242°C.

Calc'd. for $C_{24}H_{23}N_3$: C, 81.6; H, 6.6; N, 11.9. Found: C, 81.6; H, 6.6; N, 11.7.

Irradiation of methanol or DMF solutions of the above with UV light gave solutions with very weak near infrared absorption.

3. Di/perimidyl-(2)/methane
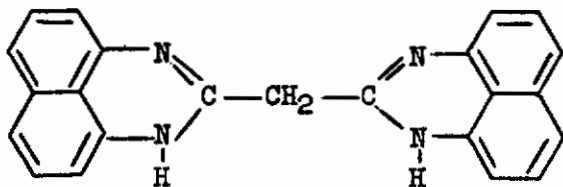
Obtained by reaction of 1,8-diaminonaphthalene with diethyl-malonate[25]. This compound showed no absorption in the near infrared.

4. Metal Complexes of Pyridine N-Oxide

These compounds were prepared by reaction of the corresponding metal perchlorate with pyridine N-oxide[4].

$$[Cu(C_5H_5NO)_4] (ClO_4)_2$$

λ_{max} (mμ)   750

a_{max} 0.188

$$[Ni(C_5H_5NO)_4] (ClO_4)_2$$
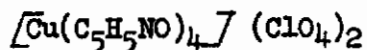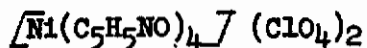
λ_{max} (mμ)   1100

a_{max} 0.016

5. Nickel Complex of Triphenyl Phosphine

$$(\phi_3P)_2NiBr_2$$
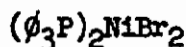
The above was obtained as a green solid by reaction of $NiBr_2 \cdot 3H_2O$ and triphenyl phosphine[28] and showed only very weak absorption in the near infrared.

λ_{max} (mμ)   925   620

a_{max} 0.18   0.18

6. Free Radical of 4,4'-Bis(2,6-di-t-butylhydroxyphenyl)amine
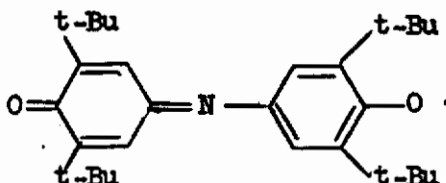
Prepared by the method reported by Coppinger[8], this compound showed no absorption in the near infrared.

24

7. Oxidation Studies

a. Tris(p-dimethylaminophenyl)amine was oxidized with dichromate and the product isolated as the $ZnCl_2$ salt. Its solution in water showed the following absorption peaks, but the product itself decomposed on standing.

λ_{max} (mμ)	a_{max}
895	130
790	91
400 (not max)	15.2

b. 2,7-Bis(dimethylamino)fluorene* was oxidized with silver perchlorate. The salt obtained showed the following absorption, but decomposed on standing.

λ_{max} (mμ)	a_{max}
970	69.9
850	33.7
455	62.0

8. Pyrylocyanines
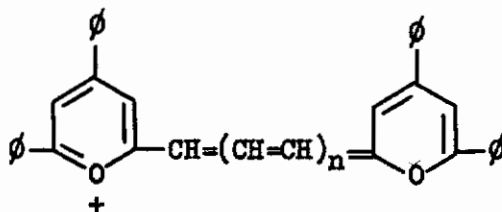
Attempts to prepare the above compounds by reaction of 2-methyl-4,6-diphenylpyrylium perchlorate[9] with ethyl orthoformate (n=1), 1,3,3-trimethoxypropene (n=2), or glutacondialdehyde (n=3) were unsuccessful. In the case where n=1, the crude product obtained showed weak absorption peaks at 800 (a = 2.9) and 725 mμ (a = 3.1) but the material could not be obtained in a pure form.

9. Unsuccessful Preparations

In addition to the syntheses reported above, unsuccessful attempts were made to prepare several other compounds that might show near infrared absorption. As a matter of record the desired final compounds are shown on the following page:

*Susi, P. V., American Cyanamid Company, Unpublished results (1959).
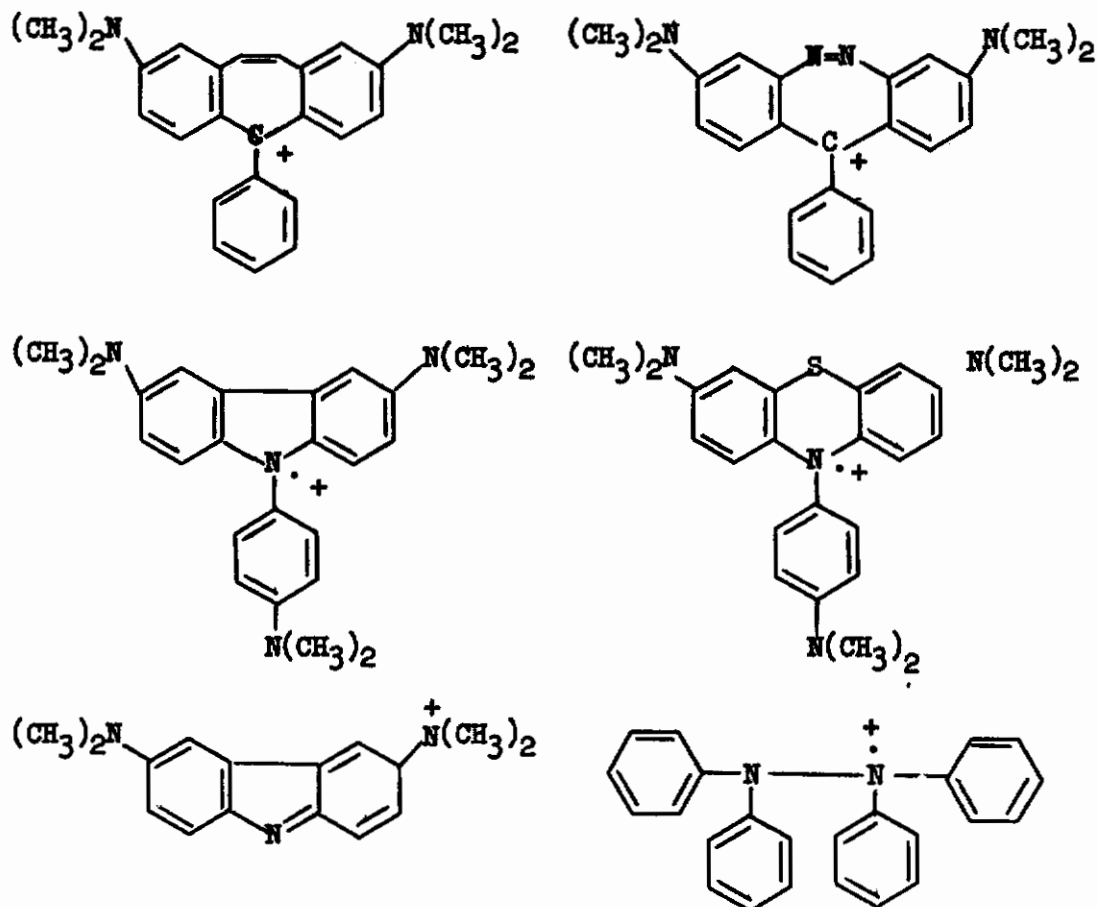
V. EVALUATION AND APPLICATION

A. Screening

The compounds screened for near-infrared absorption properties were obtained not only from the synthesis effort in this project but also from files of other research programs within the company and, for the most part, from the company's growing central or "CL" file of compounds. For the past five or six years a great many compounds in the file have been tested for near-infrared absorption in the course of previous interest in this area of research. For the Government-sponsored program, the perusal of the CL file was continued with at least three co-workers making selections from several thousands of compounds to be screened from the same distribution list of newly submitted samples to the file. The following types of substances were chosen: (1) green solids, (2) charged resonating structures, (3) metallo-organic compounds containing especially iron, manganese, nickel and copper, and (4) several phthalocyanines.

The actual testing entailed dissolving enough of the substance in a few milliliters of a suitable solvent to produce fairly strong color, as most of the compounds were colored. The colorless compounds were dissolved in large excess. The usual solvent used was spectro-quality

dimethylformamide (DMF). However, when interaction was suspected or when unexpected color changes occurred with DMF, other solvents such as 3A alcohol, methanol, or acetone were used. The spectral absorption curves of these solutions were obtained by use of a Beckman DK-2 recording spectrophotometer operating over the range 400 to 1700 mμ with the corresponding solvents used as references.

Of the better than 500 compounds selected from the CL file, only three showed near-infrared absorption properties; namely, the manganous complex of beta-nitroso-alpha-naphthol (λ_{max} = 825 mμ, a_{max} = 2.63), 3-(p-nitrophenylazo)-1,2-dimethylimidazo$\overline{[}$1,2-a$\overline{]}$ pyridinium iodide (λ_{max} = 1030 mμ, $a_{max} \simeq$ 13; absorbs only in DMF and pyridine, the absorbing species being destroyed within one-half to one hour), and dihydroxygermanium phthalocyanine, PcGe(OH)$_2$, (λ_{max} = 925 mμ). The remaining compounds showing no near-infrared absorption are listed in Appendix III by empirical formula, according to the system used in the CL files, together with their chemical nomenclature.

B. Evaluation

Quantitative spectral curves were determined for compounds showing near-infrared absorption bands. These included for the most part those compounds prepared in the synthesis program of this investigation. These curves also were run from 400 mμ to 1700 mμ on the DK-2 spectrophotometer with solvent as reference using matched 1.00-cm thick silica cells. The absorptivity, a, at each wavelength of maximum absorption for a given absorber was determined using the relationship,

$$A = abc \tag{1}$$

where A = absorbance at a λ_{max}; b, the cell path in centimeters; and c, concentration in grams/liter. The symbols and spectral definitions used are those adopted by the American Chemical Society, Analytical Chemistry Advisory Board $\overline{[}$Anal. Chem. 33, 1968 (1961)$\overline{]}$.

C. Application

Attempts were made to incorporate the near-infrared-absorbing compounds into plastics and as coatings on glass or plastic by one or more of the following six methods:

(1) solution-cast coatings
(2) hot milling and molding
(3) monomer casting
(4) burnishing
(5) dyeing
(6) vacuum evaporation

Most of these studies were made with various forms of poly(methyl

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methacrylate).* The specific plastics and monomers used in this investigation were as follows:

> Plexiglas, poly(methyl methacrylate), VM-100 crystal - Rohm and Haas
> Lucite 30, poly(methyl methacrylate), bead polymer - du Pont
> Acryloid B-72, 40% PMMA in toluene - Rohm and Haas
> Methyl methacrylate monomer, practical - Matheson, Coleman and Bell
> Cellulose acetate powder - Eastman
> "CR-39," allyl diglycol carbonate - Columbia Southern
> Laminac 4123, polyester in styrene - American Cyanamid
> Polystyrene - Dow Chemical

Solution-cast coatings on glass microscope slides were prepared by dipping a slide into a 50-cc portion of a stock solution of plastic in which portion was dissolved enough absorber to produce a desired amount of near-infrared absorbance on the dry coating at the λ_{max}, as measured with a Spectronic 20 colorimeter. The slide was allowed to dry slowly at about 55°C. for 15 to 30 minutes. Second and third dippings of the same slide were often required to increase the absorbance to a proper level for light stability measurements, this level being between about 5% transmittance (A = 1.30) and 30% transmittance (A = 0.52). The films or coatings produced on both sides of the glass slide were each in the order of 3 to 10 mils in thickness. The stock solutions of plastic used were usually 20% cellulose acetate or 22% Plexiglas in A.C.S. acetone, but some experiments were also performed with 30% Lucite in A.C.S. toluene and 38% polystyrene in tetrahydrofuran (b.p. = 65.5-66.5°C.).

By means of hot roll milling followed by hot compression molding, thick Plexiglas plates of about 60 to 100 mils thickness and thin films of about 10 to 20 mils were prepared. The absorber (0.02 to 0.2 gm) was added to 100 gm of semi-molten plastic on a two-roll laboratory mill heated to about 170°C. (front roll at about 180°C., back roll at about 160°C.). Mixing was effected by continuously stripping off with a doctor blade and passing the material between the rolls for about 20 to 40 times, depending on the ability of the absorber to withstand these temperatures. A portion of the resulting thick mass was then hot compression molded into a smooth transparent plate or film. The molding conditions were as follows: temperature, 170°C.; sample preheated for 5 minutes with no pressure; 20 tons force applied on a 4- by 5-inch sample, for about 3 minutes while hot and for about 4 minutes while cooling.

Methyl methacrylate, allyl diglycol carbonate (CR-39), and Laminac 4123 polyester resin were polymerized in molds into 1/8-inch clear plates. This was done by pouring the catalyzed monomer or monomer-polymer mixture into a mold consisting of two smooth 1/4-inch thick Pyrex glass plates separated with a 1/8-inch vinyl gasket placed along three edges, the mold being held together with fairly heavy-duty binder clips. The catalysts used were either 3 to 5% benzoyl peroxide or 3% 2,2'-azobis(2-methylpropionitrile), "Porophor N," for CR-39; 0.5% benzoyl peroxide or Porophor N, for Laminac;

*Recommended by the Air Force (private communication) because of the suitable optical and physical properties of high-impact poly(methyl methacrylate).

and 0.01 to 0.2% Porophor N, for methyl methacrylate. The curing conditions were 100°C. for one hour for Laminac and 55° ± 5°C. for about 20 hours for CR-39 and methyl methacrylate. None of the major classes of near-infrared absorbers could withstand the systems described for Laminac and CR-39. The bulk of the monomer-cast work was confined, therefore, to methyl methacrylate.

The practical grade of monomeric methyl methacrylate required a washing treatment to remove added inhibitors. This process[5] involved two ten-minute washings with 20 parts by weight of 5% sodium hydroxide - 20% sodium chloride in water per 100 parts of monomer, a third wash with about 20 parts of distilled water followed by suction filtration through at least three layers of fine filter paper.

The formulation, containing the absorber, adopted for monomer casting the methacrylate was a solution of the following constituents:

Monomeric methyl methacrylate	80 gm
Poly(methyl methacrylate) (Lucite)	20 gm
Porophor N	0.01 to 0.05 gm
Near-infrared absorber	0.01 to 0.1 gm

The burnishing technique[22] was used to form very thin reflecting coatings of the pure solid infrared absorber on glass or plastic surfaces. This was done by merely hand rubbing the finely divided (-325 mesh) solid absorber onto the surface with soft 80-square cotton cloth. In general, the samples obtained were hazy and nonuniform. Attempts to overcome these faults by mechanical buffing, that is with a buff wheel attachment on a 1/4-inch drill, failed. Overcoating improved the transparency somewhat. An advantage to burnishing is the marked increase in light stability attained over other methods of incorporation due to the particulate or solid state of the absorber. In cases where the overcoat partially dissolved the absorber, for example, an alkyd resin in xylene on the tris(p-dimethylaminophenyl)aminium perchlorate (XXVII), the light stability was sharply reduced (Table XIII). Of the overcoatings tried, only mineral oil could be used for this absorber. The best sample obtained by means of burnishing with respect to transparency with fair near-infrared attenuation was one with the tetracyanonickelate salt of the 3,6-bis(dimethylamino)-9-phenylfluoren-9-ol (XVI) burnished on cellulose acetate and overcoated with Acryloid B-72, the spectrophotometric curve being shown in Fig. 5.

Several dyeing techniques were attempted which are described below (Tables VII and XIV).

Pieces of monomer-cast poly(methyl methacrylate), with Shore D hardness of 86, and polymerized allyl diglycol carbonate (CR-39), Shore D of 45, were dyed with the zinc chloride salt of 3,6-bis(dimethylamino)-9-phenylfluoren-9-ol (XIV) and with tris(p-dimethylaminophenyl)aminium perchlorate (XXVII). The dye baths consisted of about 50% 3A alcohol with about 2 to 3 ml of Cyanatex Dyeing Assistant EM (methyl salicylate emulsion) plus a small amount (0.1 to 0.5 gm per 50 ml. dye bath) of the absorber. The dyeings were effected at steam bath temperatures within one to two hours. After the dyeings the Shore D hardness of the poly(methyl methacrylate) had decreased to 75.

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Harder samples of CR-39 (Shore D = 83) could not be dyed with the above two absorbers. Even after pretreatment overnight with various chlorinated hydrocarbons (carbon tetrachloride, chlorobenzene, and 1,2-dichloroethane) and 3A alcohol the dyeings were unsuccessful. These hard CR-39 samples, however, could be very easily dyed with an acetate dye, Calcosyn Green 2B and with a disperse dye, Calcosperse Blue 3R, using a dye bath with about 1 ml of Cyanatex Dyeing Assistant EM per 100 cc water or 50% 3A alcohol.

Other techniques used which may be classified as dyeings were investigated. One scheme was to incorporate about 2% p-toluenesulfonic acid in Laminac 4123 via monomer casting. A chip of this was immersed into an acetone solution of 3,6-bis(dimethylamino)-9-phenylfluorene-9-ol (V). Immediately the solution turned a deep yellow indicating that the sulfonic acid had leached out into the solution, which effect was opposite to that hoped for. Another experiment was somewhat successful using perfluorooctanoic acid incorporated into monomer-cast CR-39. This plastic sample when immersed into a solution of the fluorenol in warm hexane assumed the yellow coloration of the infrared-absorbing color salt of the fluorenol, with a transmittance of 33% at 960 mμ and 83% at 600 mμ. A third attempt was to incorporate silver trifluoroacetate into a plate of poly-(methyl methacrylate) by monomer casting followed by treatment with a solution of tris(p-dimethylaminophenyl)amine in petroleum ether in hopes of forming the infrared-absorbing aminium salt on the surface of the plastic. However, the solution turned green with black elemental silver coating the plastic surface. A similar experiment was performed but in reverse; that is, the amine was incorporated into Plexiglas (0.1 gm/100 gm) by milling and molding which was later treated with a 0.2 N AgClO$_4$ in acetone solution. In this case, there was no evidence of a chemical combination either on the plastic or in the solution.

An advantage to a dyeing procedure is that it would be a means of incorporation in or on monomer-cast plastic sheets or plates those absorbers which normally could not chemically withstand the curing catalysts and temperatures used in monomer casting. In addition surface dyeings would give uniform attenuation to samples of variable thickness.

Vacuum evaporation or sublimation is a common method for purification and for the production of thin smooth layers of certain solid compounds. For example, solid cationic dyes can be vacuum evaporated as thin specular coatings on glass surfaces[30]. Attempts were made, therefore, to vacuum evaporate three absorbers representing three major classes onto glass microscope slides. These compounds were the fluoborate salt of 3,6-bis(dimethylamino)-9-phenylfluoren-9-ol (XVII); 1,1,5,5-tetrakis(p-dimethylaminophenyl)divinylcarbonium perchlorate (XXXII); and tris(p-dimethylaminophenyl)aminiumfluoborate (XXVIII). The procedure involved first the preparation of small pressed pellets of the compound. Then a portion of this pellet was placed in a tungsten boat clamped between two electrodes. A glass microscope slide was clamped upright with its long edge horizontal on a bracket about one-half inch from and slightly higher than the top edge of the boat. This assembly was contained in an inverted bell jar connected to a fore pump in series with a diffusion pump. The pressure obtained was about 10^{-3} to 10^{-2} mm of Hg. The current through the boat was increased slowly to a point such that its temperature was just below the melting point of the contents, as judged visually; that is, the current was slightly decreased when the first signs of melting were noticed.

In all three cases no evaporation or coating on the glass slide occurred after 2 to 3 hours at this setting. The current had to be increased to a point where the contents of the boat melted before any evaporation took place. All three compounds, however, became dark or black. The divinyl-carbonium compound (XXXII) and the trisphenylaminium salt (XXVIII) gave white diffuse coatings, and the fluorenol salt (XVII) gave an orange diffuse coating. None showed near-infrared-absorbing or reflecting properties.

One attempt was made to attenuate near-infrared radiation by means of scattering or diffuse reflection and yet transmit visible radiation as in a Christiansen filter[21]. The Christiansen filter effect was demonstrated by incorporating finely powdered KCl (-325 mesh) at a concentration of about 33% into monomer-cast poly(methyl methacrylate). Because of the equivalence of refractive indices of the KCl and plastic for yellow light (n_D = approximately 1.49), the resulting 1/8-inch plastic plate transmitted yellow from any strong white light source and the scattered blue and violet radiations could be seen about its edges. A spectrophotometric curve, Fig. 35, however, showed only slight attenuation in the near infrared as compared to the peak visible transmittance at about 620 mμ, the greater part of the scattering being in the blue and violet region.

Table XXVIII is presented as a summary of the applicability of the various classes of infrared absorbers to the six different methods of formulation described above.

The spectrophotometric curves of all the plastic samples containing infrared absorbers were determined with a Beckman KD-2 recording spectrophotometer with air as a reference.

Light stability measurements were performed mostly on solution-cast coatings on glass slides; however, several milled and molded, and burnished samples were also included. Usually a standard Fade-Ometer was used to expose the samples, but in some of the later experiments an Atlas Xenon Weather-Ometer without water spray was also used. The data were reported as % remaining activity defined as:

$$\% \text{ Remaining} = \frac{c_t}{c_o} \times 100 \qquad (2)$$

where c_o denotes the original concentration and c_t, the concentration after exposure. By application of equation (1), equation (2) then reduces to

$$\% \text{ Remaining} = \frac{A_t}{A_o} \times 100$$

where A_o is the original absorbance at a λ_{max} and A_t, the absorbance at the same λ_{max} after exposure. In determining A_t and A_o allowance was made for scattering and/or reflection. No statistical analysis for the precision of the test was made, but it is felt the reproducibility to be of the order of 10%.

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D. Compilation of Data

        The spectrophotometric curves of the infrared absorbers in
solution, in plastics, and on plastic or glass surfaces are compiled in
Appendix II. All the curves with one exception, are reproduced as %
transmittance vs. wavelength in millimicrons. The % transmittance
ordinate was chosen for practical reasons since the data obtained
directly from the DK-2 is in % transmittance vs. wavelength. The one
exception, that of the 3,6-bis(dimethylamino)-9- phenylfluoren-9-ol in
glacial acetic (Fig. 1), was plotted as log ε vs. wavelength, since all
the other salts investigated showed the same spectral properties, i.e.,
the same λ_{max} and theoretically the same ε_{max} values in the visible and
near infrared.

        Light stability data and other spectral properties are presented
in Appendix I.

VI. CONCLUSIONS

        Considerable progress was made towards developing plastic or
coating compositions having strong near-infrared attenuation and high
visible transmittance. This was accomplished by incorporating into a
resin small amounts of organic compounds having strong near-infrared
absorption. These compounds represent most of the classes of compounds
found to possess strong absorption in the near infrared. None of them,
however, absorb significantly above 1100 mμ.

        Other properties of these substances, such as light stability and
compatibility, can be improved by introducing longer alkyl groups and, in
the case of salts, by using certain anions such as fluoborate or perfluoro-
alkanoates. Since photodegradation is due mainly to ultraviolet wave-
lengths, the light stability can also be increased by using protective
ultraviolet absorbers.

        The composition with the best spectral properties developed
during this work is poly(methyl methacrylate) containing tris(p-diethyl-
aminophenyl) aminium fluoborate (Fig. 12).

        Success can probably be achieved in protecting the human eye
against high-energy thermal radiation by using infrared-attenuating coatings
or plastics with retention of visible transmission.

        Complete attainment of the spectral properties required presents
some challenging problems. One involves the 85% transmission requirement in
the visible region. It is almost inevitable that a visibly-transparent,
infrared-attenuating medium will absorb somewhat near the 400 and 700 mμ
regions since absorption curves never have perpendicular slopes. However,
low transmittances near the edges may be tolerable because the eye is less
sensitive in these regions. Realistically, the 85% value is exceptionally
high when high attenuation at 750 mμ is required, since perfectly clear
and smooth coatings will transmit only 90-94% of visible light. The 6 to
10% loss is due to normal reflectance. This permits only 9 to 5% attenua-
tion in the visible region by the infrared-absorbing additives.

Another problem is finding compounds that can be made into a filter that attenuates completely over the 750 to 2000 mμ range particularly between 1200 and 2000 mμ. Further work towards uncovering compounds that absorb in this area is required.

