MS, m/e 300 (11), 134 (100), 121 (15).

3-Methoxyestra-1,3,5(10),9(11)-tetraen-17-one (15). A mixture of compound **14** (80 mg, 0.27 mmol), acetic acid (3.0 mL), and perchloric acid 70% (80 μL) was left standing at room temperature for 4 h. Water (15 mL) was added, and the mixture was extracted with ether (3 × 10 mL). The extract was washed with saturated aqueous potassium bicarbonate and dried ($MgSO_4$); after filtration followed by evaporation of solvent, it was chromatographed on a silica gel column using a hexane–ethyl acetate (5:1) mixture as an eluent, affording compound **15** (65 mg, 86%): mp 144–147 °C; $[\alpha]^{20}_D$ +287° (c 0.5, $CHCl_3$); IR (KBr) 1737 cm^{-1}; ^1H NMR δ 0.94 (s, 3 H, CH_3), 2.80–3.0 (m, 2 H), 6.07–6.22 (m, 1 H, $C_{11}H$), 6.63 (d, 1 H, J = 2.8 Hz, C_4H), 6.74 (dd, 1 H, J = 8.8 Hz, J = 2.8 Hz, C_2H), 7.53 (d, 1 H, J = 8.8 Hz, C_1H); UV (EtOH) λ_{max} 264 (ϵ 18 300), 298 (3200), 310 nm (2300); MS, m/e 282. Literature:[7] mp 142.5–144 °C; $[\alpha]_D$ +290.92° (c 0.5, $CHCl_3$); IR ($CHCl_3$) 1735 cm^{-1}; ^1H NMR δ 0.92 (s, 3 H, CH_3), 3.76 (s, 3 H, CH_3O), 6.13 (m, 1 H, $C_{11}H$), 6.68 (m, 2 H, aromatic H), 7.52 (d, 1 H, aromatic H); UV (95% EtOH) λ_{max} 263 (ϵ 19 300), 297 (3400), 310 nm (2220).

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Registry No. **1**, 17553-86-5; **3** (isomer 1), 113726-22-0; **3** (isomer 2), 113726-23-1; **4**, 113726-27-5; **5**, 3907-67-3; **6**, 116503-14-1; **7** (isomer 1), 116503-15-2; **7** (isomer 2), 116503-16-3; **8**, 116503-17-4; **9**, 116503-18-5; **10**, 116503-19-6; **11**, 116503-20-9; **12**, 116503-21-0; **13**, 116503-22-1; **14**, 968-98-9; **15**, 1670-49-1; m-$CH_3OC_6H_4CH_2CHO$, 65292-99-1; m-$CH_3OC_6H_4CH_2COCl$, 6834-42-0.

Synthesis and Reactions of Cyanovinyl-Substituted Benzenediazonium Salts for Nonlinear Optics

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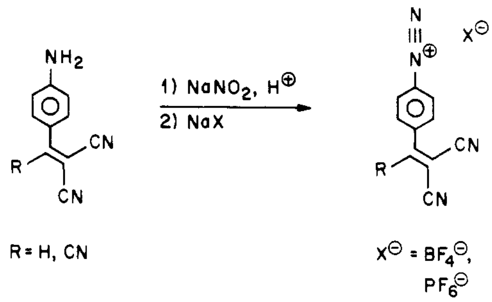
A renewed interest in azo dyes has been sparked by recent efforts to find organic second-order nonlinear optical (NLO) materials suitable for applications such as harmonic generation and optical switching.[1,2] The active components in these materials typically consist of conjugated electron systems connecting terminal electron donor and electron acceptor moieties. By incorporating different end groups and changing the effective length and type of conjugating group, the nonlinear optical activity (molecular susceptibility) of a particular system can be adjusted with some degree of predictability.[3] Azo dyes are of particular interest because they can be fairly readily prepared with a wide range of donor and acceptor groups, and the π system provides excellent conjugation[4] that is necessary for high molecular susceptibilities.[1,2]