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APPENDIX I

Tables of Data

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TABLE I

3,6-BIS(DIMETHYLAMINO)-9-PHENYLFLUOREN-9-OL SALTS
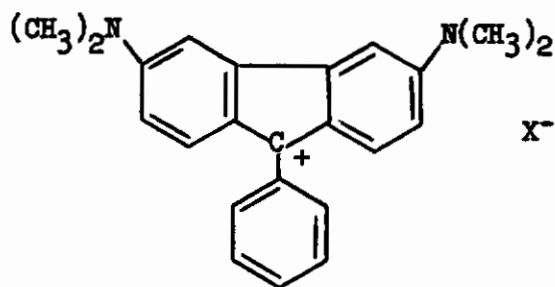
	X	a_{max} (Calc'd)[b] 960 mμ	a_{max} (Found)[a]			
		960 mμ	960 mμ	850 mμ	505 mμ	480 mμ
XII	Acetate	47.2	47.2	38.3	55.0	53.5
XIII	Oxalate	39.4	43.7	35.8	50.2	49.5
XIV	Tetrachlorozincate	42.4	24.9	20.7	29.8	29.7
XV	Phosphomolybdate[c]	19.5	12.6	10.9	-	-
XVI	Tetracyanonickelate	44.7	33.9	26.9	40.4	39.5
XVII	Fluoborate	44.1	47.0	38.2	52.1	51.0
XVIII	Trichloroacetate	37.3	33.9	28.2	40.0	39.8
XIX	Perfluorobutyrate	34.4	26.3	21.6	30.1	29.2
XX	Ethanesulfonate	41.8	42.7	34.9	49.3	47.9
XXI	Diphenyl-4,4'-disulfonate	28.1	25.0	20.5	28.4	27.6
XXII	Trifluoroacetate	41.4	37.2	30.8	42.8	41.2
XXIII	Perchlorate	42.7	48.2	39.8	52.3	50.9

(a) Curves run in acetic acid.

(b) Calc'd. from the molar absorptivity (18,230) of the pure fluorenol in acetic acid.

(c) Curve run in 10% aqueous DMF - anion assumed to be $(P_2O_8 \cdot 24\ MoO_3)^{-6}$.

<u>TABLE II</u>

3-DIMETHYLAMINO-9-(p-DIMETHYLAMINOPHENYL)FLUOREN-9-OL SALT
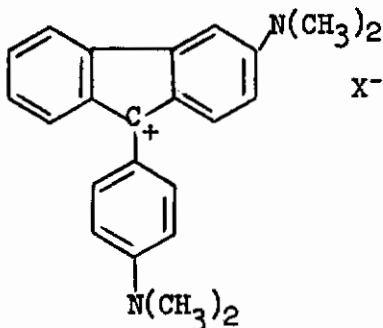
	X	780 mμ	720 mμ	565 mμ	470 mμ	440 mμ
				a_{max}(a)		
XXIV	Acetate	140	128	42.0	36.0	24.2

(a) Curve run in acetic acid.

<u>TABLE III</u>

LIGHT STABILITY OF SOLUTION-CAST COATINGS OF CELLULOSE ACETATE CONTAINING
SALTS OF 3,6-BIS(DIMETHYLAMINO)-9-PHENYLFLUOREN-9-OL

	Salt	10 Hrs.	20 Hrs.	30 Hrs.	40 Hrs.	60 Hrs.
		% Remaining after Fade-Ometer Exposures:				
XIV-	Tetrachlorozincate	22	13	-	-	-
XIX	Perfluorobutyrate	15.8	11.6	8.7	-	-
XXII	Trifluoroacetate	14.5	10.5	9	-	-
XXIII	Perchlorate	73.5	69	57.5	55	44.5
XVII	Fluoborate	48.5	36.8	27.5	22.4	16.5
XXI	Diphenyl-4,4'- disulfonate	- cloudy, insoluble				
XVIII	Trichloroacetate	- hydrolyzed to colorless species				

TABLE IV

SAMPLES BURNISHED WITH COLOR SALTS OF 3,6-BIS(DIMETHYLAMINO)-9-PHENYLFLUOREN-9-OL

	Salt	Surface Burnished*	% Remaining After Fade-Ometer Exposures		
			100 Hrs.	145 Hrs.	250 Hrs.
XIV	Tetrachlorozincate	Cell. Acetate	-	-	65
"	"	Plexiglas	-	44	-
"	"	Glass Slide (one side)	-	44	-
"	"	Glass Slide (two sides)	-	66	-
XVI	Tetracyanonickelate	Cell. Acetate	48.5	-	-

*Overcoated with Acryloid B72.

TABLE V

MONOMER-CAST POLY(METHYL METHACRYLATE) CONTAINING COLOR SALTS OF 3,6-BIS-(DIMETHYLAMINO)-9-PHENYLFLUOREN-9-OL

	Salt	Conc.	Maximum NIR Absorption		Peak Visible Transmittance	
			% T	m μ	% T	m μ
XIX	Perfluorobutyrate	0.2%	1.3 2	960 850	83	625*
"	"	(>0.2%)	0	800-975	57.5	625*
XXII	Trifluoroacetate	0.2%	25 25.6	960 850	86	650*
"	"	(>0.2%)	12.5 16.8	960 850	77.2	650*
XVII	Fluoborate	0.04%	2.5 4	960 850	62	650**

*Dark orange, very clear
**Dark orange, hazy

TABLE VI

MILLED AND MOLDED POLY(METHYL METHACRYLATE) CONTAINING COLOR SALTS OF
3,6-BIS(DIMETHYLAMINO)-9-PHENYLFLUOREN-9-OL

	Anion	Conc.	Maximum NIR Absorption		Peak Visible Transmittance	
			% T	m μ	% T	m μ
XVII	Fluoborate	0.1%	1.8 2	960 850	21	650*
XVIII	Trichloracetate	0.1%	None		53	700**
XIX	Perfluorobutyrate	0.1%	29 31.6	960 850	56	650***
XX	Ethanesulfonate	0.1%	7 8.5	960 850	47	650***
XXI	Diphenyl-4,4'- disulfonate	0.1%	13.5 15	960 850	55	650***
XXII	Trifluoroacetate	0.1%	51 51	960 850	59	675***

*Haze
**Slight haze
***Very slight haze

SAMPLES OF PLASTIC DYED WITH COLOR SALTS OF 3,6-BIS(DIMETHYLAMINO)-
9-PHENYLFLUOREN-9-OL

Method of Application	Plastic	Shore D Hardness	% Transmittance at 960 mμ	at 650 mμ
a	CR-39	45	10	-
a	PMMA	75	30	80
b	CR-39	55	33 (21 at 850 mμ)	83
c	Plexiglas	85	26	49.5
d	Plexiglas	85	(very weak peak at 900 mμ)	
e	Plexiglas	85	(no absorption peaks)	

a. Monomer cast poly(methyl methacrylate) dyed with tetrachlorozincate salt (XIV) from 50% alcohol containing 2 to 3% Cyanatex Dyeing Assistant.

b. Monomer cast CR-39 containing perfluorooctanoic acid treated with solution of the fluorenol base in hexane.

c. p-Toluene sulfonic acid (0.2%) milled in Plexiglas followed by addition of the free fluorenol (0.1%) and combination milled and molded.

d. Same process as c, except colloidal Dowex 50 (0.3%) and the free fluorenol (0.15%) used.

e. Same process as c, except phosphomolybdic acid (0.4%) and the free fluorenol (0.15%) used.

TABLE VIII

SOLUTION-CAST CELLULOSE ACETATE COATINGS CONTAINING THE TETRACHLOROZINCATE
SALT OF 3,6-BIS(DIMETHYLAMINO)-9-PHENYLFLUOREN-9-OL (XIV)

	% Remaining Protective Screen	
Fade-Ometer Hours	12-Mil C.A.*	UV Abs. 12-Mil C.A.**
3	23	73.5
13	7.7	38

* 12-mil cellulose acetate
** 12-mil cellulose acetate absorbing 95% of near UV

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TABLE IX

SOLUTION CAST POLY(METHYL METHACRYLATE) COATINGS CONTAINING COLOR SALTS OF SUBSTITUTED 9-PHENYLFLUOREN-9-OLS

	Substitution	Salt	% Remaining After 15.6 Hours in Xenon Weather-Ometer
XIV	3,6-Bis(dimethylamino)-	$ZnCl_4^=$	87
XVII	3,6-Bis(dimethylamino)-	BF_4^-	76
XVI	3,6-Bis(dimethylamino)-	$Ni(CN)_4^=$	*
XXIV	3-Dimethylamino-9-(p-dimethylamino)	CH_3COO^- (HAc added in situ)	0

* Sample could not be prepared due to slow hydrolysis to colorless free base.

TABLE X

TRIARYLAMINIUM FREE RADICAL SALTS

$$\left(R_2N-\!\!\left\langle \bigcirc \right\rangle \right)_3 \overset{+}{\underset{\cdot}{N}} \quad X^-$$
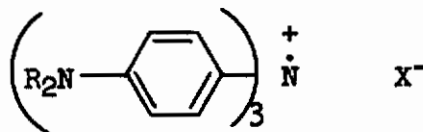
	R	X	$\lambda_{max}\ (m\mu)^{(a)}$	a_{max}	ε_{max}
XXVII	CH_3	ClO_4	960	80.1	38,000
XXVIII	CH_3	BF_4	960	119	54,800
XXVIII	CH_3	$BF_4^{(b)}$	960	82.4	37,900
XXIX	C_2H_5	ClO_4	960	56.9	31,800
XXX	C_2H_5	BF_4	960	77	42,000
XLVII	$C_4H_9^{(c)}$	BF_4	980	44	31,400

(a) Curves run in methanol
(b) Curve rerun after one month
(c) This derivative also had a peak in the visible; $a_{max} = 21$ at 400 mμ

TABLE XI

SOLUTION-CAST COATINGS OF CELLULOSE ACETATE CONTAINING TRIS(p-DIALKYL-AMINOPHENYL)AMINIUM SALTS AND THEIR RELATIVE LIGHT STABILITY WITH AND WITHOUT UV LIGHT PROTECTION

A. Tris(p-dimethylaminophenyl)aminium Perchlorate (XXVII)

Transmittance at 960 mμ(%)	Transmittance at 550 mμ(%)	Protective Screen	% Remaining After 13 Hours in Fade-Ometer
9	92.5	None	15 (12 hours)
20	85	12-mil cellulose acetate	39
22	85.5	12-mil cellulose acetate absorbing 95% of near UV	52

B. Relative Light Stability to Fade-Ometer Exposures

	Alkyl Group	Anion	% Transmittance at 960 mμ	% Transmittance at 550 mμ	After 15 Hrs.	% Remaining Behind Yellow Filter 20 Hrs.	% Remaining Behind Yellow Filter 120 Hrs.
XXVII	Methyl	ClO_4^-	15	88	40	-	-
XXVIII	Methyl	BF_4^-	27	88.5	43.5	79	50
XXX	Ethyl	BF_4^-	18	90	60	92	72

C. Relative Light Stability to Xenon Weather-Ometer

	Alkyl Group	Anion	% Transmittance at 960 mμ	% Transmittance at 550 mμ	% Remaining 27.3 Hrs.	% Remaining 54.3 Hrs.	% Remaining 82 Hrs.
XXVII	Methyl	ClO_4^-	18	84	45	25.2	16
XXVIII	Methyl	BF_4^-	15.3	89	52	31.4	22
XXX	Ethyl	BF_4^-	6.9	92.2	54.5	37	27
XXIX	Ethyl	ClO_4^-	3.0	89.8	72	59	52

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TABLE XII

MILLED AND MOLDED PLASTIC CHIPS CONTAINING TRIS(p-DIALKYLAMINOPHENYL)- AMINIUM SALTS

	Alkyl Group	Anion	Plastic	Conc. Wt. - %	Thickness (mils)	% Transmittance 960 mμ	% Transmittance 550 mμ
XXVII	Methyl	ClO_4^-	Cel. Acet.	0.02	109*	4	43
XXVII	Methyl	ClO_4^-	Plexiglas	0.05	65	5	52
XXVII	Methyl	ClO_4^-	Plexiglas	0.20	65	6	56
XXVIII	Methyl	BF_4^-	Plexiglas	0.05	67**	8	71
XXX	Ethyl	BF_4^-	Plexiglas	0.05	67**	0	80

* Light stability: 49% remaining after 20 hours in Fade-Ometer.
** Light stability on 10-mil chips: XXVIII = 55%, XXX = 51% remaining after 50 hours Fade-Ometer.

TABLE XIII

TRIS(p-DIALKYLAMINOPHENYL)AMINIUM SALTS BURNISHED ON PLASTIC AND GLASS SURFACES

	Alkyl Group	Anion	Surface Burnished	Overcoat	Transmittance Visible % T	Transmittance Visible mμ	Transmittance Near Infrared % T	Transmittance Near Infrared mμ
XXVIII	Methyl	BF_4^-	Poly(methyl-methacrylate)	None	29	575	0.5	900-1050
XXVII	Methyl	ClO_4^-	Plexiglas*	Alkyd Resin	72	575	42	970
XXVII	Methyl	ClO_4^-	Glass Slide**	***	72	650	36	980

* Light Stability: 40% remaining after 20 hours in Fade-Ometer.
** Light Stability: 60% remaining after 50 hours in Fade-Ometer.
*** Mineral Oil + second glass slide.

TABLE XIV

SAMPLES DYED WITH TRIS(p-DIMETHYLAMINOPHENYL)AMINIUM PERCHLORATE (XXVII)

Plastic Dyed	% Transmittance at 960 mμ
Poly(methyl methacrylate)	5
CR-39 (Shore D = 45)	0
CR-39 (Shore D = 45)	10

TABLE XV

TETRAPHENYLVINYLCARBONIUM SALTS
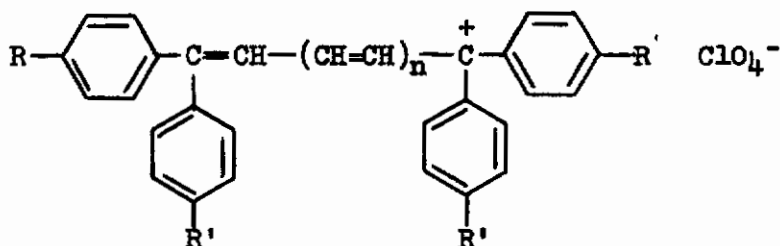
	R	R'	n	λ max (mμ)[a]	α max
XXXI	$(CH_3)_2N$	$(CH_3)_2N$	0	730 635 570	62.3 102 84.8
XXXII	$(CH_3)_2N$	$(CH_3)_2N$	1	800 630	159 56.8
XXXIII	$(CH_3)_2N$	$(CH_3)_2N$	2	900 675	195 69.4
XXXIV	$(CH_3)_2N$	H	1	825 550 400	232 26.8 26.8
XXXV	$(CH_3)_2N$	H	2	900 575 440	154 20.4 19.4
XXXVI	CH_3O	CH_3O	2	750 495	177 26.4

(a) Curves determined in acetone.

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TABLE XVI

XANTHYLOMETHINES

	R	X	n	λ_{max} (mμ)[a]	a_{max}
XXXVII	H	O	1	700	56.9
XXXVIII	H	O	2	780	74.5
XXXIX	H	O	3	855	124
XL	CH_3O	O	3	900	52.8
				800	54.1
				443	37.4

(a) Curves run in acetone.

TABLE XVII

TETRAPHENYLVINYLCARBONIUM PERCHLORATES IN SOLUTION-CAST COATINGS

$$(CH_3)_2N-\bigcirc-C=CH-(CH=CH)_n-\overset{\oplus}{C}-\bigcirc-N(CH_3)_2 \quad ClO_4^{\ominus}$$
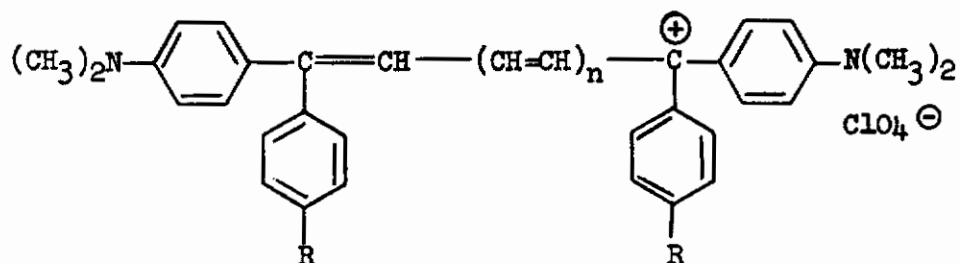
	n	R	λmax	Lucite % Remaining After Xenon Hours 15.6	Cellulose Acetate % Remaining After Fade-Ometer Hours 20	40
XXXI	0	$-N(CH_3)_2$	730	80	72.5	56.5
XXXII	1	"	800	25	34	21
XXXIII	2	"	900	2.9	19	10
XXXIV	1	H	825	27.6	(65)*	-
XXXV	2	H	900	15	(66)*	-

* Interpolated values.

TABLE XVIII

SOLUTION-CAST CELLULOSE ACETATE CONTAINING MISCELLANEOUS POLYMETHINES

	Compound	λmax	% Remaining After Fade-Ometer Hours 5	15	25
XXXVIII	Bis(xanthene)pentamethine ClO_4^-	780	49	14.5	7.9
XXXIV	Bis(xanthene)heptamethine ClO_4^-	855	9.7	-	-
XLI	Azulene Derivative*	860	34.4	12.3	7

* 5-Isopropyl-1-/5-(5-isopropyl-3,8-dimethylazulen-1-yl)penta-2,4-
dienylidene7-3,8-dimethylazulenium perchlorate.

TABLE XIX

MILLED AND MOLDED POLY(METHYL METHACRYLATE) CONTAINING POLYMETHINE TYPE ABSORBERS

	Compound	Conc. (Wt. - %)	Maximum Near-Infrared Absorption		Peak Visible Transmittance	
			% T	$\lambda(m\mu)$	% T	$\lambda(m\mu)$
XXXI	1,1,3,3-Tetrakis(p-dimethylaminophenyl)-vinylcarbonium perchlorate	0.05	5	730	50 blue (sl. haze)	430
XXXII	1,1,5,5-Tetrakis(p-dimethylaminophenyl)-divinylcarbonium perchlorate	0.05	0	800	29 blue (sl. haze)	470
XXXIII	1,1,7,7-Tetrakis(p-dimethylaminophenyl)-trivinylcarbonium perchlorate	0.05	2	900	24 blue (hazy)	465
XXXIV	1,5-Bis(p-dimethyl-aminophenyl-1,5-di-phenyldivinylcarbonium perchlorate	0.05	0	660-850	14 10 purple (haze)	450 575
XXXV	1,7-Bis(p-dimethylamino-phenyl)-1,7-diphenyl-trivinylcarbonium perchlorate	0.05	7	925	30 gray-blue (hazy)	625
XXXVII	Bis(xanthene)trimethine perchlorate	0.05	22.8	705	34 green (hazy)	530
XXXVIII	Bis(xanthene)penta-methine perchlorate	0.05	no peak		34.7 Olive (hazy)	565
XXXIX	Bis(xanthene)hepta-methine perchlorate	0.05	9	800-1000	12 (very hazy)	660
XLI	Azulene derivative	0.05	11	875	22.8 purple (hazy)	630

TABLE XX

POLYMETHINES IN MONOMER-CAST POLY(METHYL METHACRYLATE)

	Compound	Conc. (Wt. - %)	Near-Infrared Absorption % T	Near-Infrared Absorption $\lambda(m\mu)$	Peak Visible Transmittance % T	Peak Visible Transmittance $\lambda(m\mu)$
XXXI	1,1,3,3-Tetrakis(p-dimethylaminophenyl)-vinylcarbonium perchlorate	0.05	4	730	47 blue (clear)	440
XXXII	1,1,5,5-Tetrakis(p-dimethylaminophenyl)-divinylcarbonium perchlorate	0.05	0	800	21 blue (clear)	520
XXXIII	1,1,7,7-Tetrakis(p-dimethylaminophenyl)-trivinylcarbonium perchlorate	0.05	4	900	72 (insoluble)	520
XXXIV	1,5-Bis(p-dimethyl-aminophenyl)-1,5-diphenyldivinylcarbonium perchlorate	0.05	0	640-850	10 9 purple (clear)	575 455
XXXV	1,7-Bis(p-dimethyl-aminophenyl)-1,7-diphenyltrivinyl-carbonium perchlorate	0.05	1.5	925	37 yellow-green (insol. matter)	550
XXXVII	Bis(xanthene)trimethine perchlorate	0.05	2.5	720	64 pale green	525
XXXVIII	Bis(xanthene)pentamethine perchlorate	0.05	none		greenish-yellow	
XXIX	Bis(xanthene)hepta-methine perchlorate	0.05	(insoluble)			
XLI	Azulene derivative	0.05	4	860	51 dull green (insol. matter)	620

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TABLE XXI

SOLUTION-CAST PLASTIC COATINGS ON GLASS SLIDES CONTAINING THE NICKEL COMPLEX OF THE FORMAZAN (XLIII)

Plastic	% Transmittance at		Protective Screen	Fade-Ometer Hours	% Remaining
	825 mμ	640 mμ			
Cell. Acet.	25	74	none	434	90
Cell. Acet.	20.6	69.8	12-mil Cell. Ac. Film	484	93
Cell. Acet.	20	71	UV Abs. 12-mil Cell. Ac. Film	484	97
Plexiglas	20	57.5	none	25	100
Polystyrene	18.4	60.8	none	25	100

TABLE XXII

MONOMER-CAST POLY(METHYL METHACRYLATE) CONTAINING 0.05% OF THE NICKEL COMPLEX OF THE FORMAZAN (XLIII)

Wavelength (Millimicrons)	% Transmittance
200-480	0
640	31.8
790-825	0

TABLE XXIII

MILLED AND MOLDED POLY(METHYL METHACRYLATE) CONTAINING 0.2% OF THE NICKEL COMPLEX OF THE FORMAZAN (XLIII)

Wavelength (Millimicrons)	% Transmittance	
	17 mil Film	62 mil Chip
400	12	0
500	15	0
650	33.5	5.5
700 ($\lambda_{min.}$)	37	7
825 ($\lambda_{max.}$)	24	1.5

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TABLE XXIV

SAMPLE OF CELLULOSE ACETATE BURNISHED WITH THE NICKEL COMPLES OF THE
FORMAZAN (XLIII)

Wavelength (Millimicrons)	% Transmittance
400-475	2
650 ($\lambda_{min.}$)	26
840 (λ_{max})	3.7

TABLE XXV

LIGHT STABILITY OF PIGMENTS IN POLY(METHYL METHACRYLATE)

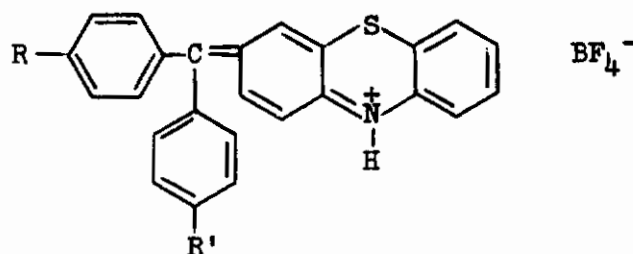| Compound	Conc. (%)	% Remaining - After Hours in Fade-Ometer					
		95	200	245	445	500	700
THN Green	0.1	84	-	86	86	-	-
Pigment Green B	0.1	98	-	73	54.5	-	-
Cupric Oleate	5.0	(>81)	88	-	(86)	-	-
PcGe(OH)$_2$ (Fig. 30)	0.1	(>92)	91.5	-	-	(88)	89

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TABLE XXVI

PIGMENTS MILLED AND MOLDED IN POLY(METHYL METHACRYLATE)

Compound	Conc. %	Near-Infrared Absorption	Visible Peak Transmittance Sample Appearance
Cupric oleate	3.0	no max.	21% T at 535 mμ green (clear)
Cupric stearate	1.0	no max.	30% T at 535 mμ lt. green (clear)
Copper phthalocyanine green (THN Green)	0.1	no max.	28% T at 545 mμ blue-green (clear)
Ferric complex of 1-nitroso-2-naphthol	0.1	none	18% T at 700 mμ olive (clear)
Pigment Green B (Fig. 36)	0.1	cut off between 600 and 850 mμ	22% T at 550 mμ green (clear)
Vanadyl Phthalo-cyanine (Fig. 31)	0.1	0% T between 600 and 1000 mμ	10% at 525 mμ blue-green (v. hazy)
Cobaltous Phthalocyanine	0.1	no max.	33% T at 515 mμ blue (v. hazy)
(Chloroiron) Phthalocyanine	0.1	0% T between 600 and 905 mμ	4% T at 565 mμ green (v. hazy)
Dihydroxygermanium Phthalocyanine (Fig. 29)	0.1	0% T between 560 and 925 mμ	29% T at 485 mμ green-blue (sl. haze)
Diphenoxygermanium Phthalocyanine (Fig. 29)	0.1	0% T between 615 and 700 mμ	30% T at 535 mμ green (sl. haze)
Copper Tetrabromo phthalocyanine (α)	0.2	<22% T from 700 to 1000 mμ	28% T at 535 mμ blue-gray (very hazy, large agglomerates)
Copper Tetrabromo phthalocyanine (β)	0.1	<9% T from 700 to 1000 mμ	30% T at 525 mμ blue (very hazy)
CuSO$_4$ (325 mesh)	0.5	none	very hazy
FeCl$_2$·3H$_2$O (oxidation to ferric evident)	0.5	none	very hazy
Auralux M (Ferric Oxide Pigment)	0.4	none	very hazy
Auralux L (Ferric Oxide Pigment)	0.3	none	very hazy

TABLE XXVII

PHENOTHIAZINE DERIVATIVES

R—⟨benzene⟩—C=⟨phenothiazine ring system, S at top, N⁺H at bottom⟩ BF_4^-

with R' on lower phenyl group

	R	R'	$\lambda_{max}\ (m\mu)$ [a]	a_{max}
XLIV	H	H	680	17.2
			442	13.4
XLV	$(CH_3)_2N$	H	715	48.0
			485	28.4
XLVI	CH_3O	CH_3O	720	59.5
			445	67.8

(a) Curves run in acetic acid.