We have determined the hyperpolarizabilities of a number of azo dyes, including some with better electron acceptor groups than the nitro group (e.g., di- and tricyanovinyl).[3] These strong electron withdrawing groups conjugated to an amino donor via a diphenylazo linkage provide compounds with some of the largest molecular susceptibilities reported to date. Because the typical synthesis of these compounds may require several steps, which involve severe reaction conditions,[5–7] we have investigated new synthetic methods that will simplify the preparation of such azo dyes and allow the preparation of materials that are otherwise inaccessible (e.g. polymeric dyes). We have isolated benzenediazonium salts derived from (dicyanovinyl)- and (tricyanovinyl)anilines and successfully coupled them to N,N-disubstituted anilines to give azo dyes of interest for NLO studies.

Results

Diazotization Reactions. The benzenediazonium salts are isolated by following the general procedure described by Roe[8] for simple benzene derivatives. This involves diazotization of the aniline in HCl/H_2O with sodium nitrite followed by precipitation of the fluoroborate salt by addition of sodium tetrafluoroborate (eq 1). Due to the low solubility of di- and (tricyanovinyl)aniline in $HCl/H_2O/NaNO_2$, we find that HCl/HOAc is a better reaction medium for (tricyanovinyl)aniline. The dicyanovinyl derivative requires H_2SO_4/HOAc for complete dissolution. While the diazonium salts can be formed and isolated in HCl/H_2O, the yield and activity of the salt is greatly improved if the aniline is completely dissolved after addition of the nitrite. It is also important to control the rate of nitrite addition in order to maintain the solution temperature below ~10 °C to minimize side reactions. During the formation of the (dicyanovinyl)benzenediazonium salts, a second product is formed if reaction conditions are not carefully controlled. We have isolated a red solid whose spectral characteristics (NMR, MS) are consistent with [[(dicyanovinyl)phenyl]azo]malononitrile. We believe this side product may be formed by electrophilic attack of the diazonium ion on the dicyanovinyl group of the starting aniline, followed by hydrolytic loss of the aniline moiety.



(1)

Via a similar procedure, we have also prepared hexafluorophosphate salts. In a comparison of tetrafluoroborate versus hexafluorophosphate as the counter ion, we find the PF_6^- diazonium salts to be less soluble in the reaction mixture than the corresponding BF_4^- salts, making their isolation easier. Therefore we have usually isolated the PF_6^- salts, keeping the amount of solvent to a minimum. The salts are not purified after isolation since recrystallization is difficult due to low decomposition temperatures and because of the similar solubilities of the salts and other components. However, the activity of the

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† AT&T Bell Laboratories, Summer Research Program participant, 1987.

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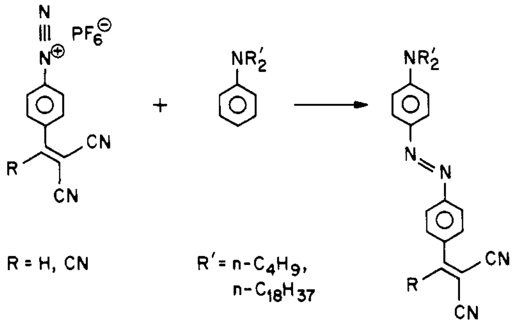
reagent can be determined by NMR integration so that a quantitative or excess amount of reagent can be used for the coupling reaction. With care the diazonium salts can be obtained with >98% activity.

Azo Coupling Reactions. The coupling of the diazonium salts to a substituted aniline involves a one-step procedure in HOAc/(H_2O)/NaOAc solutions (eq 2).