TABLE XXVIII

METHODS OF FORMULATION

| Absorber Type	Solution Cast	Mill & Mold	Monomer Cast	Burnish	Dye	Vacuum Evap.
Fluorenol Salts	yes	yes	yes	yes	yes	yes
Triarylaminium Salts	yes	yes	no	yes	yes	no
Polymethine	yes	yes	yes	yes	–	no
Ni-Formazan	yes	yes	yes	(yes)	–	–
Pigments	–	yes	(no)	(yes)	no	(yes)

55

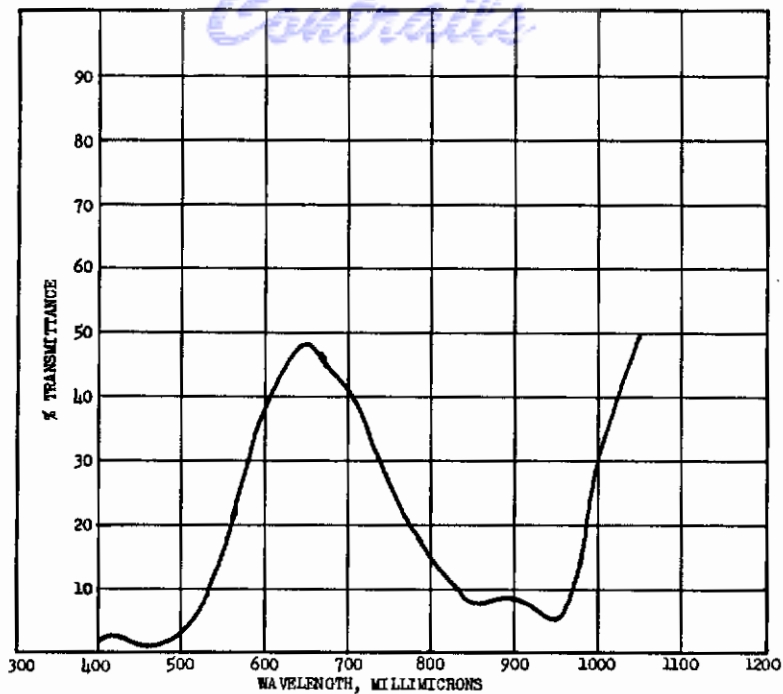APPENDIX II

Spectrophotometric Curves
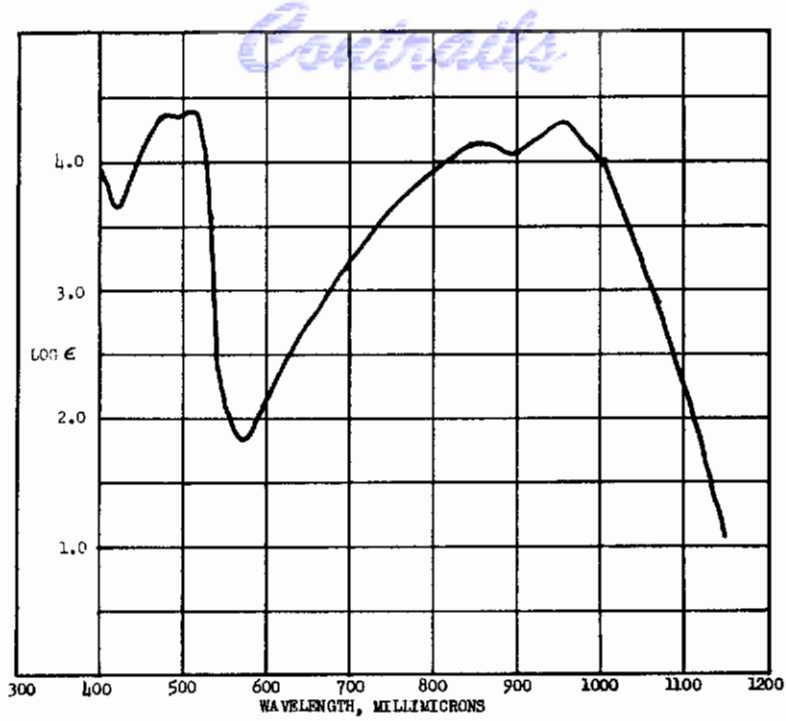
Spectral curve of 3,6-bis(dimethylamino)-9-phenylfluoren-9-ol (XII) in glacial acetic acid.
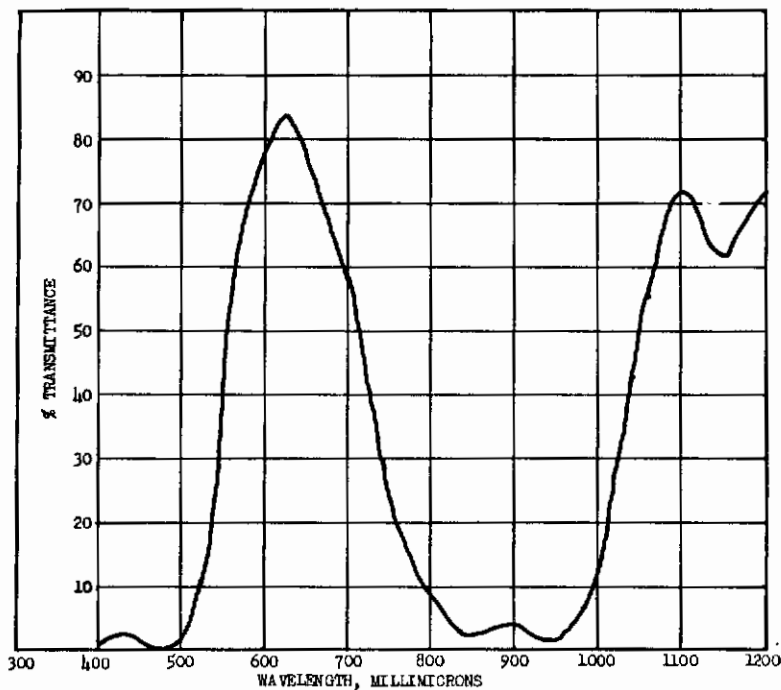
Figure 1
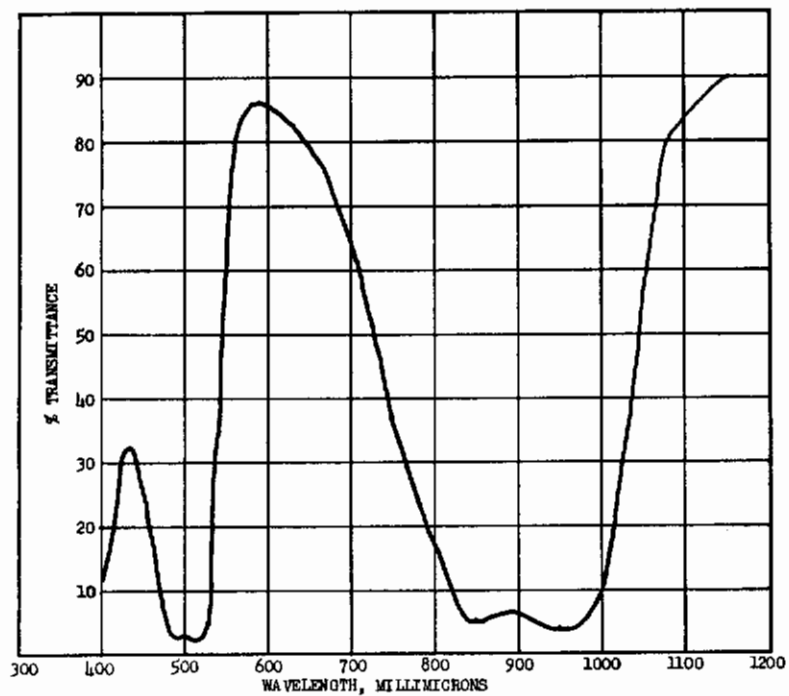
Spectral curve of the perchlorate salt (XXIII) of 3,6-bis(dimethylamino)-9-phenylfluoren-9-ol in solution-cast cellulose acetate coating on glass.
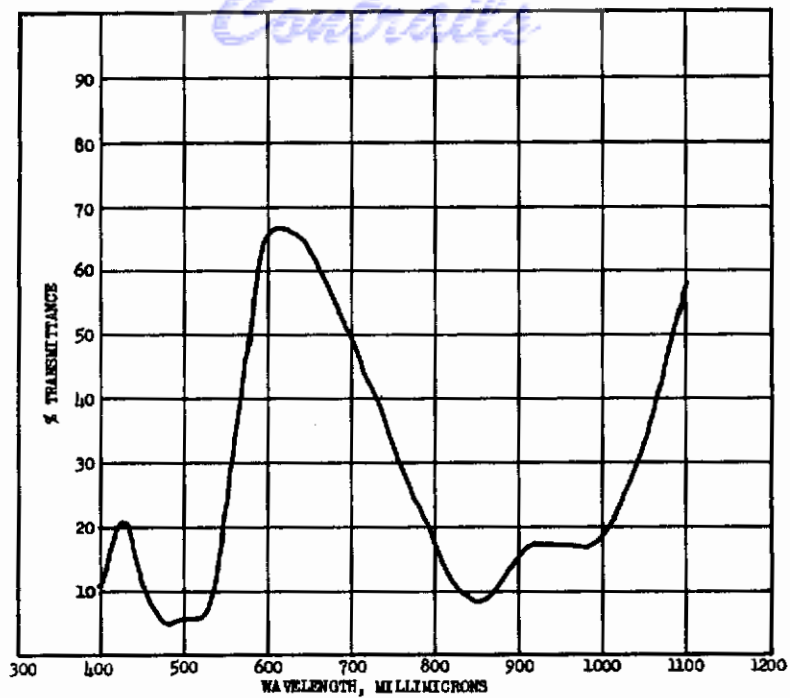
Figure 2

59

Spectral curve of the ethanesulfonate salt (XX) of 3,6-bis(dimethylamino)-9-phenylfluoren-9-ol in milled and molded poly(methyl methacrylate). Conc., 0.1%; thickness, 77 mils.

Figure 3

Spectral curve of the perfluorobutyrate salt (XIX) of 3,6-bis(dimethyl-amino)-9-phenylfluoren-9-ol in monomer-cast poly(methyl methacrylate). Conc., 0.2%; thickness, 113 mils.

Figure 4

60

Spectral curve of the tetracyanonickelate salt (XVI) of 3,6-bis(dimethyl-amino)-9-phenylfluoren-9-ol burnished on cellulose acetate film and overcoated with Acryloid B-72 resin.

Figure 5
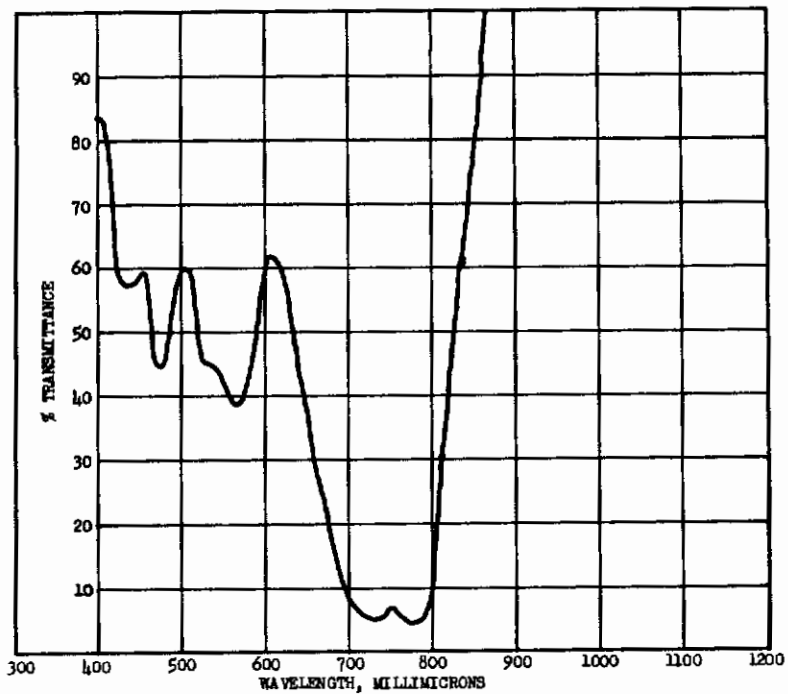
Spectral curve of 3-dimethylamino-9-(p-dimethylaminophenyl)fluoren-9-ol (XXIV) in glacial acetic acid. Conc., 0.01gm/l; cell, 1.00 cm.

Figure 6

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Legend:
— Natural sunlight, noon June
— — Weather-Ometer, 6000-watt xenon tube with clear glass filter
······· Fade-Ometer, single enclosed violet carbon, carbon arc 20-inch specimen rack

Spectral distribution of the Fade-Ometer and Xenon Weather-Ometer versus sunlight.*

* Data obtained from <u>Atlas Fade-Ometers and Weather-Ometers</u>, Atlas Electric Devices Company.

Figure 7
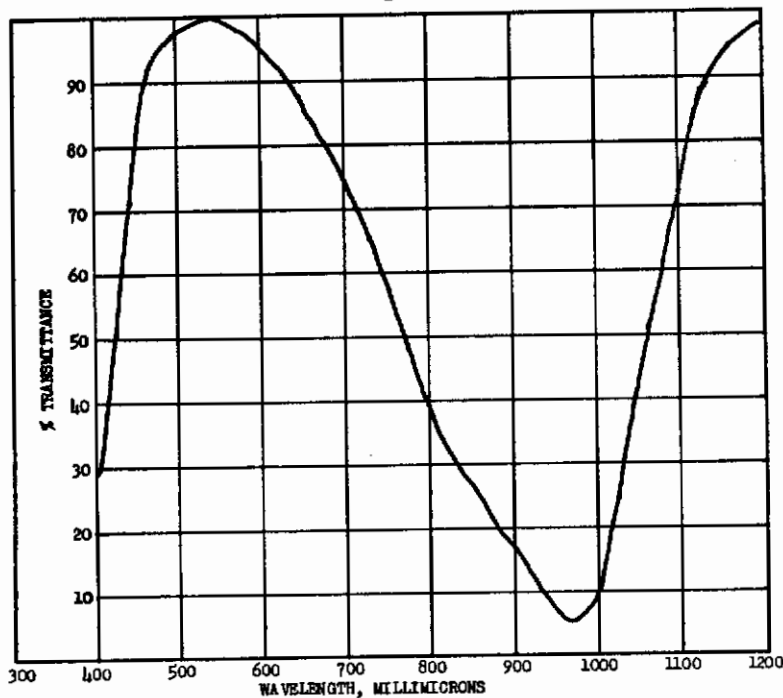
Spectral curve of tris(p-dimethylaminophenyl)aminium fluoborate (XXVIII) in methanol. Conc., 0.01gm/l; cell, 1.00 cm.

Figure 8
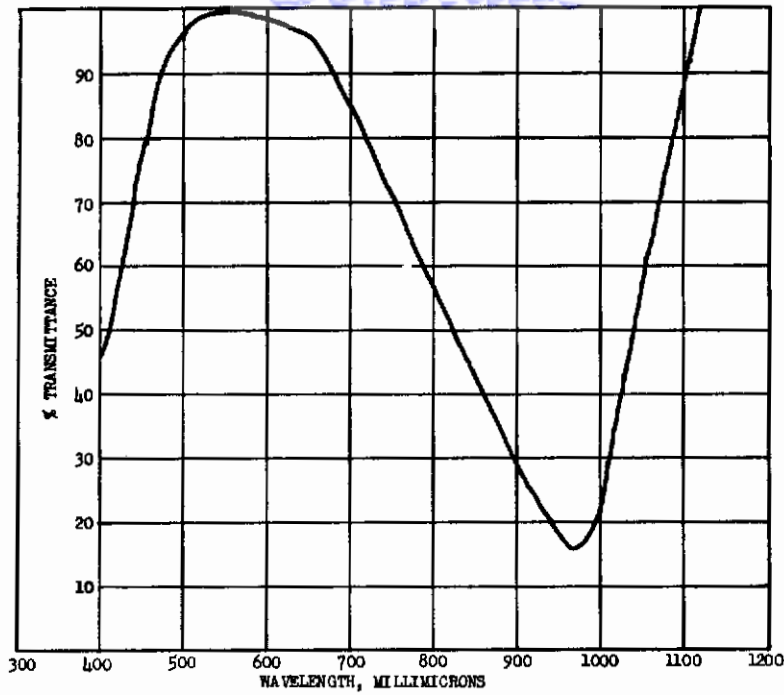
Spectral curve of tris(p-diethylaminophenyl)aminium fluoborate (XXX) in methanol. Conc., 0.01gm/l; cell, 1.00 cm.

Figure 9
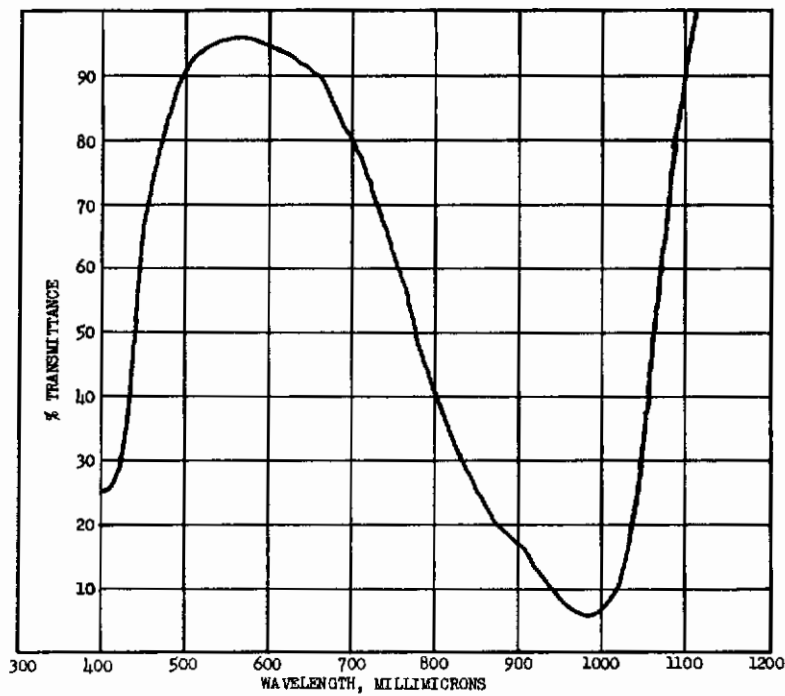
Spectral curve of tris(p-dibutylaminophenyl)aminium fluoborate (XLVII) in methanol. Conc., 0.028gm/l; cell, 1.00 cm.

Figure 10

63
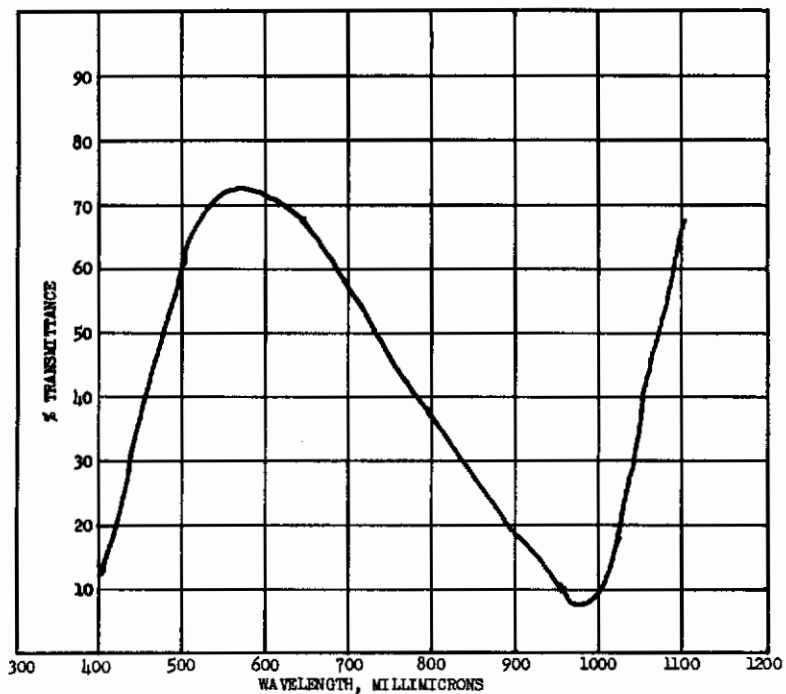
Spectral curve of tris(p-dimethylaminophenyl)aminium fluoborate (XXVIII) in milled and molded poly(methyl methacrylate). Conc., 0.05%; thickness 75 mils.
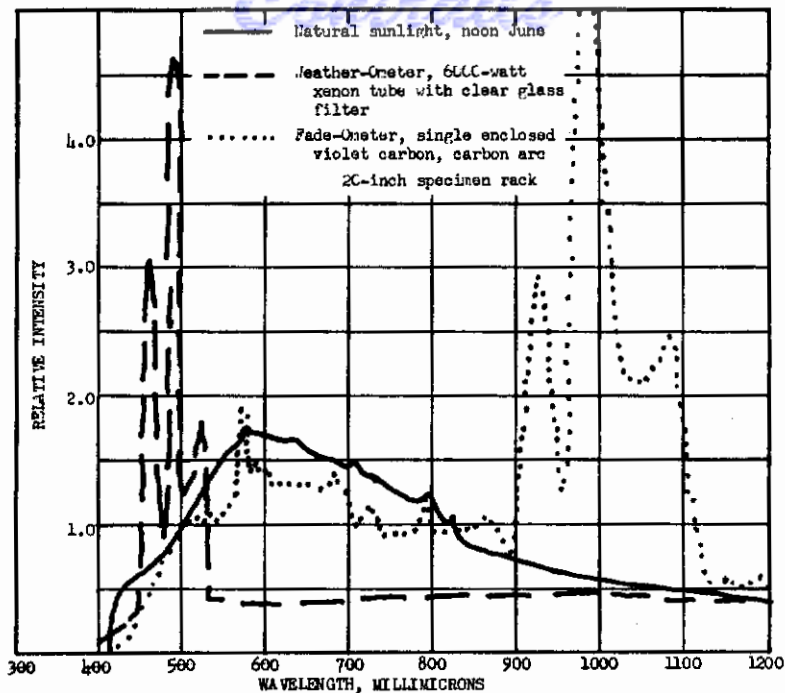
Figure 11
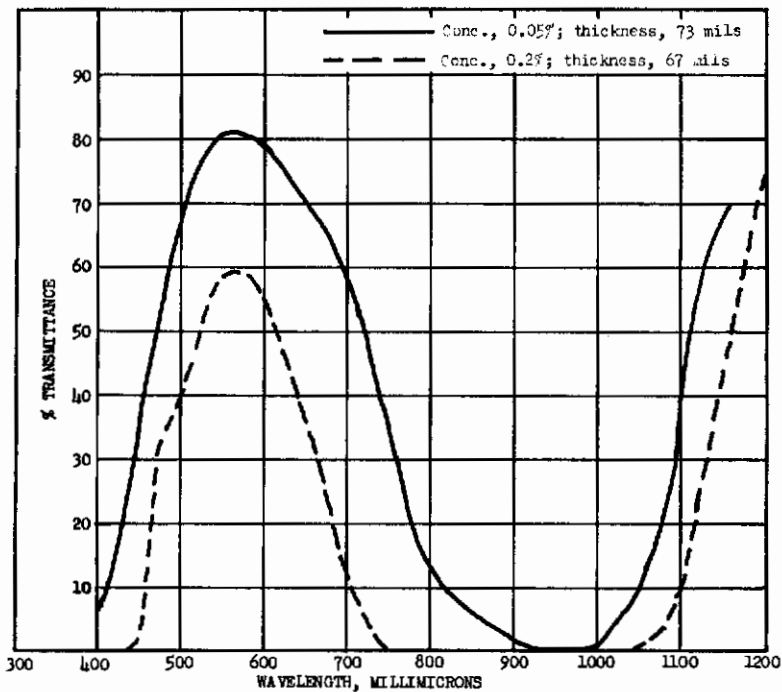
Spectral curve of tris(p-diethylaminophenyl)aminium fluoborate(XXX) in milled and molded poly(methyl methacrylate).

Figure 12

64
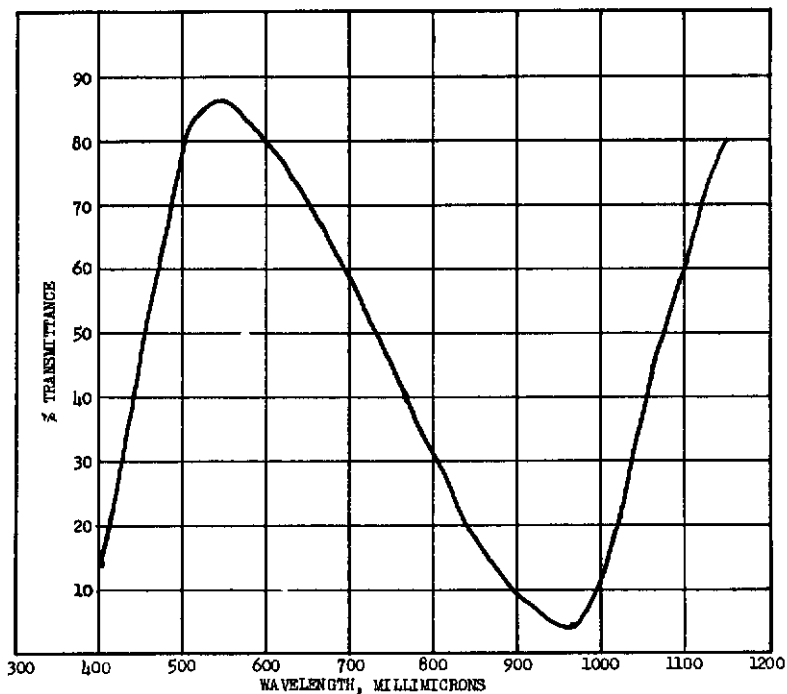
Spectral curve of tris(p-dimethylaminophenyl)aminium perchlorate (XXVII)
in solution-cast cellulose acetate coating on glass.

Figure 13
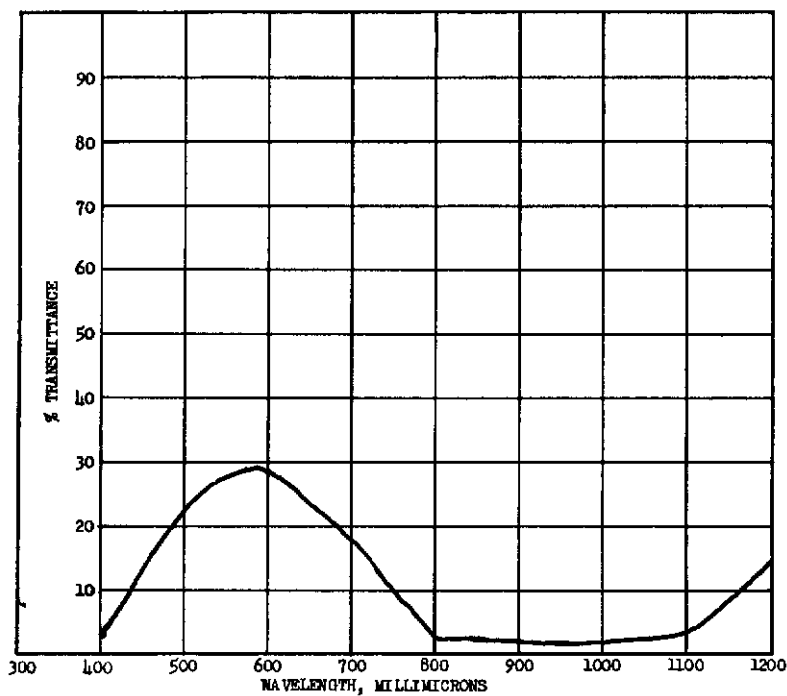
Spectral curve of tris(p-dimethylaminophenyl)aminium fluoborate (XXVIII)
burnished on poly(methyl methacrylate).

Figure 14
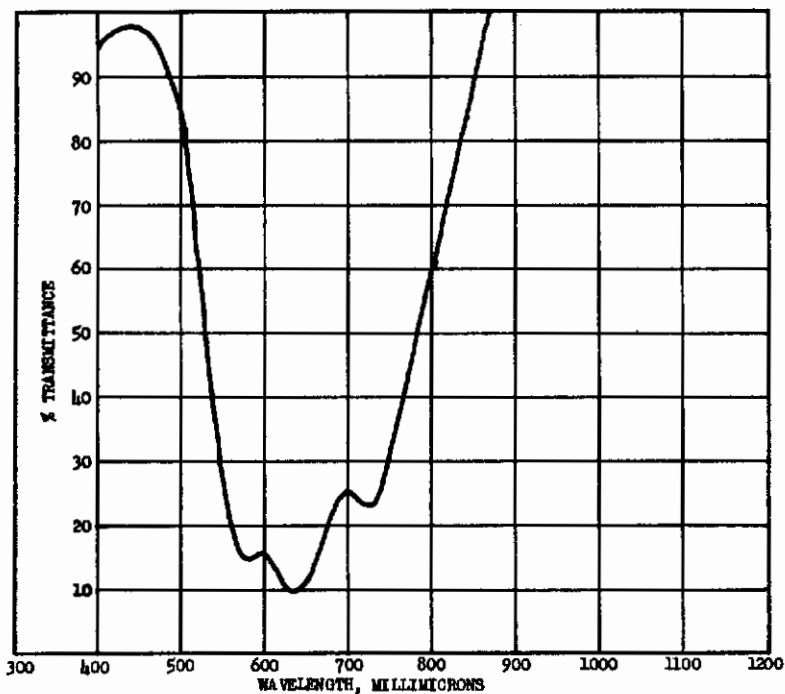
65

Spectral curve of 1,1,3,3-tetrakis(p-dimethylaminophenyl)vinylcarbonium perchlorate (XXXI) in acetone. Conc., 0.01gm/1; cell, 1.00 cm.

Figure 15
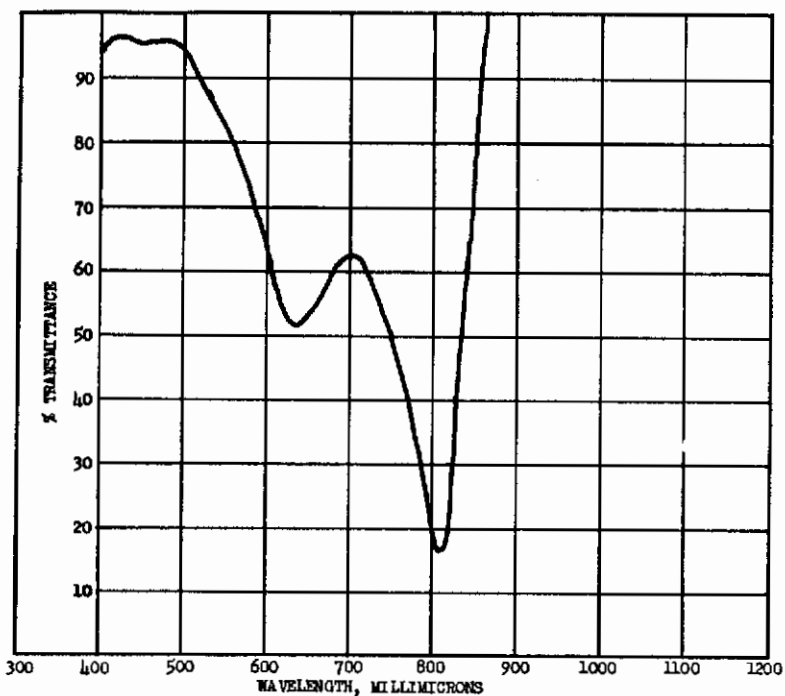
Spectral curve of 1,1,5,5-tetrakis(p-dimethylaminophenyl)divinylcarbonium perchlorate (XXXII) in acetone. Conc., 0.005gm/1; cell, 1.00 cm.

Figure 16
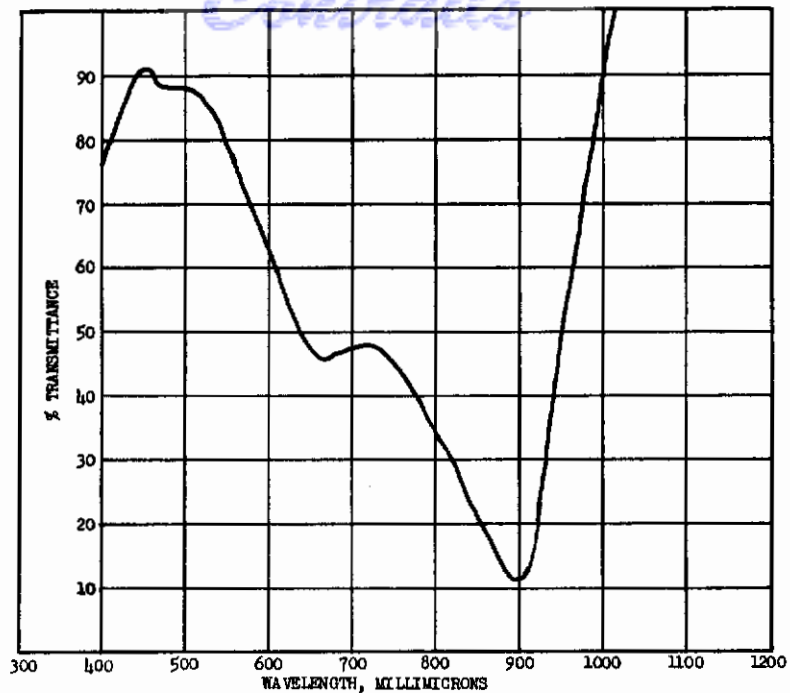
Spectral curve of 1,1,7,7-tetrakis(p-dimethylaminophenyl)trivinyl-carbonium perchlorate (XXXIII) in acetone. Conc., 0.005gm/l; cell, 1.00 cm.

Figure 17
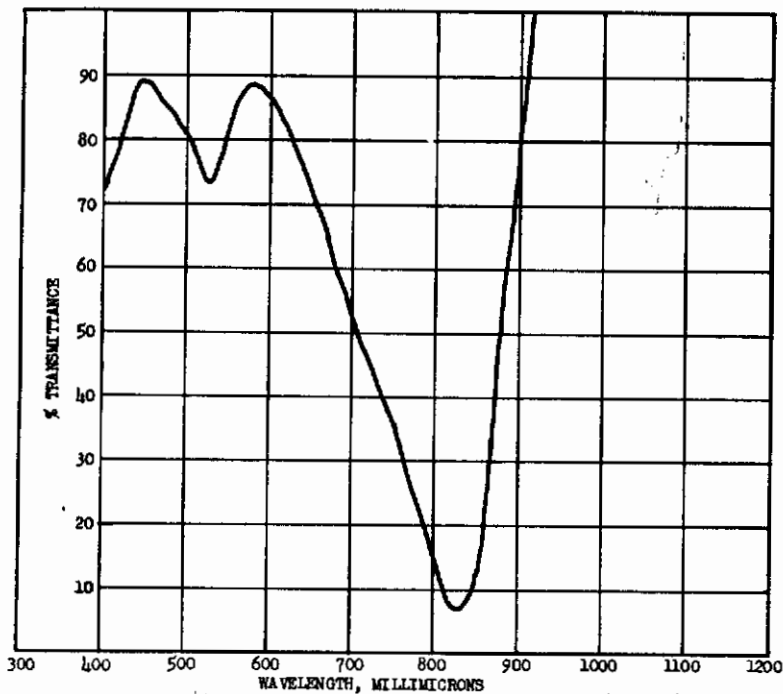
Spectral curve of 1,5-bis(p-dimethylaminophenyl)-1,5-diphenyldivinyl-carbonium perchlorate (XXXIV) in acetone. Conc., 0.005gm/l; cell, 1.00 cm.

Figure 18

67
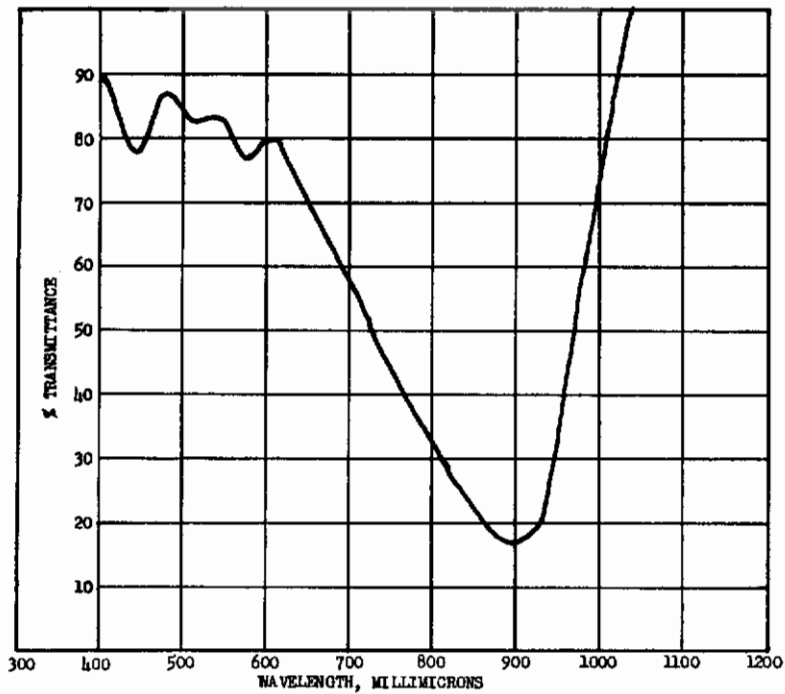
Spectral curve of 1,7-bis(p-dimethylaminophenyl)-1,7-diphenyltrivinyl-carbonium perchlorate (XXXV) in acetone. Conc., 0.005gm/l; cell, 1.00 cm.

Figure 19
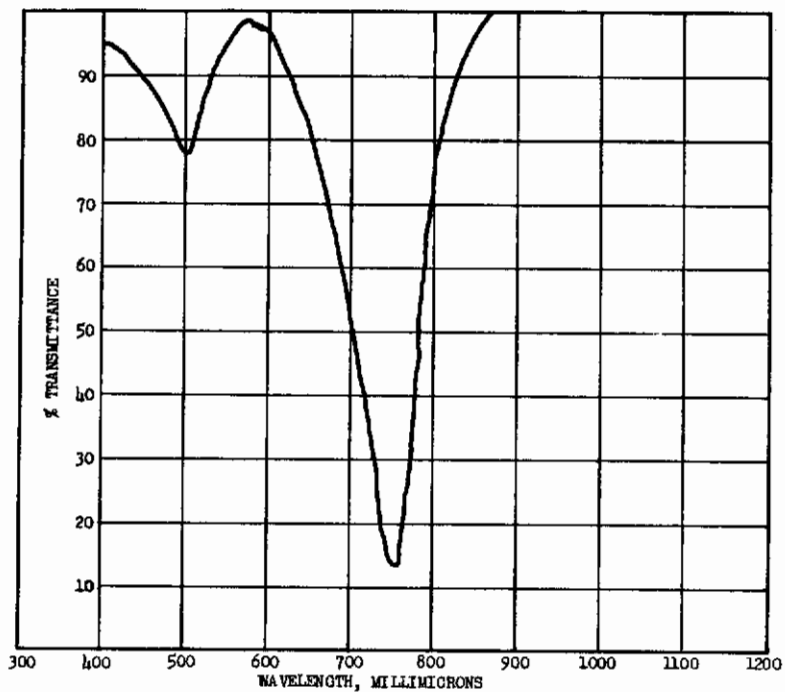
Spectral curve of 1,1,7,7-tetrakis(p-methoxyphenyl)trivinylcarbonium per-chlorate (XXXVI) in glacial acetic acid. Conc., 0.005gm/l; cell, 1.00 cm.

Figure 20
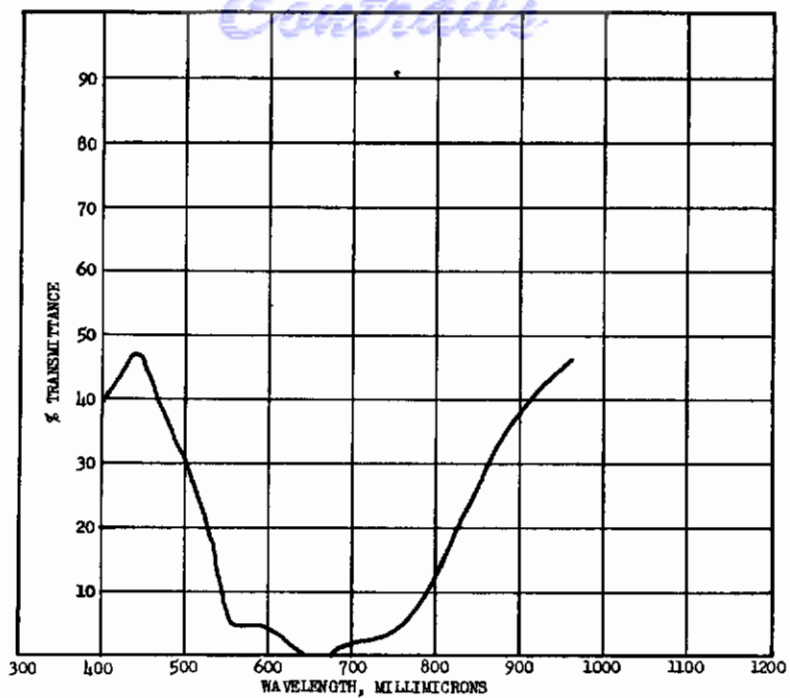
Spectral curve of 1,1,3,3-tetrakis(p-dimethylaminophenyl)vinylcarbonium perchlorate (XYXI) in monomer-cast poly(methyl methacrylate). Conc., 0.05%; thickness 133 mils.

Figure 21
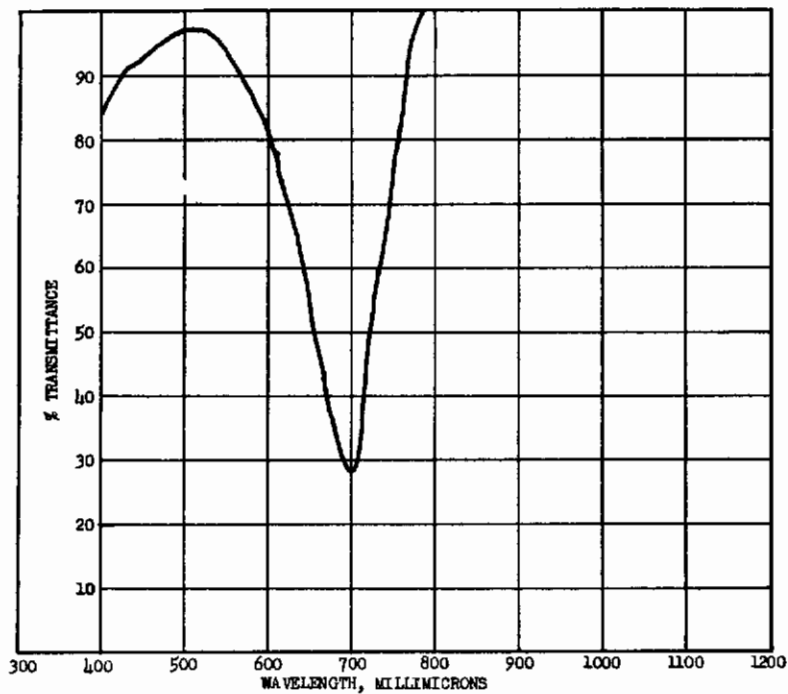
Spectral curve of bis(xanthene)trimethine perchlorate (XXXVII) in acetone. Conc., 0.01gm/1; cell, 1.00 cm.

Figure 22

69
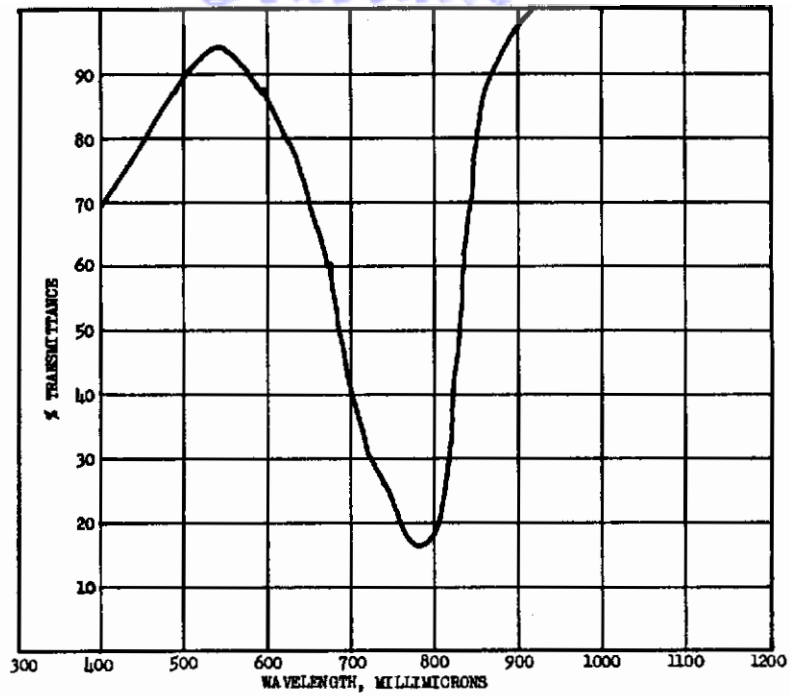
Spectral curve of bis(xanthene)pentamethine perchlorate (XXXVIII) in acetone. Conc., 0.01gm/l; cell, 1.00 cm.

Figure 23
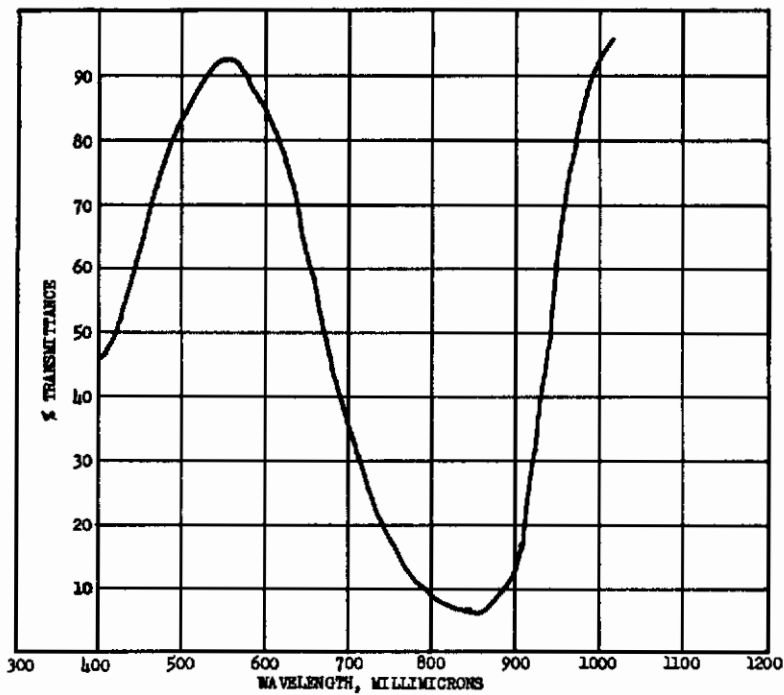
Spectral curve of bis(xanthene)heptamethine perchlorate (XXXIX) in aceto-nitrile. Conc., 0.01gm/l; cell, 1.00 cm.

Figure 24

70
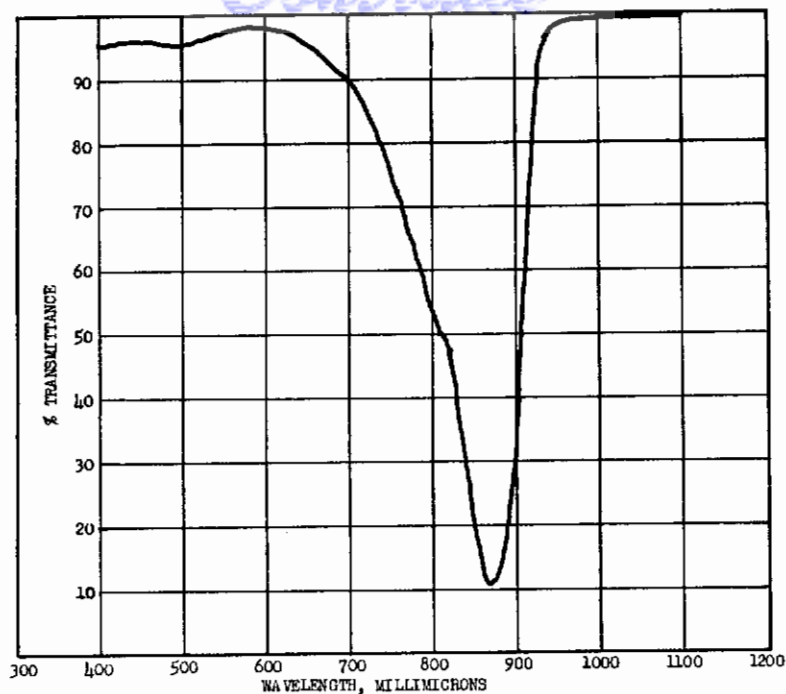
Spectral curve of bis(3,6-dimethoxyxanthene)heptamethine perchlorate (XL) in acetone. Conc., 0.01gm/l; cell, 1.00 cm.

Figure 25
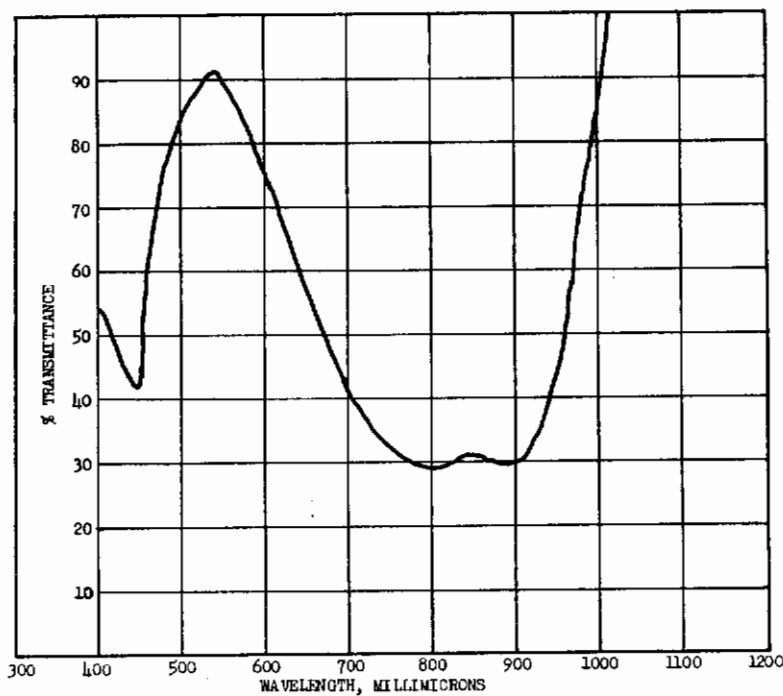
Spectral curve of 5-isopropyl-1-/5-(5-isopropyl-3,8-dimethylazulene-1-yl)- penta-2,4-dienylidene7-3,8-dimethylazulenium perchlorate (XLI) in acetic acid. Conc., 0.002gm/l; cell, 1.00 cm.

Figure 26
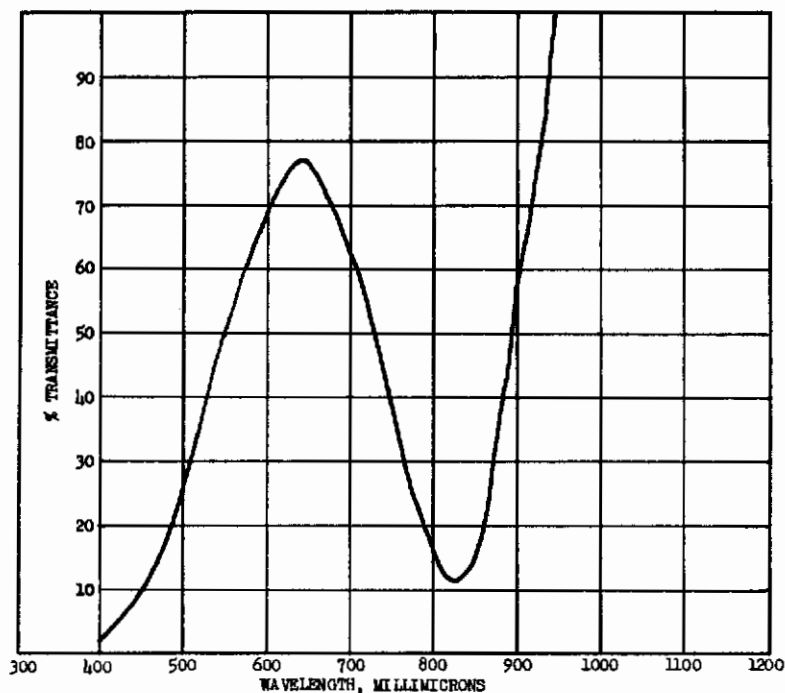
71

Spectral curve of the nickel complex of N-phenyl-N'-(2-methoxyphenyl)-C-phenyl formazan (XLIII) in benzene. Conc., 10^{-4}M; cell, 1.00 cm.

Figure 27
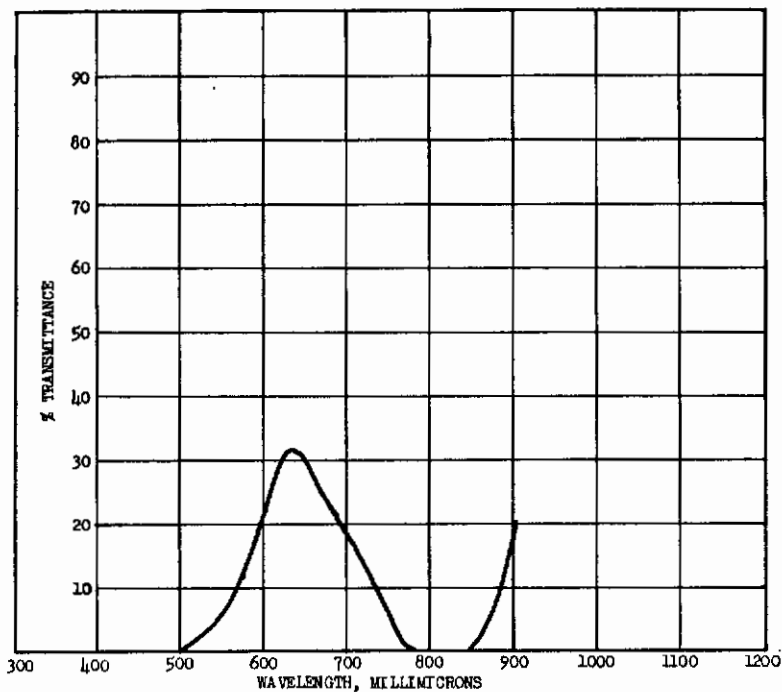
Spectral curve of the nickel complex of N-phenyl-N'-(2-methoxyphenyl)-C-phenyl formazan (XLIII) in monomer-cast poly(methyl methacrylate). Conc., 0.05%; thickness, 131 mils.

Figure 28

72
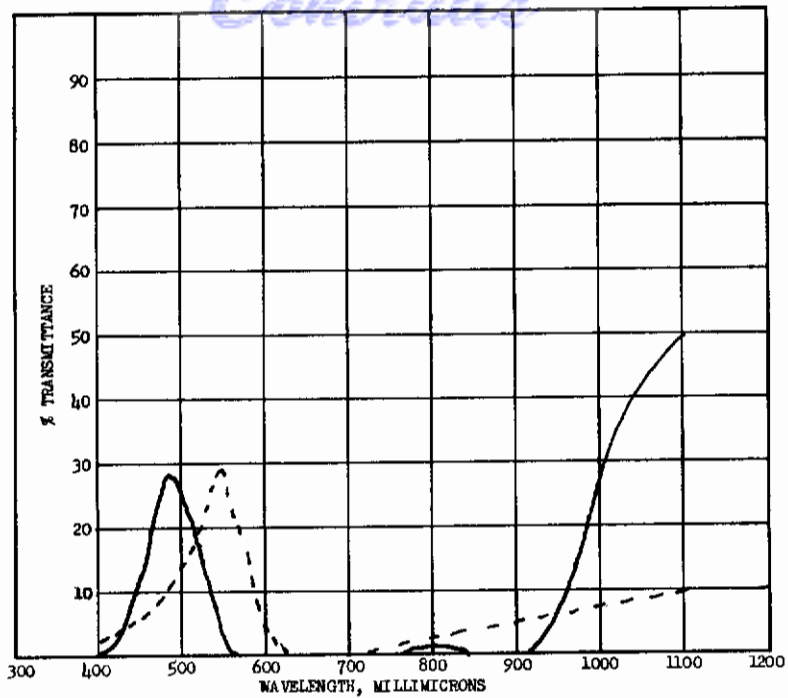
Spectral curve of dihydroxygermanium phthalocyanine (solid curve) and of diphenoxygermanium phthalocyanine (dashed curve) in milled and molded poly-(methyl methacrylate). Conc., 0.1% each; thickness, 65 mils each.

Figure 29
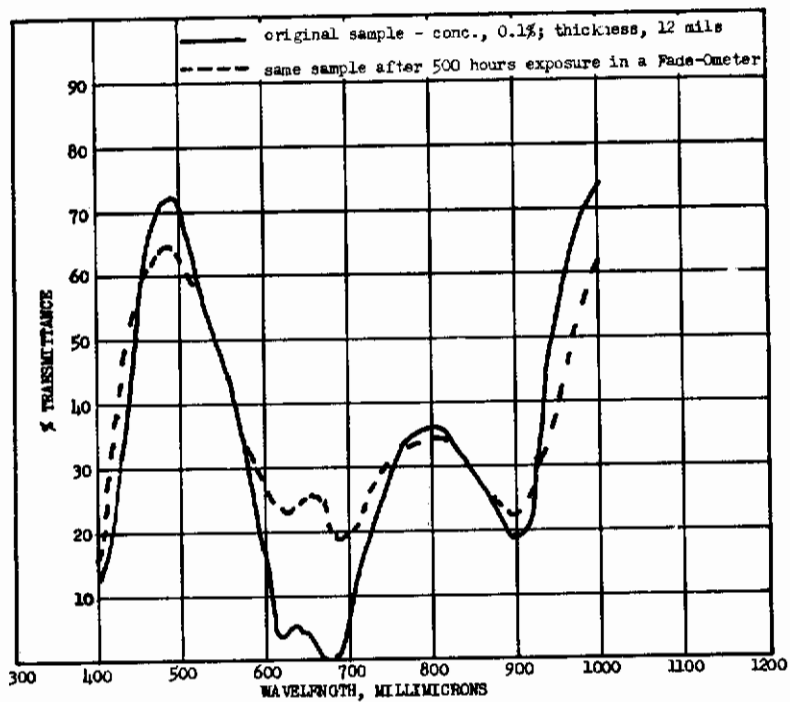
Light stability of dihydroxygermanium phthalocyanine in poly(methyl-methacrylate).

Figure 30

73
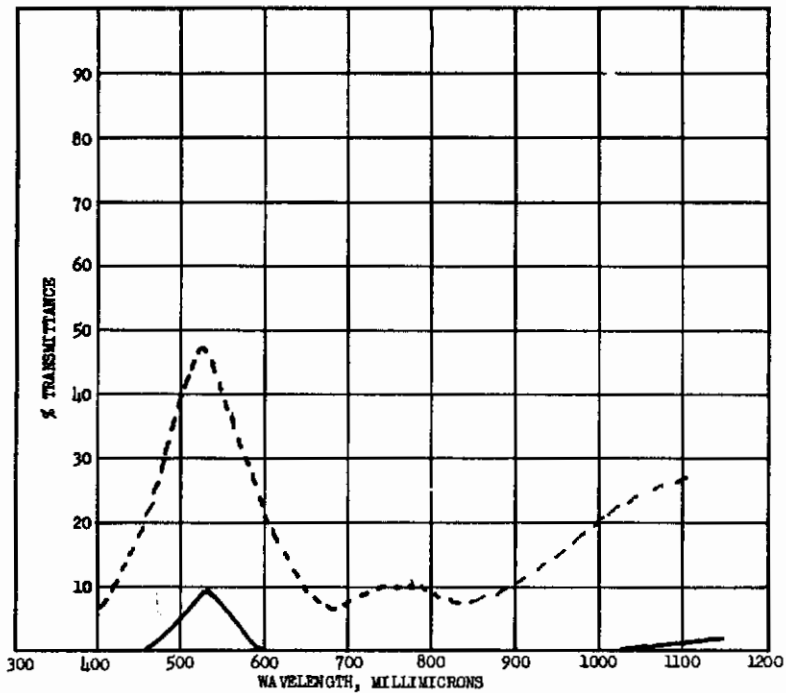
Spectral curves of vanadyl phthalocyanine in milled and molded poly(methyl-methacrylate). Solid curve: conc., 0.1%; thickness, 57 mils. Dashed curve: conc., 0.1%; thickness, 17 mils.

Figure 31
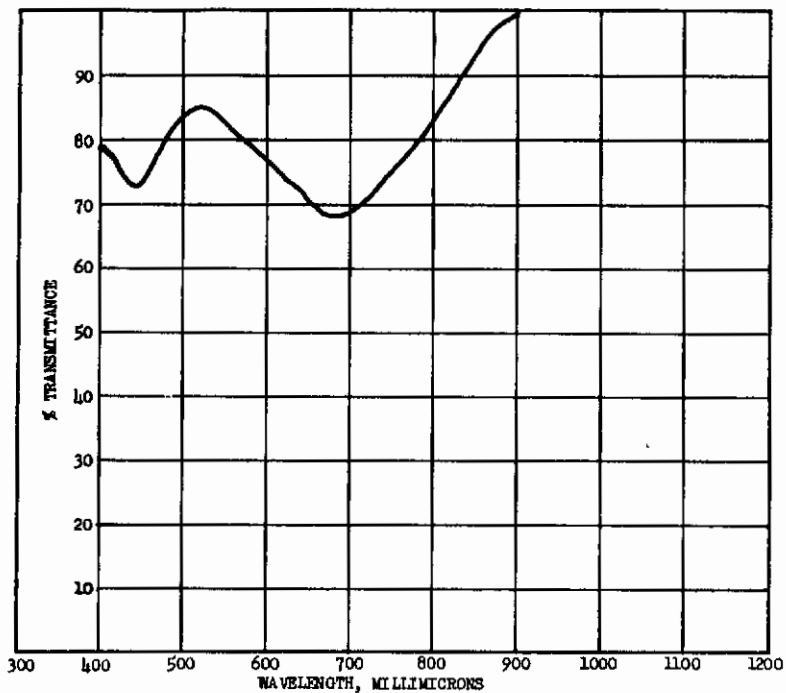
Spectral curve of the phenothiazine derivative from benzophenone (XLIV) in glacial acetic acid. Conc.; 0.01gm/l; cell, 1.00 cm.

Figure 32

74
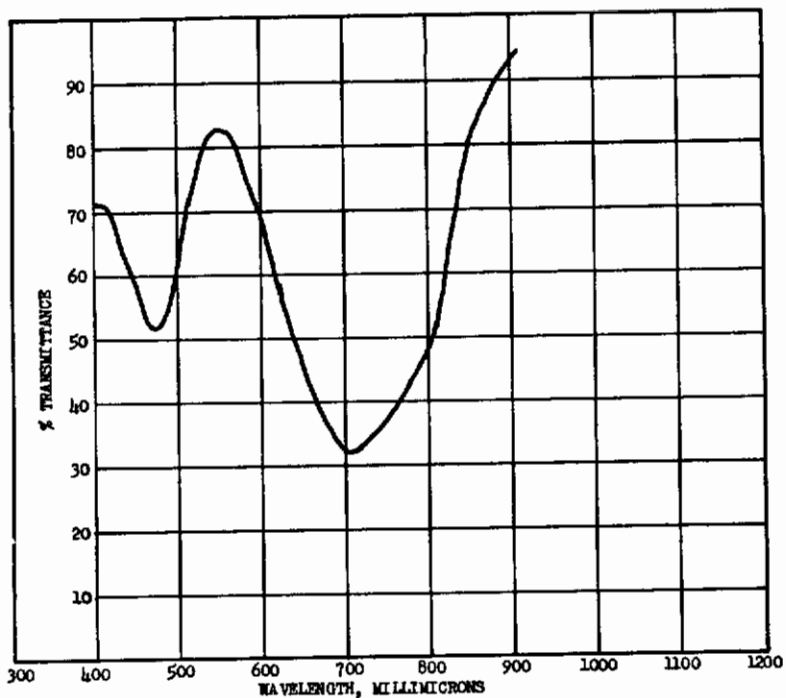
Spectral curve of the phenothiazine derivative from p-dimethylaminobenzo-
phenone (XLV) in glacial acetic acid. Conc., 0.01gm/l; cell, 1.00 cm.
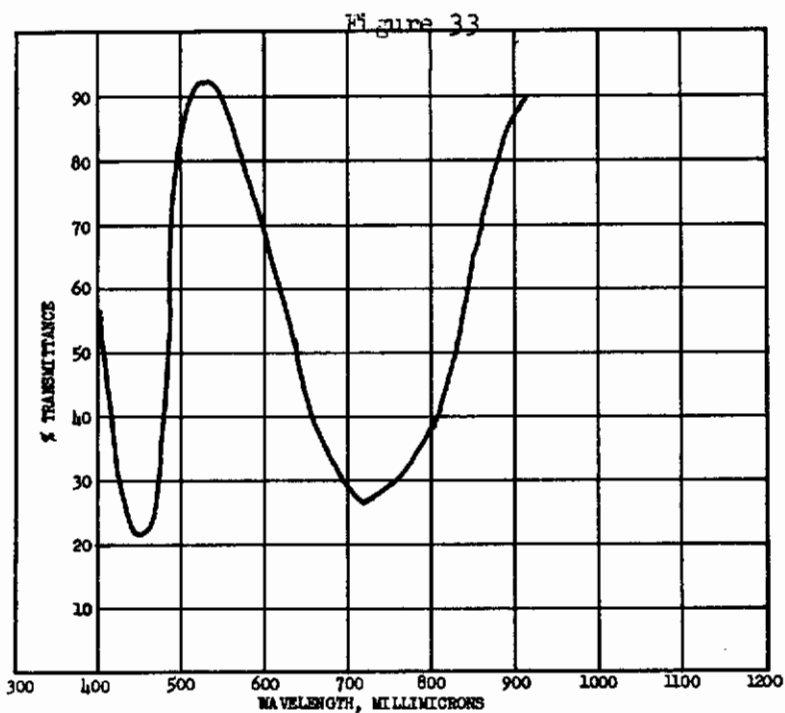
Figure 33

Spectral curve of the phenothiazine derivative from p,p'-dimethoxybenzo-
phenone (XLVI) in glacial acetic acid. Conc., 0.01gm/l; cell, 1.00 cm.

Figure 34

75
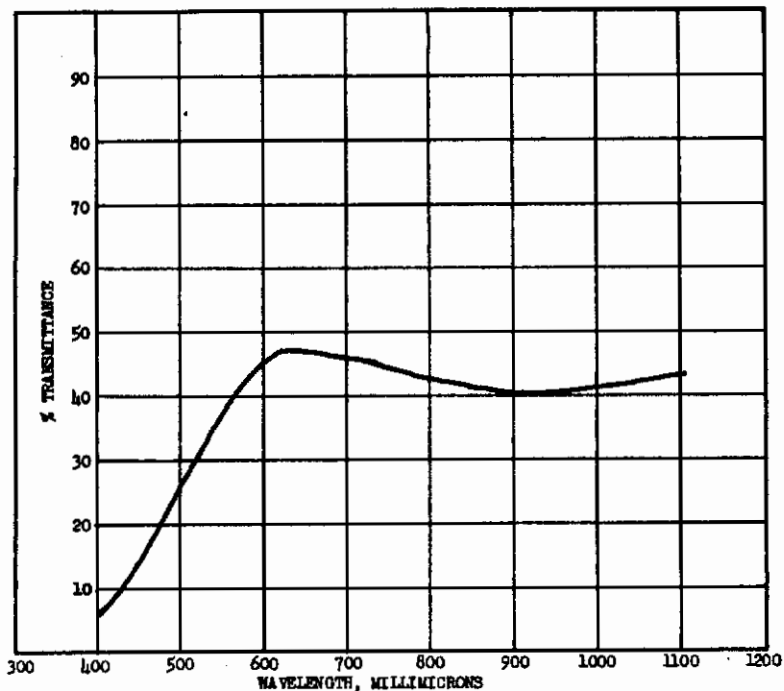
Spectral curve of finely ground potassium chloride (-325 mesh) dispersed in monomer-cast poly(methyl methacrylate). Conc., 33%; thickness, 130 mils.

Figure 35
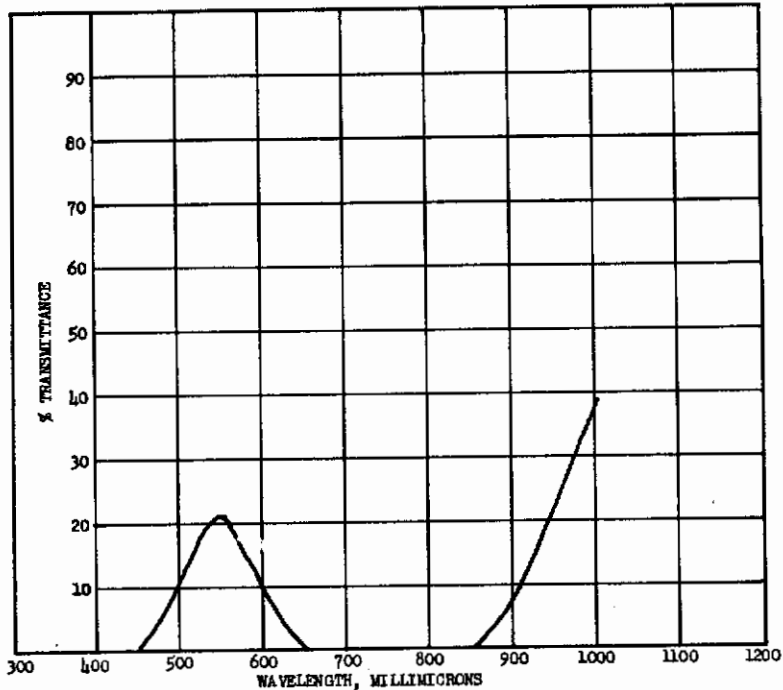
Spectral curve of Pigment Green B in milled and molded poly(methyl-methacrylate). Conc., 0.1%; thickness, 60 mils.

Figure 36

APPENDIX III

Compounds with no Near-Infrared Absorption

Formula	Name
$AsN_3C_3C_{16}H_{14}$	Benzenearsonic acid, p-(2-amino-1-naphthylazo)-
$AsO_3CH_3Ag_2$	Methanearsonic acid, disilver salt
AsO_3CH_3Cu	Methanearsonic acid, dicopper[II] salt
$BiCl_3C_{12}H_{10}$	Bismuth chloride (H_2BiCl_3), diphenyl-
$C_{10}H_8$	Azulene
$C_{21}H_{15} \cdot BF_4$	Cyclopropenacarbonium fluoborate-1,2,3-triphenyl
$CoHgN_4S_4C_4$	Cobalt mercury thiocyanate $[Co(NCS)_4Hg]$
$FC_{16}H_9$	Fluoranthene, 3-fluoro-
$F_2C_{13}H_8$	Fluorene, 2,7-difluoro-
$GeC_{24}H_{20}$	Germane, tetraphenyl-
$IC_{12}H_{10}Br$	Iodonium bromide, diphenyl
$I_2C_{12}H_8$	Biphenyl, 4,4'-diiodo-
$NC_6H_7 \cdot N_3O_7C_6H_3$	3-Picoline, picrate
$NC_6H_{14} \cdot I$	Pyrrolidinium iodide, 1,1-dimethyl-
$NC_{11}H_{12} \cdot SO_4CH_3$	Quinaldinium methyl sulfate, 1-methyl
$NC_{13}H_{16} \cdot I$	Ammonium iodide, trimethyl (2-naphthyl)
$NC_{21}H_{38} \cdot I$	Ammonium iodide, benzyldodecyldimethyl-
$NOBrC_{13}H_8$	9-Fluorenone, 2-amino-3-bromo-
$NOC_9H_6Cu_{1/2}$	8-Quinolinol, copper[II] derivative
$NOC_{19}H_{17} \cdot ClO_4$	Perinaphthoxolium perchlorate
$NOC_{20}H_{17}$	Formamide, N-trityl
$NOC_{25}H_{22} \cdot ClO_4$	3-(p-Dimethylamino)-2,6-diphenylpyrylium perchlorate
$NOClF_3C_{14}H_8Ni_{1/2}$	o-Cresol, α-(6-chloro-α,α,α-trifluoro-m-tolylimino)-, nickel[II] derivative
$NO_2BrC_{10}H_5Cu_{1/2}$	1,2-Naphthoquinone, 3-bromo-, 1-oxime, copper[II] derivative
$NO_2Br_2C_{10}H_4Cu_{1/2}$	1,2-Naphthoquinone, 3,6-dibromo-, 1-oxime, copper[II] derivative
$NO_2C_2H_4Cu_{1/2}$	Glycine, copper[II] salt
$NO_2C_7H_7 \cdot NC_5H_5 \cdot HCl$	Pyridinium hydroxide, 1-(carboxymethyl)-, inner salt, compound with pyridine, hydrochloride
$NO_2C_{10}H_7$	1,2(and 1,4)-Naphthoquinone, 2(and 1)-oxime
$NO_2C_{13}H_{19}$	Benzoic acid, p-dimethylamino-, isobutyl ester
$NO_2C_{14}H_7$	6H-Anthra-$[1,9$-cd$]$-isoxazole-6-one
$NO_2C_{14}H_9$	Phenanthrenequinone, oxime
$NO_2C_{14}H_{22} \cdot Br$	Ammonium bromide, (p-carboxybenzyl)triethyl-
$NO_2C_{17}H_{20} \cdot Br$	Ammonium bromide, benzyl(p-carboxybenzyl)-dimethyl-
$NO_2ClC_{10}H_6$	1,2-Naphthoquinone, 4-chloro-, 2-oxime

Formula	Name
$NO_3C_6H_5$	Resorcinol, 4-nitroso-
$NO_3C_{10}H_7$	1,2-Naphthoquinone, 6-hydroxy-, 2-oxime
$NO_3C_{10}H_7$	1,2-Naphthoquinone, 3-hydroxy-, 1-oxime
$NO_3C_{11}H_9$	1,2-Naphthoquinone, 7-methoxy-, 1-oxime
$NO_3C_{11}H_{12}Cu_{1/2}$	Glycine, N-salicylidene-, ethyl ester, copper[II] derivative
$NO_3C_{14}H_{10}Ni_{1/2}$	Benzoic acid, p-salicylideneamino-, nickel[II] derivative
$NO_3C_{14}H_{14} \cdot ClO_4$	Dianisyl nitroxide perchlorate
$NO_3C_{22}H_{17}$	Anthraquinone, 1-hydroxy-4-(2,4-xylidino)-
$NO_3C_{27}H_{17}$	4-Biphenylcarboxamide, N-1-anthraquinonyl
$NO_3Cl_4C_{15}H_5$	Pyridinium hydroxide, 1-(5,6,7,8-tetrachloro-1,4-dihydro-3-hydroxy-1,4-dioxo-2-naphthyl)-, inner salt
$NO_3IC_7H_6$	Anisole, 2-iodo-4-nitro
$NO_4BrC_{21}H_{12}$	Anthraquinone, 1-(5-bromosalicylamido)-
$NO_4BrC_{21}H_{12}$	Anthraquinone, 1-(3-bromosalicylamido)-
$NO_4Br_2C_{21}H_{11}$	Anthraquinone, 1-(3,5-dibromosalicylamido)-
$NO_4Br_4C_{21}H_9$	Anthraquinone, 1-(3,4,5,6-tetrabromosalicylamido)-
$NO_4C_{12}H_9$	2-Naphthoic acid, 3,4-dihydro-3,4-dioxo-, methyl ester, 4-oxime
$NO_4C_{12}H_{11}$	1,2-Naphthoquinone, 5,6-dimethoxy-, 1-oxime
$NO_4C_{17}H_{11}$	1,2-Naphthoquinone, 6-hydroxy-, 1-oxime, 6-benzoate
$NO_4C_{21}H_{13}$	Spiro/2H-1-benzopyran-2,3'-/3H/naphtho/2,1-b/-pyran, 6-nitro-
$NO_4C_{22}H_{15}$	Anthraquinone, 1-benzamido-4-methoxy-
$NO_4C_{22}H_{15}$	2,3-Cresotamide, N-1-anthraquinonyl-
$NO_4C_{22}H_{15}$	Anthraquinone, 1-m-anisamido-
$NO_4C_{22}H_{17}$	Anthraquinone, 1-hydroxy-4-/p-(2-hydroxyethyl)-anilino/-
$NO_4C_{23}H_{17}$	Anthraquinone, 1-(o-ethoxybenzamido)-
$NO_4C_{25}H_{15}$	2-Naphthamide, N-1-anthraquinonyl-1-hydroxy-
$NO_4C_{27}H_{17}$	Salicylamide, N-1-anthraquinonyl-5-phenyl-
$NO_4C_{27}H_{17}$	Salicylamide, N-1-anthraquinonyl-3-phenyl
$NO_4ClC_{21}H_{12}$	Anthraquinone, 1-(4-chlorosalicylamido)-
$NO_4ClC_{21}H_{12}$	Anthraquinone, 1-(5-chlorosalicylamido)-
$NO_4ClC_{21}H_{12}$	Anthraquinone, 1-(3-chlorosalicylamido)-
$NO_4ClC_{22}H_{14}$	Anthraquinone, 1-p-chlorobenzamido-4-methoxy
$NO_4ClC_{22}H_{16}$	Anthrarufin, 8-chloro-4-(2,3-xylidino)-
$NO_4Cl_2C_{21}H_{11}$	Anthraquinone, 1-(3,4-dichlorosalicylamido)-
$NO_4Cl_2C_{21}H_{11}$	Anthraquinone, 3-chloro-1-(3-chlorosalicylamido)-
$NO_4Cl_2C_{21}H_{11}$	Anthraquinone, 4-chloro-1-(3-chlorosalicylamido)-

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Formula	Name
$NO_4Cl_2C_{21}H_{11}$	Anthraquinone, 6-chloro-1-(3-chlorosalicylamido)-
$NO_4Cl_2C_{21}H_{11}$	Anthraquinone, 8-chloro-1-(3-chlorosalicylamido)-
$NO_4Cl_2C_{21}H_{11}$	Anthraquinone, 1-(3,5-dichlorosalicylamido)-
$NO_4Cl_3C_{20}H_{10}$	Anthrarufin, 4-chloro-8-(3,4-dichloroanilino)-
$NO_4Cl_3C_{21}H_{10}$	Anthraquinone, 6,7-dichloro-1-(3-chlorosalicylamido)-
$NO_4FC_{21}H_{12}$	Anthraquinone, 1-(3-fluorosalicylamido)-
$NO_4I_2C_{21}H_{11}$	Anthraquinone, 1-(3,5-diiodosalicylamido)-
$NO_5C_{21}H_{13}$	Gentisamide, N-1-anthraquinonyl-
$NO_5C_{21}H_{13}$	β-Resorcylamide, N-1-anthraquinonyl-
$NO_5C_{21}H_{13}$	γ-Resorcylamide, N-1-anthraquinonyl-
$NO_5C_{22}H_{15}$	Anthraquinone, 1-methoxy-4-salicylamido)-
$NO_5C_{22}H_{15}$	Anthraquinone, 1-(2-hydroxy-p-anisamido)-
$NO_5C_{23}H_{17}$	Anthraquinone, 1-m-anisamido-4-methoxy-
$NO_5ClC_{21}H_{11}$	Anthraquinone, 1-(3-chlorosalicylamido)-4-hydroxy-
$NO_5Cl_2C_{22}H_{13}$	Anthraquinone, 1-(3,5-dichlorosalicylamido)-4-methoxy-
$NSC_{25}H_{22} \cdot C_4Cl$	Thiopyrylium perchlorate, 4-(p-dimethylaminophenyl)-2,6-diphenyl-
$NSOC_{13}H_{26}Ni_{1/2}$	Nickel complex of dihexyl dithiocarbamate
$NSO_4C_{11}H_{11}$	Isoxazolium hydroxide, 2-ethyl-5-m-sulfophenyl-, inner salt
$NSC_5C_{10}H_6Na$	1-Naphthalenesulfonic acid, 3,4-dihydro-3,4-dioxo-, 3-oxime, sodium salt
$NS_2C_6H_4Ni_{1/2}$	Nickel complex of mercaptobenzothiazole
$NS_2C_9H_{10}Zn_{1/2}$	Zinc, N-benzyl-N-methyldithiocarbamate
$NS_2C_{10}H_{12} \cdot SO_3C_7H_9$	Benzothiazolium p-toluenesulfonate-3-ethyl-2-(mercaptomethyl)
$NS_2C_{11}H_{12} \cdot HSO_4$	1,2-Dithiolium H sulfate, 5-(p-dimethylaminophenyl)-
$NS_2C_{13}H_{22}Zn_{1/2}$	Zinc, N,N-dicyclohexyl dithiocarbamate
$NS_2C_{14}H_{18}Zn_{1/2}$	Zinc, N-benzyl-N-cyclohexyl dithiocarbamate
$NS_2C_{15}H_{14}Ba_{1/2}$	Barium, dibenzyldithiocarbamate
$NS_2C_{15}H_{14}Ca_{1/2}$	Calcium, dibenzyldithiocarbamate
$NS_2C_{15}H_{14}Cl_{1/2}$	Cadmium, dibenzyldithiocarbamate
$NS_2C_{15}H_{14} \cdot ClO_4$	3,4-Benzo-5-(p-N,N-dimethylaminophenyl)-1,2-dithiolium perchlorate
$NS_2C_{15}H_{14}Co_{1/2}$	Cobalt, dibenzyldithiocarbamate
$NS_2C_{15}H_{14}Cu_{1/2}$	Copper, dibenzyldithiocarbamate
$NS_2C_{15}H_{14}Fe_{1/2}$	Iron, dibenzyldithiocarbamate
$NS_2C_{17}H_{16} \cdot ClO_4$	4-Phenyl-5-(p-N,N-dimethylaminophenyl)-1,2-dithiolium perchlorate
$NS_2C_{17}H_{34}Ni_{1/2}$	Bis(N,N-di-2-ethylhexyldithiocarbamate)nickel (II)
$NS_2C_{17}H_{34}Zn_{1/2}$	Zinc, di-n-octyl dithiocarbamate

Formula	Name
$NS_2C_{19}H_{18} \cdot ClO_4$	1,2-Dithiolium perchlorate, 5-(p-dimethylaminostyryl)-3-phenyl-
$NS_2C_{19}H_{18} \cdot ClO_4$	1,2-Dithiolium perchlorate, 5-(p-dimethylaminostyryl)-4-phenyl
$NS_2C_{22}H_{18} \cdot ClO_4$	1,2-Dithiolium perchlorate, 5-(p-N-methylanilinophenyl)-3-phenyl
$NS_2O_8C_{10}H_5Na_2$	2,7-Naphthalenedisulfonic acid, 3,4-dihydro-3,4-dioxo-, 4-oxime, disodium salt
$N_2C_2H_8 \cdot 1/2O_3ICCo$	Ethylenediamine, compound with $CoCO_3I$
$N_2C_8H_{14}$	Pyrazine, 2,5-dihydro-2,2,5,5-tetramethyl
$N_2C_{11}H_{11} \cdot BF_4$	Benz-5,7-dimethyl-1,4-diazepinium fluoborate
$N_2C_{13}H_{10}$	1-Phenalenone, hydrazone
$N_2C_{15}H_{16}$	2,3-Fluorenediamine, N^2, N^2-dimethyl-
$N_2C_{18}H_{17} \cdot Cl \cdot HCl$	Quinolinium chloride, 2-(p-aminostyryl)-1-methyl-, hydrochloride
$N_2C_{19}H_{17} \cdot NO_3$	Sempervirine nitrate
$N_2C_{20}H_{16}$	Aniline, N,N'-(p-phenylenedimethylidyne)di-
$N_2C_{20}H_{17} \cdot SO_3C_6H_5$	Quinolinium benzenesulfonate, 2-/2-(3-indolyl)vinylene/-1-methyl-
$N_2C_{21}H_{25} \cdot Cl$	3H-Pseudoindolium chloride, 2-(p-dimethylaminostyryl)-1,3,3-trimethyl-
$N_2C_{22}H_{20}$	p-Toluidine, N,N-(p-phenylenedimethylidyne)-di-
$N_2C_{22}H_{23} \cdot I$	1H-Cyclopenta/b/quinolinium iodide, 3-(p-dimethylaminobenzylidene)-2,3-dihydro-4-methyl-
$N_2C_{25}H_{30}$	Quinoline, 6,6'-methylenebis/1,2-dihydro-2,2,4-trimethyl
$N_2ClC_{22}H_{24} \cdot Cl$	Ammonium chloride, /4-α-(o-chlorophenyl)-p-dimethylaminobenzylidene-2,5-cyclohexadineylidene dimethyl
$N_2ClC_{25}H_{27} \cdot HCl$	2,4-Xylidene, α^4-(o-chlorophenyl)-N-ethyl-α^4-/4-(ethylimino)-3-methyl-2,5-cyclohexadienylidene/-, hydrochloride
$N_2Cl_2C_{20}H_{14}$	Aniline, N,N-(p-phenylenedimethylidynebis/4-chloro-
$N_2OC_9H_{12}$	m-Toluidine, N,N-dimethyl-4-nitroso
$N_2OC_{11}H_{14}$	2H-1,5-Benzodiazepin-2-one, 1,3,4,5-tetrahydro-7,8-dimethyl-
$N_2OC_{14}H_8$	Anthra/1,9-cd/pyrazol-6(1H)-one
$N_2OC_{14}H_9 \cdot K$	Potassium salt of phenylcyanomethylenequinoneoxime
$N_2OC_{15}H_{10}$	Anthra/1,9-cd/pyrazol-6(2H)-one, 2-methyl-
$N_2OC_{15}H_{15} \cdot Cl$	Xanthylium chloride, 3,6-bis(methylamino)-
$N_2OC_{16}H_{17} \cdot I$	Morpholinium iodide, 1-benz/cd/indol-2-yl-1-methyl-
$N_2OC_{20}H_{14}$	1-Naphthol, 4-(1-naphthylazo)-
$N_2OC_{21}H_{23} \cdot I$	Quinolinium iodide, 2-(p-dimethylaminostyryl)-6-methoxy-1-methyl-
$N_2OC_{27}H_{20}$	4-Imidazolin-2-one, 1,3,4,5-tetraphenyl-
$N_2OClC_{14}H_{22} \cdot Cl$	Ammonium chloride, (p-chlorophenylcarbamoylmethyl)triethyl-
$N_2OClC_{17}H_{20} \cdot Cl$	Ammonium chloride, benzyl(p-chlorophenylcarbamoylmethyl)-dimethyl-
$N_2O_2C_8H_6$	Isatin, 3-oxime

Formula	Name
$N_2O_2C_8H_{10} \cdot HCl$	o-Benzoquinone, 4-dimethylamino-, 1-oxime
$N_2O_2C_{13}H_{21} \cdot Cl$	Ammonium chloride, triethyl(p-nitrobenzyl)-
$N_2O_2C_{14}H_{14}$	Hydantoin, 5-methyl-5-styryl-3-vinyl
$N_2O_2C_{16}H_{12}$	2-Naphthol, 1-(o-hydroxyphenylazo)-
$N_2O_2C_{16}H_{19} \cdot Cl$	Ammonium chloride, benzyl(p-nitrobenzyl)dimethyl-
$N_2O_2C_{17}H_{14}Ni \cdot NC_5H_5$	Benzoic acid, γ-hydroxy-α-methylcinnamylidenehydrazone, nickel[II] derivative, nickel[II] salt, compound with pyridine
$N_2O_2C_{18}H_{15} \cdot Cl$	Benzo[a]phenazoxonium chloride, 9-dimethylamino-2-hydroxy-
$N_2O_2C_{20}H_{12}$	Indeno[1,2-b]fluorene-6,12-dione, 2,8-diamino-
$N_2O_2C_{21}H_{14}$	Acridine, 9-(p-nitrostyryl)
$N_2O_2C_{22}H_{20}$	p-Anisidine, N,N'-(p-phenylenedimethylidyne)-di-
$N_2O_2Cl_2C_{21}H_{12}$	2-Anthraquinonecarboximidoyl chloride, 1-amino-N-(m-chlorophenyl)-
$N_2O_3BrC_{21}H_{15}$	Anthraquinone, 1-amino-2-bromo-4-(p-anisidino)-
$N_2O_3Br_2C_{21}H_{16}$	Benzamide, N,N'-(3,5-dibromosalicylidene)bis
$N_2O_3C_{12}H_{10}$	1,2-Naphthoquinone, 5-acetamido-, 1-oxime
$N_2O_3C_{13}H_8$	Benzonitrile, 2-[2-(2-furyl)vinyl]-5-nitro-
$N_2O_3C_{18}H_{21} \cdot 1/2 C_2$	4-Biphenyldiazonium oxalate-2,4',5-triethoxy
$N_2O_3C_{21}H_{14}$	Anthraquinone, 1-(p-aminobenzamido)
$N_2O_3C_{27}H_{26}$	Benz[g]indolo[2,3-a]quinolizin-2(1H)-one, 3,4,4a,5,7,8,13,13b,14,14a-decahydro-3-(hydroxymethylene)-, benzoate
$N_2O_3C_{28}H_{31} \cdot Cl$	Xanthylium chloride, 9-(o-carboxyphenyl-3,6-bis(ethylamino)-2,7-dimethyl ethyl ester
$N_2O_3ClC_{21}H_{11}$	Naphth[2,3-c]acridine-5,14-dione, 6-amino-10-chloro-8-hydroxy-
$N_2O_3ClC_{21}H_{13}$	Anthraquinone, 4-amino-1-benzamido-6-chloro-
$N_2O_4C_{13}H_{10}$	Anthranilic acid, 6-nitro-N-phenyl
$N_2O_4C_{14}H_{10}$	Chrysazin, 4,5-diamino
$N_2O_4C_{16}H_{14}$	Anthrarufin, 4,8-bis(methylamino)-
$N_2O_4C_{16}H_{14}$	Chrysazin, 4,5-bis(methylamino)
$N_2O_4C_{20}H_{14}$	Anthrarufin, 4-amino-8-anilino
$N_2O_4C_{20}H_{22}$	Benzamide, N-(3-indolylmethyl)-3,4,5-trimethoxy-N-methyl-
$N_2O_4C_{20}H_{31} \cdot I$	Pyrrolidinium iodide, 1-methyl-1-[1-(3,4,5-trimethoxybenzoyl)-4-piperidyl]-
$N_2O_4C_{21}H_{33} \cdot I$	Piperidinium, 1-methyl-1-[1-(3,4,5-trimethoxybenzoyl)-4-piperidyl]-
$N_2O_4C_{22}H_{16}$	Benzoic acid, p-(4-methylamino-1-anthraquinonylamino)-
$N_2O_4C_{22}H_{16}$	Acetanilide, 4'-(4-hydroxy-1-anthraquinonylamino)-
$N_2O_4C_{22}H_{16}$	Anthraquinone, 1-amino-4-(o-methoxybenzamido)-
$N_2O_4C_{22}H_{22}$	2-Naphthoic acid, 3-hydroxy-4-(6-methoxy-m-tolylazo) isopropyl ester
$N_2O_4C_{23}H_{18}$	Acetanilide, 4'-(4-hydroxy-1-anthraquinonylamino)-N-methyl-

Formula	Name
$N_2O_4C_{23}H_{20}$	Anthrarufin, 4-(N-ethylanilino)-8-(methylamino)-
$N_2O_4C_{30}H_{26}$	Anthraquinone, 1,4-bis/p-(2-hydroxyethyl)anilino/-
$N_2O_4C_{40}H_{22}$	Cinnamamide, N-(5,10,15,16-tetrahydro-5,10,15-trioxoanthra-/2,1,9-mna/naphth/2,3-/acridin-9-yl)-
$N_2O_4ClC_{20}H_{13}$	Anthrarufin, 4-amino-8-(m-chloroanilino)
$N_2O_4ClC_{28}H_{17}$	Anthraquinone, 1-benzamido-4-p-chlorobenzamido
$N_2O_4ClC_{28}H_{17}$	Anthraquinone, 1-benzamido-4-m-chlorobenzamido
$N_2O_4ClC_{38}H_{19}$	Anthra/2,1,9-mna/naphth/2,3-/acridine-5,10,15(16H)-trione, 9-(p-chloro-benzamido)-
$N_2O_4Cl_3C_{20}H_{11}$	Anthrarufin, 4-amino-8-(2,4,6-trichloroanilino)-
$N_2O_4IC_6H_3$	Benzene, 1-iodo-3,5-dinitro-
$N_2O_5C_{13}H_6$	9-Fluorenone, 2,6-dinitro-
$N_2O_5C_{14}H_{20}$	Oxanilic acid, 4'-amino-2',5'-diethoxy-, ethyl ester
$N_2O_5C_{21}H_{12}$	Anthraquinone, 1-(p-nitrobenzamido)-
$N_2O_5C_{28}H_{18}$	Anthraquinone, 1-benzamido-4-salicylamido-
$N_2O_5C_{28}H_{18}$	Benzanilide, 4'-(1-anthraquinonylcarbamoyl)-3'-hydroxy-
$N_2O_5C_{29}H_{20}$	Anthraquinone, 1-m-anisamido-4-benzamido-
$N_2O_5C_{32}H_{20}$	2-Naphthamide, N-(4-benzamido-1-anthraquinonyl)-1-hydroxy-
$N_2O_5C_{35}H_{20}$	Anthraquinone, 5-benzamido-1,1'-iminodi-
$N_2O_5ClC_{21}H_{15}$	Anthrarufin, 4-amino-8-(5-chloro-o-anisidino)
$N_2O_5ClC_{28}H_{17}$	Salicylamide, N-(4-benzamido-1-anthraquinonyl)-3-chloro-
$N_2O_5Cl_2C_{28}H_{16}$	Anthraquinone, 1-benzamido-4-(3,5-dichlorosalicylamido)-
$N_2O_5Cl_2C_{31}H_{14}$	Naphtho/2,3-b/pyrrocoline-12-carboxamide, N-(1,3-dichloro-2-anthraquinonyl)-6,11-dihydro-6,11-dioxo
$N_2O_6C_{20}H_{12}$	Chrysazin, 4-anilino-5-nitro-
$N_2O_6C_{20}H_{12}$	Anthrarufin, 4-anilino-8-nitro-
$N_2O_6C_{21}H_{14}$	Chrysazin, 4-(N-methylanilino)-5-nitro-
$N_2O_6C_{21}H_{14}$	Anthrarufin, 4-(N-methylanilino)-8-nitro-
$N_2O_6C_{20}H_{18}$	Anthraquinone, 1-cyclohexylamino-4,8-dihydroxy-5-nitro
$N_2O_6C_{21}H_{14}$	Anthrarufin, 8-nitro-4-(p-toluidino)-
$N_2O_6C_{22}H_{16}$	Chrysazin, 4-(N-ethylanilino)-8-nitro
$N_2O_6C_{22}H_{16}$	Anthrarufin, 4-(p-ethylanilino)-8-nitro-
$N_2O_6C_{22}H_{16}$	Anthrarufin, 4-nitro-8-(3,4-xylidino)
$N_2O_6C_{22}H_{16}$	Anthrarufin, 4-nitro-8-xylidino
$N_2O_6C_{22}H_{16}$	Anthrarufin, 4-(N-ethylanilino)-8-nitro-
$N_2O_6C_{23}H_{18}$	Benzoic Acid, p-(5-amino-4,8-dihydroxy-1-anthraquinonylamino ethyl ester

Formula	Name
$N_2O_6C_{24}H_{18}$	Benzo/I,2-b·5,4-b/diindolizine-12,14-dicarboxylic acid, 6,13-dihydro-6,13-dioxo-, diethyl ester
$N_2O_6C_{24}H_{20}$	Benzoic acid, p-(4,8-dihydroxy-5-methylamino-1-anthraquinone
$N_2O_6C_{25}H_{22}$	Anthrarufin, 4-(N-butyl-m-toluidino)-8-nitro-
$N_2O_6C_{28}H_{18}$	Anthraquinone, 1,4-disalicylamido-
$N_2O_6C_{32}H_{18}$	Fumaramide, N,N'-di-1-anthraquinonyl
$N_2O_6C_{36}H_{20}$	Terephthalamide, N,N'-di-2-anthraquinonyl-
$N_2O_6ClC_{20}H_{11}$	Anthrarufin, 4-(p-chloroanilino)-8-nitro-
$N_2O_6ClC_{20}H_{11}$	Anthrarufin, 4-(m-chloroanilino)-8-nitro-
$N_2O_6Cl_2C_{20}H_{10}$	Anthrarufin, 4-(2,5-dichloroanilino)-8-nitro
$N_2O_6Cl_2C_{28}H_{16}$	Anthraquinone, 1,4-bis(3-chlorosalicylamido)-
$N_2O_6Cl_2C_{28}H_{16}$	Anthraquinone, 1,5-bis(3-chlorosalicylamido)-
$N_2O_6Cl_4C_{36}H_{16}$	Phthalamide, N,N'-di-1-anthraquinonyl-3,4,5,6-tetrachloro
$N_2O_6F_3C_{21}H_{11}$	Chrysazin, 4-nitro-5(α,α,α-trifluoro-m-toluidino
$N_2O_7C_{23}H_{18}$	Benzoic acid, m-(5-amino-4,8-dihydroxy-1-anthraquinonylamino)-, 2-hydroxyethyl ester
$N_2O_7C_{25}H_{22}$	Anthrarufin, 4-/N-(4-methoxybutyl)-anilino/-8-nitro
$N_2O_7ClC_{21}H_{13}$	Anthrarufin, 4-(5-chloro-o-anisidino)-8-nitro-
$N_2O_8C_{21}H_{12}$	Benzoic acid, m-(4,5-dihydroxy-8-nitro-1-anthraquinonylamino)-
$N_2O_8C_{21}H_{12}$	Benzoic acid, m-(4,8-dihydroxy-5-nitro-1-anthraquinonylamino)-
$N_2O_8C_{22}H_{14}$	Benzoic acid, p-(4,8-dihydroxy-5-nitro-1-anthraquinonylamino-, methyl ester
$N_2O_8C_{22}H_{14}$	Benzoic acid, p-(4,5-dihydroxy-8-nitro-1-anthraquinonylamino)-, methyl ester
$N_2O_8C_{23}H_{16}$	Benzoic acid, p-(4,8-dihydroxy-5-nitro-1-anthraquinonylamino)-, ethyl ester
$N_2O_8C_{36}H_{20}$	Phthalamide, N,N'-di-1-anthraquinonyl-3,6-dihydroxy-
$N_2O_8C_{43}H_{26}$	Salicylamide, 5,5'-methylenebis/N-1-anthraquinonyl
$N_2O_{10}C_{24}H_{16}$	Phthalic acid, 4-(4,8-dihydroxy-5-nitro-1-anthraquinonylamino)-, dimethyl ester
$N_2SC_{12}H_9Ni_{1/2}$	Picolinamidic acid, N-phenylthio-, nickel[II] salt
$N_2SC_{18}H_{19}·I$	Benzothiozolium iodide, 2-(p-dimethylaminostyryl)-3-methyl
$N_2SO_2Cl_2C_{12}H_8$	3,7-Dibenzothiophenediamine, 2,8-dichloro-, 5,5-dioxide
$N_2SO_4C_8H_{10}$	Methanesulfonic acid, (N-methyl-p-nitrosoaniline)
$N_2SO_5C_{16}H_{11}Na$	1-Phenol-4-sulfonic acid, 2-(2-hydroxy-1-naphthylazo)-sodium salt
$N_2SO_5C_{12}H_{14}$	2-Naphthol-4-sulfonic acid, 1-(6-hydroxy-m-tolylazo)
$N_2SO_5C_{33}H_{22}$	2-Anthraquinonesulfonamide, 5-(7-oxo-7H-benz/de/anthracen-3-ylamino)-N,N-dimethyl-
$N_2SO_6C_{18}H_{13}Na$	5H-Benzo/a/phenoxazine-ar[1]-sulfonic acid, 9-dimethylamino-6,12-dihydro-5,6-dioxo-, sodium salt
$N_2SO_6C_{21}H_{16}$	Chrysazin, 4-amino-8-(m-methylsulfonylanilino)-

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Formula	Name
$N_2SO_6C_{23}H_{18}$	Salicylamide, N-1-anthraquinonyl-5-(dimethylsulfamoyl)
$N_2SO_6C_{44}H_{24}$	Anthra/2,1,9-mna7naphth/2,3-_7acridine-5,10,15(16H)-trione, 9-/(p-phenylsulfonyl)benzamido7-
$N_2SO_6ClC_{21}H_{15}$	Anthrarufin, 4-amino-8-(p-chloromethylsulfonylanilino)-
$N_2SO_6Cl_2C_{20}H_{14}$	Acetoacetamide, N-(7-acetoacetamido-2,8-dichloro-3-dibenzothiophenyl)-, S,S-dioxide
$N_2SO_8C_{21}H_{14}$	Anthrarufin, 4-(m-methylsulfonylanilino)-8-nitro
$N_2SO_8C_{21}H_{14}$	Chrysazin, 4-(m-methylsulfonylanilino)-5-nitro-
$N_2SO_8ClC_{21}H_{13}$	Anthrarufin, 4-(p-chloromethylsulfonylanilino)-8-nitro-
$N_2S_2C_{19}H_{17} \cdot ClO_4$	1,2-Dithioliumperchlorate, 3-{p-/(2-cyanoethyl)methylamino7phenyl}-4-phenyl
$N_2S_2C_{19}H_{12} \cdot ClO_4$	1,2-Dithioliumperchlorate, 3-{p-/(2-cyanoethyl)methylamino7phenyl}-5-phenyl
$N_2S_2Cl_2C_{12}H_{10}$	Aniline, 2,2'-dithiobis/4-chloro-
$N_2S_2O_4ClC_{39}H_{30}Na$	Ammonium hydroxide, {4-{α-(o-chlorophenyl)-4-/ethyl-(m-sulfobenzyl)amino7-2-methylbenzylidene}-3-methyl-2,5-cyclohexadienylidene} ethyl(m-sulfophenyl)-, inner salt, sodium salt
$N_2S_2O_6C_{27}H_{31}Na$	Ammonium hydroxide, diethyl{4-/α-(p-dimethylaminophenyl)-2,4-disulfobenzylidene7-2,5-cyclohexadienylidene}-, 2-(inner salt), 4-(sodium salt)
$N_2S_2O_7C_{20}H_{12}Na_2$	1-Naphthol-4-sulfonic acid, 2-(4-sulfo-1-naphthylazo disodium salt
$N_2S_2O_7C_{37}H_{35}Co_{1/2}$	Patent Blue A
$N_2S_2O_8C_{20}H_{12}Na_2$	2-Anthraquinonesulfonic acid, 1-amino-4-p-sulfoanilino-, disodium salt
$N_2S_2O_8C_{22}H_{17}Na$	2-Anthraquinonesulfonic acid, 1-amino-4-/m-(2-hydroxyethylsulfonyl)anilino7-, sodium salt
$N_2S_3O_8C_{12}H_9Na$	2,8-Dibenzothiophenedisulfonic acid, 3,7-diamino-, 5,5-dioxide, monosodium salt
$N_2S_3O_8C_{12}H_{10} \cdot NC_5H_5$	2,8-Dibenzothiophenedisulfonic acid, 3,7-diamino-5,5-dioxide, monosalt with pyridine
$N_2S_3O_9C_{37}H_{34}Na_2$	Ammonium hydroxide, ethyl{4-{p-/ethyl(ar-sulfobenzyl)-amino-α-p-sulfophenylbenzylidene}-2,5-cyclohexadienylidene}-ar-sulfobenzyl-, ar-(inner salt), ar', p-(disodium salt)
$N_2S_4C_8H_{14}Zn$	Zinc, N,N'-diethylethylenebis(dithiocarbamate)
$(N_3C_4H)_x$	Cyanaform polymer
$N_3C_4H_{13} \cdot 1/2CuCl_2$	Diethylenetriamine, compound with $CuCl_2$
$N_3C_{10}H_{14} \cdot Cl \cdot 1/2ZnCl_2$	Benzenediazonium chloride, p-diethylamino-, compound with $ZnCl_2$
$N_3C_{11}H_{14} \cdot ClO_4$	Pyrazolium perchlorate, 4-(p-aminophenyl)-1,2-dimethyl-
$N_3C_{16}H_{21} \cdot 1/2ZnCl_2$	Bindschedler's Green
$N_3C_{19}H_{20} \cdot I$	Quinolinium iodide, 2-/(p-dimethylaminophenylimino)-methyl7-1-methyl-
$N_3C_{22}H_{26} \cdot I$	Quinolinium iodide, 6-dimethylamino-2-(p-dimethylaminostyryl)-1-methyl-
$N_3C_{24}H_{20} \cdot Cl$	Benzo/a7phenazinium chloride, 9-dimethylamino-7-phenyl-
$N_2C_{25}H_{25} \cdot Cl$	Quinolinium chloride, 1-ethyl-2-/3-(1-ethyl-2(1H)-quinolylidene)propylenyl7-

Formula	Name
$N_3C_{29}H_{32} \cdot Cl$	Ammonium chloride, {4-[p-dimethylamino-α-(4-ethylamino-1-naphthyl)-benzylidene]-2,5-cyclohexadienylidene} dimethyl
$N_3C_{31}H_{42} \cdot Cl$	Ammonium chloride, {4-[bis(p-diethylaminophenyl)methylene]-2,5-cyclo-hexadienylidene} diethyl-
$N_3OC_{17}H_{15}$	2-Naphthylamine, 1-(p-methoxyphenylazo)-
$N_5O_2BrC_{22}H_{16}$	Anthraquinone, 2-bromo-3-(p-dimethylaminophenylazo)-
$N_3C_2C_{11}H_{14}Cl \cdot 1/2ZnCl_2$	Benzenediazonium chloride, 2-methoxy-4-morpholino-, compound with $ZnCl_2$
$N_3O_2C_{13}H_9$	Imidazo-[1,5-a]-pyridine, 3-(p-nitrophenyl)-
$N_3O_2C_{17}H_{13}$	Benzoic acid, p-(2-amino-1-naphthylazo)-
$N_3O_2C_{18}H_9$	Acenaphtho-[1,2-b]-quinoxaline, 9-nitro-
$N_3O_2C_{19}H_{17}$	Benzoic acid, p-(2-amino-1-naphthylazo)-, ethyl ester
$N_3O_2ClC_{16}H_{10}$	1-(2-Chloro-4-nitrophenylazo)-azulene
$N_3O_3C_{22}H_{17}$	Acetanilide, p-(4-amino-1-anthraquinonylamino)-
$N_3O_3C_{23}H_{19}$	Acetanilide, 4'-(4-amino-1-anthraquinonylamino)-N-methyl-
$N_3O_3C_{28}H_{17}$	Benzamide, N-1-anthraquinonyl-o-(2-benzimidazolyl)-Anthraquinone-1-[o-(2-benzimidazolyl)benzamido]-
$N_3O_4BrC_{21}H_{16}$	m-Toluenesulfonamide, 6-(4-amino-3-bromo-1-anthraquinonylamino)-
$N_3O_4C_{17}H_{18} \cdot Cl$	Phenazoxonium chloride, 1-carbamoyl-7-diethylamino-3,4-dihydroxy-
$N_3O_4C_{23}H_{19}$	Acetanilide, 4'-(4-amino-3-methoxy-1-anthraquinonylamino
$N_3O_4C_{27}H_{25}$	Benzanilide, 2',5'-diethoxy-4'-(2-hydroxy-1-naphthylazo)-
$N_3O_4Cl_2C_{14}H_7$	Anthraquinone, 1,4-diamino-2,3-dichloro-5-nitro-
$N_3O_6C_{28}H_{17}$	Anthraquinone, 1-benzamido-4-(p-nitrobenzamido)-
$N_3O_6C_{35}H_{19}$	Benzamide, N-(1-anthraquinonyl)-p-(5,8-dihydro-2-hydroxy-8-dioxo-1-naphthylazo)-
$N_3O_8C_{21}H_{11}$	Salicylamide, N-1-anthraquinonyl-3,5-dinitro-Anthraquinone, 1-(3,5 dinitrosalicylamido)-
$N_3O_8ClC_{36}H_{16}$	2-Anthraquinone carboxamide, N-(10-chloro-5,14-dihydro-8-hydroxy-5,14-dioxo-naphth[2,3-c]acridin-6-yl)-1-nitro
$N_3O_9C_{21}H_{13}$	Anthrarufin, 4-nitro-8-(5-nitro-o-anisidino)-
$N_3O_9C_{51}H_{27}$	Trimellitamide, N,N',N''-tri-1-anthraquinonyl-
$N_3SC_{14}H_{14} \cdot Cl$	Azure A
$N_3SC_{15}H_{16} \cdot Br$	Phenazathionium bromide, 3-dimethylamino-7-methylamino-
$N_3SO_4C_{21}H_{25}$	2-Anthraquinonesulfonamide, 5-(ethylamino)-N-(3-dimethylaminopropyl)-
$N_3SO_5C_6H_7$	1-Phenol-2-sulfonamide, 6-amino-4-nitro-
$N_3SO_5C_{12}H_7$	Phenothiazine, 2,8-dinitro-, 5-oxide
$N_3SO_5C_{20}H_{14}Na$	2-Anthraquinonesulfonic acid, 1-amino-4-(p-aminoanilino)-, sodium salt
$N_3SO_5C_{20}H_{15}$	2-Anthraquinonesulfonic acid, 1-amino-4-(p-aminoanilino)-

Formula	Name
$N_3SO_6C_{22}H_{15}$	Acetonitrile, /N-(5-amino-4,8-dihydroxy-1-anthraquinonyl)sulfanilyl
$N_3SO_7C_{20}H_{12}Na$	2-Naphthol-4-sulfonic acid, 1-(2-hydroxy-1-naphthylazo)-6-nitro-, sodium salt
$N_3SO_7C_{20}H_{12}Na$	Eriochrome Black T
$N_3S_2C_{15}H_{12} \cdot Cl$	1,2-Dithiolium chloride, 5-(p-aminophenylazo)-3-phenyl-
$N_3S_2C_{16}H_{14} \cdot I$	Benzothiazolium iodide, 3-methyl-2-(3-methyl-2-benzothiazolinylidene-amino)-
$N_3S_2OC_{15}H_{13}$	Ferric complex of 4,5-benzo-1,2-dithiol-3-one, p-acetamidophenylhydrazone
$N_3S_2O_6C_{37}H_{28}Na$	Benzenesulfonic acid, α-(4-phenylimino-2,5-cyclohexadienylidene)-α-/p-(ar-sulfoanilino)phenyl7-p-toluidino-, sodium salt
$N_3S_2O_6C_{37}H_{36}Na$	Ammonium hydroxide, dimethyl-{4-{α-(p-dimethylaminophenyl)-m-/bis(m-sulfobenzyl)amino7benzylidene}-2,5-cyclohexadienylidene}-, inner salt, sodium salt
$N_3S_2O_6C_{41}H_{44}Na$	Ammonium hydroxide, ethyl {4-{α-(p-diethylaminophenyl)-p-/ethyl(ar-sulfobenzyl)amino7benzylidene}-2,5-cyclohexadienylidene}-(ar-sulfobenzyl)-, inner salt, sodium salt
$N_3S_2O_8C_{20}H_{13}Na_2$	2-Anthraquinonesulfonic acid, 1-amino-4-(4-amino-2-sulfoanilino)-, disodium salt
$N_3S_2O_8C_{22}H_{16}Na$	2-Anthraquinonesulfonic acid, 4-(N⁴-acetylsulfanilamido)-1-amino-, sodium salt
$N_3S_2O_9C_{23}H_{15}Na_2$	2-Anthraquinonesulfonic acid, 1-amino-4-(4-acrylamido-2-sulfanilino)-, disodium salt
$N_3S_2O_9ClC_{22}H_{14}Na_2$	2-Anthraquinonesulfonic acid, 1-amino-4-/4-(2-chloroacetamido)-s-sulfo-anilino7-, disodium salt
$N_3S_3C_3Cu_{3/2}$	s-Triazine-2,4,6-trithiol, tricopper^{II} derivative
$N_3S_3C_3Fe$	s-Triazine-2,4,6-trithiol, monoiron^{III} derivative
$N_3S_3C_3Zn_{3/2}$	s-Triazine-2,4,6-trithiol, trizinc derivative
$N_3S_3O_7C_{24}H_{14}Na_2$	1-Naphthol-4-sulfonic acid, 2-/p-(6-methyl-7-sulfo-2-benzothiazolyl)phenylazo7, disodium salt
$N_4BrC_{11}H_{11}$	s-Tetrazine, 3-bromo-6-p-cumenyl-
$N_4C_4H_3Co_{1/2}$	Acetamidine, N,N'-dicyano-, copper^{II} derivative
$N_4C_{14}H_{17} \cdot SC_4CH_3$	Pyridinium methyl sulfate, 2-(p-dimethylaminophenylazo)-1-methyl-
$N_4C_{15}H_{17} \cdot I$	Ammonium iodide, trimethyl(2-phenyl-2H-benzotriazol-5-yl)-
$N_4C_{15}H_{19} \cdot I$	3-Picolinium iodide, 6-(p-dimethylaminophenylazo)-1-methyl-
$N_4C_{15}H_{19} \cdot I$	4-Picolinium iodide, 2-(p-dimethylaminophenylazo)-1-methyl-
$N_4C_{16}H_{12}$	2,2'-Stilbenedicarbonitrile, 4,4'-diamino-
$N_4C_{16}H_{21} \cdot I$	Pyridinium iodide, 4-(4-amino-3,5-diethylphenylazo)-1-methyl-
$N_4C_{16}H_{21} \cdot I$	3-Picolinium iodide, 6-(4-dimethylamino-o-tolylazo)-1-methyl-
$N_4C_{18}H_{19} \cdot Cl$	Quinolinium chloride, 5-(p-dimethylaminophenylazo)-1-methyl
$N_4C_{18}H_{19} \cdot SO_4CH_3$	Quinolinium methyl sulfate, 5-(p-dimethylaminophenylazo)-1-methyl-
$N_4C_{18}H_{25} \cdot I$	Pyridinium iodide, 4-(p-dipropylaminophenylazo)-1-methyl-

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Formula	Name
$N_4C_{19}H_{16}$	N-,N'-Diphenyl-c-(4-pyridyl)-formazan
$N_4C_{19}H_{19} \cdot I$	Pyridinium iodide, 1-methyl-2-/p-(N-methylanilino)phenylazo/-
$N_4C_{20}H_{14}$	Imidazo-/1,5-a/-pyridine, 3,3'-(p-phenylene)bis-
$N_4C_{22}H_{20}$	Benzimidazole, 2,2'-vinylenebis/1-allyl-
$N_4ClC_9H_4K$	Benzamide, p-chloro-N,N'-dicyano-, potassium derivative
$N_4Cl_2C_5HHg$	Purine 2,6-dichloro-ar[7]-mercury derivative
$N_3OC_{16}H_{14} \cdot SO_4CH_3$	Pyridinium methyl sulfate, 3-(4-hydroxy-1-naphthylazo)-1-methyl-
$N_4OC_{16}H_{17} \cdot I$	1H-Imidazo/1,2-a/pyridinium iodide, 3-(p-methoxyphenylazo)-1,2-dimethyl-
$N_4OC_{18}H_{25} \cdot I$	Pyridinium iodide, 2-(4-diethylamino-2-ethoxyphenylazo)-1-methyl-
$N_4OClC_{10}H_9$	s-Tetrazine, 3-(p-chlorophenyl)-6-ethoxy-
$N_4OClC_{14}H_{12} \cdot I$	1H-Diimidazo/1,2-a/pyridiniumiodide, 3-(p-chlorophenylazo)-2-hydroxy-1-methyl-
$N_4OClC_{16}H_{16} \cdot I$	1H-Imidazo/1,2-a/pyridinium iodide, 3-(5-chloro-2-methylphenylazo)-1,2-dimethyl-
$N_4O_2C_{14}H_{17} \cdot I$	Pyridinium iodide, 4-(4-amino-2,5-dimethoxyphenylazo)-1-methyl-
$N_4O_2C_{14}H_{17} \cdot I$	Pyridinium iodide, 4-(4-amino-2,5-dimethoxyphenylazo)-1-methyl-
$N_4O_2C_{15}H_{15} \cdot I$	Imidazo/1,2-a/pyridinium iodide, 2-hydroxy-3-(p-methoxyphenylazo)-1-methyl-
$N_4O_2C_{18}H_{16}Ni$	Benzoic acid, (dimethylethanediylidene)dihydrazone, dinickel[II] salt
$N_4O_2C_{18}H_{18}$	Quinoline, 1,2-dihydro-2,2,4-trimethyl-6-(p-nitrophenylazo)-
$N_4O_2C_{19}H_{16}$	Carbanilic acid, p-2H-naphtho/1,2-d/triazol-2-yl-ethyl ester
$N_4O_2C_{28}H_{12}$	9H, 18H-Bisbenz/6,7/indozolo-/2,3,4-m,n,o,2',3',4'-f,g,h/phenazine-9,18-dione
$N_4O_2C_{28}H_{44} \cdot 2Cl$	Ammonium chloride, /p,p'-biphenylenebis(iminocarbonylmethylene)/bis-/triethyl-
$N_4O_2C_{38}H_{38} \cdot 2I$	Ammonium iodide, /1,4-anthraquinonylenebis(trimethylene)/bis/trimethyl-
$N_4O_3C_{10}H_8$	Xanthine, 9-furfuryl-
$N_4O_3C_{15}H_{13} \cdot I$	Imidazo/1,2-a/pyridinium iodide, 3-(o-carboxyphenylazo)-2-hydroxy-1-methyl-
$N_4O_3C_{16}H_{18}$	o-Anisidine, 5-ethyl-4-(4-nitro-o-tolylazo)-
$N_4O_4C_{24}H_{18}$	Oxalic acid, bis/2-(2-hydroxy-1-naphthylmethylene)-hydrazide/
$N_4O_4Cl_2C_{17}H_{16}$	1(2H)-Quinoline ethanol, 6-(2,6-dichloro-4-nitrophenylazo)-3,4-dihydro-3-hydroxy-
$N_4O_5C_{12}H_8$	Azoxybenzene, 3,3'-dinitro-
$N_4O_6C_{26}H_{16}$	Anthrarufin, 4-nitro-8-/p-(phenylazo)anilino/
$N_4O_6C_{36}H_{40} \cdot Br_2$	Ammonium bromide, /4,4'-biphenylenebis-(iminocarbonylmethylene/bis/(p-carboxybenzyl)dimethyl-
$N_4O_7C_{14}H_{16}$	m-Dioxane-2-acetaldehyde, 4,6-dimethyl-5-oxo-, 2,4-dinitrophenylhydrazone
$N_4O_8C_{50}H_{30}$	Phthalamide, N,N'-bis(4-benzamido-1-anthraquinonyl)-
$N_4O_{10}C_{52}H_{26}$	Pyromellitimide, N,N'-bis(4-benzamido-1-anthraquinonyl)-

Formula	Name
$N_4SC_{13}H_{11}Hg_{1/2}$	Dithizone, mercury[II] chelate
$N_4SC_{15}H_{14}$	Benzothiazole, 2-(p-dimethylaminophenylazo)-
$N_4SOC_{19}H_{20}$	Benzothiazole, 4,6-dimethyl-2-(p-morpholinophenylazo)-
$N_4SOC_{20}H_{23} \cdot SO_3C_7H_7$	Benzothiazolium p-toluenesulfonate, 3,4,6-trimethyl-2-(p-morpholino-phenylazo)-
$N_4SO_2C_{16}H_{14}$	Benzenesulfonamide, p-(2-amino-1-naphthylazo)-
$N_4SO_2C_{17}H_{19} \cdot SO_3C_7H_7$	Benzothiazolium p-toluenesulfonate, 6-hydroxy-2-{4-[(2-hydroxyethyl)-methylamino]phenylazo}-3-methyl-
$N_4SO_2C_{18}H_{18}$	Benzothiazole, 4-methoxy-2-(p-morpholinophenylazo)-
$N_4SO_2C_{18}H_{19} \cdot SO_3C_7H_7$	Benzothiazolium p-toluenesulfonate, 4-hydroxy-3-methyl-2-(p-morpholino-phenylazo)-
$N_4SO_2C_{18}H_{19} \cdot SO_3C_7H_7$	Benzothiazolium p-toluenesulfonate, 6-hydroxy-3-methyl-2-(p-morpholino-phenylazo)-
$N_4SO_2C_{19}H_{21} \cdot SO_3C_7H_7$	Benzothiazolium p-toluenesulfonate, 4-methoxy-3-methyl-2-(p-morpholino-phenylazo)-
$N_4SO_2C_{21}H_{18}$	Benzo[h]quinolin-3-ol, 1,2,3,4-tetrahydro-6-(6-methoxy-2-benzo-thiazolylazo)-
$N_4SC_2C_{22}H_{21} \cdot SO_3C_7H_7$	Benzothiazolium p-toluenesulfonate, 6-methoxy-3-methyl-2-(1,2,3,4-tetrahydro-3-hydroxybenzo[h]quinolin-6-ylazo)-
$N_4SO_3ClC_{19}H_{22} \cdot SO_3C_7H_7$	Benzothiazolium p-toluenesulfonate, 2-{4-[bis(2-hydroxyethyl)amino]-2-chlorophenylazo}-6-methoxy-3-methyl-
$N_4SO_3C_{19}H_{20}$	1(2H)-Quinolineethanol, 3,4-dihydro-3-hydroxy-6-(6-methoxy-2-benzothiazolyl-azo)-
$N_4SO_3ClC_{18}H_{19}$	Ethanol, 2,2'-[3-chloro-4-(6-methoxy-2-benzothiazolylazo)phenylimino]di-
$N_4SO_4C_{23}H_{15}Ca_{1/2}$	2-Naphthol-6-sulfonic acid, 1-[o-(2-benzimidazolyl)phenylazo]-, calcium salt
$N_4SO_4C_{23}H_{15}Na$	2-Naphthol-6-sulfonic acid, 1-[o-(2-benzimidazolyl)phenylazo]-, sodium salt
$N_4SO_4C_{28}H_{42} \cdot 2Cl$	Compound from 3,7-bis(chloroacetamido)dibenzothiophene-5,5-dioxide and tri-ethylamine
$N_4SO_5C_{18}H_{16}$	Acetamide, N-[4-hydroxy-8-(2-hydroxy-5-sulfamoylphenylazo)-1-naphthyl]-
$N_4SO_7C_{27}H_{16}Na$	Diamond Black F
$N_4SO_{10}C_{56}H_{32}$	3,7-Dibenzothiophenedicarboxamide, N,N'-bis(4-benzamido-1-anthraquinonyl)-, 5,5-dioxide
$N_4S_2O_4C_{20}H_{22}$	p-Phenylenediamine, N,N-dimethyl-ar,ar-disulfanilyl
$N_4S_2O_4Cl_2C_{22}H_{16}$	Oxalic acid, bis[2-(6-chloro-3-hydroxy-4-methyl-2-thianaphthenylmethylene)-hydrazide]
$N_4S_2O_6C_{50}H_{22}$	6H-Anthra[9,1-cd]isothiazole-3-carboxamide, N,N'-(6,12-dihydro-6,12-dioxoindeno[1,2-b]fluorene-2,8-diyl)bis-[6-oxo-
$N_4S_2O_8C_{28}H_{26} \cdot Cl_2$	Pyridinium chloride, 1,1'-{vinylenebis[(3-sulfo-p-phenylene)imino-carbonylmethylene]}bis-
$N_4S_2O_8C_{40}H_{34} \cdot 2Cl$	Compound from 4,4'-bis(chloroacetamido-2,2'-stilbenedisulfonic acid, di-phenyl ester, and pyridine
$N_4S_2O_{10}C_{34}H_{24}Na_2$	Benzazurine G

Formula	Name

$N_4S_3O_{10}C_{18}H_{14}Na_2$ — 1-Naphthol-3,6-disulfonic acid, 8-acetamido-2-(m-sulfamoylphenylazo)-, disodium salt

$N_5Cl_2C_{15}H_{11} \cdot HCl$ — s-Triazine, 1,4-bis(p-chlorophenyl)-1,2,3,6-tetrahydro-2,6-diamino, hydrochloride

$N_5OClC_{16}H_{14}$ — s-Triazine-4-(p-chlorophenyl)-1,2,3,6-tetrahydro-2,6-diimino-1-(p-methoxyphenyl)

$N_5OClC_{31}H_{22}$ — 2-Naphtho-o-toluidide, 4-/o-(2-benzimidazolyl)phenylazo7-5'-chloro-3-hydroxy

$N_5O_2C_{15}H_{14} \cdot I$ — Imidazo/1,2-a7pyridinium iodide, 3-(p-nitrophenylazo)-1,2-dimethyl-

$N_5O_2C_{18}H_{13}$ — Perimidine, 2-methyl-6-(p-nitrophenylazo)

$N_5O_2C_{22}H_{13}Cu$ — 2,4-Quinolinediol, 3-/o-(2-benzimidazolyl)phenylazo7-copper^{II}

$N_5O_2C_{23}H_{17}Cu$ — Acetoacetanilide, 2-/o-(2-benzimidazolyl)phenylazo7-copper^{II} derivative

$N_5O_2C_{30}H_{21}$ — 2-Naphthanilide, 4-/o-(2-benzimidazolyl)phenylazo7-3-hydroxy-

$N_5O_2ClC_{27}H_{20}$ — 2H-Cyclopenta/d7pyridazine, ar^5-(2-chloro-4-nitrophenylazo)-1,4-di-p-tolyl-

$N_5SO_4C_{19}H_{13}$ — N-(2-hydroxyphenyl)-N'-(2-hydroxy-5-sulfamylphenyl)-c-phenylformazan

$N_5SO_4C_{22}H_{24}Na$ — 2-Naphthalenesulfonic acid, 6-{4-/ethyl-(2-ureidoethyl)amino7-o-tolylazo}-, sodium salt

$N_5SO_8C_{19}H_{11}Na_2$ — Mordant Brown 18

$N_5S_2O_8C_{28}H_{19}Na_2$ — 1-Naphthol-3-sulfonic acid, 6-amino-2-/4'-(4-hydroxy-2-sulfophenylazo)-4-bisphenylazo7-disodium salt

$N_5S_2O_9C_{35}H_{27}Na_2$ — 2,2'-Stilbenedisulfonic acid, 4-(p-aminobenzamido)-4'-p-(p-aminobenzamido)-benzamido-, disodium salt

$N_5S_3O_9C_{33}H_{27}Na_2$ — Phenazinium hydroxide, 3-(4-amino-3-sulfoanilino)-7-/ethyl(m-sulfobenzyl)-amino7-5-p-sulfophenyl-, 3,7-(disodium salt)

$N_5S_4O_{12}ClC_{18}H_{14}Na_2$ — 1-Naphthol-3,6-disulfonic acid, 8-acetamido-2-(5-chloro-2,4-disulfamoyl-phenylazo)-, disodium salt

$N_6C_{11}H_{13} \cdot SO_4CH_3$ — Pyridinium methyl sulfate, 3-(2,6-diamino-3-pyridylazo)-1-methyl-

$N_6C_{20}H_{21} \cdot SO_4CH_3$ — Pyridinium methyl sulfate, 2-/p-(p-dimethylaminophenylazo)-phenylazo7-1-methyl-

$N_6C_{28}H_{18}$ — 5,26: 13,18-Diimino-7,11: 20,24-dimethenodibenzo-/c,n7-/1,6,12,177tetra-azacyclododecosine

$N_6OC_{23}H_{18}$ — 5-Pyrazolol, 4-/o-(2-benzimidazolyl)-phenylazo7-3-methyl

$N_6O_2C_7H_{14}$ — s-Triazine, 2,4-bis(hydroxyamino)-6-diethylamino

$N_6O_2C_{11}H_{12}$ — 4-Pyrimidinol, 2,6-diamino-5-(p-methoxyphenylazo)-

$N_6O_2C_{16}H_{12}Ni_2 \cdot 2NC_5H_5$ — Oxalimidic acid, bis(salicylidene hydrazide)tetranickel^{II} derivative, compound with pyridine

$N_6O_4Cl_2C_{34}H_{26}$ — Acetonitrile, (3,3'-dichloro-p-biphenylylenebisazo)-bis/o-ethoxybenzoyl-

$N_6O_6C_{45}H_{24}$ — s-Triazine, 2,4,6-tris(1-amino-2-anthraquinonyl)

$N_6O_{12}C_{72}H_{42}$ — Trimellitamide, N,N',N"-tris(4-benzamido-1-anthraquinonyl)-

$N_6SO_3ClC_{24}H_{25}$ — Propionitrile, 3,3'- /3-chloro-4-(6-methoxy-2-benzothiazolylazo)phenyl-imino7bis(ethyleneoxy) di-

$N_6SO_3ClC_{25}H_{28} \cdot SO_3C_7H_7$ — Benzothiazolium p-toluenesulfonate, 2-{{4-bis/2-(2-cyanoethoxy)ethyl7amino}-2-chlorophenylazo} -6-methoxy-3-methyl-

Formula	Name

$N_6SO_6Cl_2C_{30}H_{14}C_{42}$ — 2,4-Quinolinediol, 3-[2,8-dichloro-7-(2,4-dihydroxy-3-quinolylazo)-dibenzothiophene-3-yl]azo-S,S-dioxide, copper derivative

$N_6SO_6Cl_2C_{32}H_{24}$ — Acetoacetanilide, 2-{2,8-dichloro-7-[1-(phenylcarbamoyl)acetonylazo]-dibenzothiophene-3-ylazo}-S,S-dioxide

$N_6SO_6Cl_2C_{34}H_{28}$ — o-Acetoacetotoluidide, 2-{2,8-dichloro-7-[1-(o-tolylcarbamoyl)-acetonylazo]dibenzothiophene-3-ylazo}-, S,S-dioxide

$N_6S_2O_9Cl_2C_{30}H_{16}Na_2$ — 2-Anthraquinonesulfonic acid, 1-amino-4-{4-[m-(4,6-dichloro-s-triazin-2-yl)-benzamido]-2-sulfoanilino}-, disodium salt

$N_6S_2O_{10}C_{34}H_{26}Na_2$ — 1-Naphthol-5-sulfonic acid, 2,2'-(3,3'-dimethoxy-4,4'-biphenylylene-bisazo)bis[8-amino-, disodium salt

$N_6S_4O_{16}C_{34}H_{24}Na_4$ — 1-Naphthol-3,6-disulfonic acid, 2,2'-(3,3'-dimethoxy-4,4'-biphenylylene-bisazo)bis[8-amino-, disodium salt

$N_7Cl_3C_{15}H_{12}$ — Pyridinium chloride, 2-{p-[(4,6-dichloro-s-triazin-2-yl)amino]phenyl-azo}-1-methyl-

$N_7O_2ClC_{18}H_{16}$ — Propionitrile, 3,3'-[4-(2-chloro-4-nitrophenylazo)-m-phenylenediimino]di-

$N_7S_2O_{14}C_{32}H_{17}Cu_2$ — 1-Naphthol-3-sulfonic acid, 6,6'-iminobis[2-(2-hydroxy-4-nitrophenylazo)-, tetracopper^{II} derivative

$N_7S_3O_9ClC_{26}H_{17}Na_3$ — 1,5-Naphthalenedisulfonic acid, 3-{4-[(4-chloro-6-p-sulfoanilino-s-triazin-2-yl)amino]-o-tolylazo}-, trisodium salt

$N_8C_{32}H_{16}ClFe$ — Phthalocyanine, (chloroiron)^{II} derivative

$N_8C_{32}H_{16}(GeCl_2)$ — Phthalocyanine, dichlorogermanium derivative

$N_8SO_4Cl_2C_{32}H_{22}$ — 5-Pyrazolol, 4-[2,8-dichloro-7-(5-hydroxy-3-methyl-1-phenyl-4-pyrazolylazo)-dibenzothiophene-3-ylazo]-3-methyl-1-phenyl-, S,S-dioxide

$N_8SO_5Cl_2C_{31}H_{28}$ — 2-Naphthanilide, 3'-[(4,6-dichloro-s-triazin-2-yl)amino]-4-[5-(diethyl-sulfamoyl)-2-methoxyphenylazo]-3-hydroxy-

$N_8SO_{10}Cl_2C_{32}H_{22}$ — Acetoacetanilide, 2-{2,8-dichloro-7-[1-(p-nitrophenylcarbamoyl)acetonyl-azo]dibenzothiophene-3-ylazo}-4'-nitro-, S,S-dioxide

$N_8S_3O_9ClC_{26}H_{18}Na_3$ — 1,5-Naphthalenedisulfonic acid, 3-{-4[4-chloro-6-(2-p-sulfophenylhydrazino)-s-triazin-2-yl]amino}-o-tolylazo}-, trisodium salt

$N_8S_3O_{10}C_{21}H_{17}Na_3$ — 1-Naphthol-3,6-disulfonic acid, 8-{[4,6-bis(methylamino)-s-triazin-2-yl]-amino}-2-(o-sulfophenylazo)-, trisodium salt

$N_8S_3O_{10}C_{35}H_{45}Na_3$ — 1-Naphthol-3,6-disulfonic acid, 8-{[4,6-bis(octylamino)-s-triazin-2-yl]-amino}-2-(o-sulfophenylazo)-, trisodium salt

$N_9SC_{33}H_{26}\cdot Cl$ — N,N'-Bis(p-phenylazophenyl)-C-2-benzothiazolyl)-formazan

$N_9S_2O_{11}C_{23}H_{23}Na_2$ — 1-Naphthol-3-sulfonic acid, 7-amino-8-{5-{[4,6-bis[bis(hydroxymethyl)amino]-s-triazin-2-yl]amino}-2-sulfophenylazo}-, disodium salt

$N_9S_2O_{12}C_{25}H_{25}Na_2$ — 1-Naphthol-3-sulfonic acid, 7-acetamido-2-{5-{[4,6-bis[bis(hydroxymethyl)-amino]-s-triazin-2-yl]amino}-2-sulfophenylazo}-, disodium salt

$N_9S_4O_{12}C_{25}H_{21}$ — 1-Naphthol-3,6-disulfonic acid, 8-benzenesulfonamide-2-{5-[(4,6-diamino-s-triazin-2-yl)amino]-2-sulfophenylazo}-

$N_{10}SC_7H_8$ — Guanidine [α-(1H-tetrazol-5-ylazo)-2-thenylideneamino

$N_{10}S_2C_{20}H_{10}$ — 5,24: 12,17-Diimino-7,10: 19,22-diepithiodibenzo[h,r]-[1,3,4,6,11,13,14,16]-octaazacycloeicosine

$N_{10}S_2O_8C_{23}H_{18}Cr_{1/2}$ — 2-Naphthol-4-sulfonic acid, 6-[(4,6-diamino-s-triazin-2-yl)amino]-1-(5-hydroxy-3-methyl-1-p-sulfophenyl-4-pyrazolylazo)-, chromium^{III} derivative

Formula	Name
$N_{10}S_2O_8C_{23}H_{18}Cu$	2-Naphthol-4-sulfonic acid, 6-/(4,6-diamino-s-triazin-2-yl)amino/-1-(5-hydroxy-3-methyl-1-p-sulfophenyl-4-pyrazolylazo)-, copper^{II} derivative
$N_{10}S_4O_{12}C_{39}H_{22}Cu$	Phthalocyaninetrisulfonic acid, /(p-carbanoylphenyl)sulfamoyl/-copper^{II} derivative
Na_3VO_4	Sodium vanadate (Na_3VO_4)
$OC_{23}H_{17} \cdot ClO_4$	Pyrylium perchlorate, 2,4,6-triphenyl-
$OC_{29}H_{20}$	Cyclopentadienone, tetraphenyl-
$OClC_{23}H_{16} \cdot Cl$	1-Benzopyrylium chloride, 2-(p-chlorostyryl)-4-phenyl-
$O_2C_7H_5Cu_{1/2}$	Tropolone, copper^{II} derivative
$O_2C_7H_5Cu_{1/2}$	Salicylaldehyde, copper^{II} derivative
$O_2C_{11}H_7Cu_{1/2}$	1-Naphthaldehyde, 2-hydroxy-, copper^{II} derivative
$O_2C_{15}H_{21}Ni_{1/2}$	Nickel complex of 2-ethylhexylsalicylate
$O_2C_{20}H_{10}$	Indeno/1,2-b/fluorene-6,12-dione
$O_2C_{20}H_{10}$	Indeno/2,1-a/fluorene-11,1,-dione
$O_2C_{22}H_{12}$	Dibenzo/a,h/pyrene-7,14-dione
$O_2CH(AlOH)_{1/2}$	Aluminum formate
$O_2Cl_3C_7H_3$	p-Benzoquinone, 2,3,5-trichloro-6-methyl-
$O_3C_9H_{15}Ni_{1/2}$	2-Hexenoic acid, 3-hydroxy-5-methyl-, ethyl ester, nickel^{II} derivative
$O_3C_{21}H_{25} \cdot Ni$	Nickel complex of 2-hydroxy-4-octyloxybenzophenone
$O_3C_{22}H_{16}$	Spiro/2H-1-benzopyran-2,3'-/3H/naphtho/2,1-b/-pyran, 8-methoxy-
$O_3C_{24}H_{20}$	Spiro/2H-1-benzopyran-2,3'-/3H/naphtho/2,1-b/-pyran, 8-methoxy-2',3-dimethyl-
$O_4C_{14}H_6Ti_{1/2}$	Anthraquinone, 1,2-dihydroxy-, dititanium^{IV} derivative
$O_4C_{21}H_{24}Ni$	Benzophenone, 2,2'-dihydroxy-4-octyloxy-, nickel^{II} derivative
$O_4C_{22}H_{16}$	/Δ2,2'(1H,1H')-Binaphthalene/-1,1'-dione, 4,4'-dimethoxy-
$O_4C_{26}H_{14}$	1,3-Indanedione, 2,2'-(p-phenylenedimethylidyne)di-
$O_4C_{30}H_{22}$	3-Hexynedioic acid, tetraphenyl
$O_4C_{32}H_{32}$	Pentacyclo/19.3.1.1^{3,7}.1^{9,13}.1^{15,19}/octacosa-1(25),3,5,7(28),9,11,13(27),15,17,19(26),21,23-dodecaene-25,26,27,28-tetrol-5,11,17,23-tetramethyl-
$O_6C_{22}H_{10}$	Indeno/1,2-b/fluorene-2,8-dicarboxylic acid, 6,12-dihydro-6,12-dioxo-
$O_6C_{22}H_{12}$	1,4,6,11,13-Pentacene-hexane-5,7,12,14-tetrahydro
$PC_{20}H_{20} \cdot I$	Triphenyl ethyl phosphonium iodide
$PClC_{12}H_{10} \cdot NCCu$	Phosphine, chlorodiphenyl-, compound with CuCN
$PN_3C_{24}H_{23} \cdot Cl$	Phosphonium chloride, /4,6-dimethyl-s-triazin-2-yl)methyl/triphenyl-
$PN_3SO_2C_{25}H_{18}$	Phosphine imide, N-(6-nitro-2-benzothiazolyl)-P,P,P-triphenyl-
$POC_{25}H_{26} \cdot I$	Phosphonium iodide, dimethyl(3-oxo-1,5-diphenyl-4-pentenyl)phenyl-
$PO_2C_{14}H_{16} \cdot Cl$	Phosphonium chloride, bis(hydroxymethyl)-diphenyl

Formula	Name
$PO_3C_{12}H_{27} \cdot 1/4Cr(NO_3)_3 \cdot 17_4HNO_3 \cdot 3/4H_2O$	Chromium nitrate-dibutyl butyl phosphonate complex
$PO_3C_{12}H_{27} \cdot 1/5NiN_2O_6$	Phosphonic acid, butyl-, dibutyl ester, compound with $Ni(NO_3)_2$
$PO_3C_{24}H_{51} \cdot 1/4[Cu(NO_3)_2 \cdot HNO_3 H_2O]$	Cupric nitrate-bis(2-ethylhexyl)-2-ethylhexylphosphonate complex
$PO_3C_{24}H_{51} \cdot \alpha[Ni-(NO_3)_2 \cdot CH_3O]$	Nickel nitrate-bis(2-ethylhexyl)-2-ethylhexylphosphonate complex
$PO_4C_{12}H_{27} \cdot 1/4CuN_2O_6$	Butyl phosphate $[(C_4H_9O)_3PO]$ compound with $Cu(NO_3)_2$
$PO_4C_{12}H_{27} \cdot 1/4NiN_2O_6$	Butyl phosphate $[(C_4H_9O)_3PO]$ compound with $Ni(NO_3)_2$
$PO_4C_{16}H_{34}Co_{1/2} \cdot 3/2 PO_4C_{16}H_{35}$	Hexanol, 2-ethyl-, H phosphate, Co^{II} salt, compound with bis(2-ethylhexyl) H phosphate
$PS_2C_{16}H_{34}Ni_{1/2}$	Phosphinodithioic acid, dioctyl-nickel^{II} salt
$PS_2O_2C_8H_{18}Ni_{1/2}$	Bis(di-sec-butyldithiophosphato)nickel(II)
PrN_3O_9	Praseodymium nitrate $[Pr(NO_3)_3]$
$SiC_{19}H_{18}$	Silane, methyltriphenyl-
$SiCl_2C_{12}H_{12} \cdot N_2C_{10}H_8$	2,2'-Bipyridyl complex of dichlorodiphenylsilane
$Si_3O_3C_6H_{18}$	Cyclotrisiloxane, hexamethyl-
$Si_4O_4C_{48}H_{40}$	Cyclotetrasiloxane, octaphenyl-
$Si_4TiO_4C_{72}H_{60}$	Titanium, tetrakis(triphenylsiloxy)-
$SO_2C_{28}H_{40}Ni \cdot NH_3$	Phenol, 2,2'-thiobis[4-(1,1,3,3-tetramethylbutyl)-, dinickel^{II} derivative, monoammine
$SO_2C_{28}H_{40} \cdot Ni$	Nickel complex of 2,2'-thiobis(4-t-octylphenol)
$SO_2C_{28}H_{40}Ni \cdot NH_3$	Ammine-nickel complex of 2,2'-thiobis(4-t-octylphenol)
$SO_9C_{22}H_{15}Na_3$	Salicylic acid, 5-[α(3-carboxy-5-methyl-4-oxo-2,5-cyclohexadienylidene)-o-sulfobenzylidine]-3-methyltrisodium salt
$S_2OC_{17}H_{12}$	3-Benzoylmethylene-1,2-dithiole
$S_3C_{17}H_{12}$	4,6-Diphenylthiothiophthene
$S_3C_{23}H_{15}$	4,5,6-Triphenylthiothiophthene
$S_4C_{15}H_8$	Methane, bis(3H-1,2-benzodithiol-3-ylidene-
$SnN_3C_{22}H_{15} \cdot SnOC_{16}H_{16}$	Methanetricarbonitrite-(triphenylstannyl)- compound with triphenyl hydroxide
$Sn_2O_4C_{20}H_{44}$	1,3,6,8,27-Tetroxadistannecane, 2,2,7,7-tetrabutyl
$Ti_2O_5F_6C_{24}H_{20}$	Titanium, oxybis[dicyclopentadienyl-(trifluoroacetoxy)-

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