(2)

Complete dissolution of the aniline is important for maximum yields. [[(Tricyanovinyl)phenyl]azo]-*N*,*N*-dibutylaniline is prepared by this procedure in 49% yield. The isolated product is identical with that which we have prepared via the standard multistep synthetic route[5-7] in 25% yield. The butyl groups increase the solubility of this azo dye over the corresponding diethyl analogue, for which the molecular susceptibility has already been reported.[3]

Also of interest are azo dyes derived from dioctadecylaniline[9] whose long alkyl chains make them attractive for Langmuir–Blodgett film studies.[10] However, these chains also complicate the preparation of azo dyes via a multistep synthesis because isolation and purification of products would be tedious if not impossible. In a one-step synthesis, we have effectively coupled dioctadecylaniline with the (dicyanovinyl)- and (tricyanovinyl)benzenediazonium hexafluorophosphates to give the corresponding azo dyes. Since the aniline is insoluble in water, the reactions are carried out in glacial acetic acid. The dicyanovinyl azo dye precipitates out of the reaction mixture, but in the case of the tricyanovinyl azo dye, water is added to precipitate the product. Both materials are successfully purified by column chromatography.

Finally, we have employed this azo coupling methodology in the synthesis of methacrylate copolymers with azo dye side chains. Specifically, the methacrylate ester of *N*-ethyl-*N*-(hydroxyethyl)aniline copolymerized with methyl methacrylate was converted to a polymeric azo dye in high yield with <10% unreacted aniline residues. To our knowledge, this is the first example of an azo coupling reaction performed on a polymer. Batches of 20 g or more of polymer dye have been prepared in this manner.

Discussion

Azo dyes, such as (dicyanovinyl)-or [[(tricyanovinyl)-phenyl]azo]-*N*,*N*-diethylaniline have previously been prepared[3] from the reaction of a N,N-disubstituted aniline with diazotized *p*-aminobenzaldehyde,[5] followed by modification of the formyl group by malononitrile condensation (→ dicyanovinyl)[6] and then addition of sodium cyanide followed by lead tetraacetate oxidation (→ tricyanovinyl).[7] Besides requiring several steps, some of the necessary reagents react with other parts of the molecule, e.g., in the case of highly functionalized anilines, causing side reactions. In addition, the diazotization reaction goes in inconsistent yields, often giving low yields of the dye.

Preparation and isolation of diazonium salts of the electron-withdrawing portion of the azo dye provides several advantages. The multiple steps requiring harsh reagents can be done in bulk quantities before preparation of the salts, eliminating the need for repetitive reactions. Isolation of the salts permits a quantitative determination (e.g. by NMR) of activity giving more predictable yields in the coupling reaction. Also, the diazonium salts can be coupled to a wide variety of anilines (monomeric or polymeric) in a controlled manner under moderate reaction conditions (acetic acid), reducing the possibilities of side reactions or decomposition. The tricyanovinyl group, in particular, is susceptible to degradation under basic conditions[4] and probably would not survive further reactions such as reductions or radical reactions.

Conclusion

We have developed a convenient procedure for the preparation and isolation of benzenediazonium hexafluorophosphates derived from cyanovinyl-substituted anilines. These salts are useful for the one-step synthesis of a variety of azo dyes under moderate reaction conditions. The coupling reaction appears to be general for most aniline moieties and should be useful for making dyes that are otherwise inaccessible, such as polymeric azo dyes for fabrication of nonlinear optical materials.[11]

Experimental Section

Materials. Inorganic salts ($NaNO_2$, $NaPF_6$, $NaBF_4$) were used as purchased without further purification. Dibutylaniline (Kodak) and dioctadecylamine (Fluka) were used as purchased.

Diazonium Salts. (Dicyanovinyl)benzenediazonium Hexafluorophosphate. 4-(Dicyanovinyl)aniline[7] (2.7 g, 0.016 mol) was stirred into a mixture of sulfuric acid (2.5 mL (17.8 M), 0.045 mol) and glacial acetic acid (30 mL) cooled in an ice bath to just above the freezing point (~10 °C). The solution turned dark brown, and the aniline may not have been completely dissolved. Sodium nitrite (1.1 g, 0.016 mol) in a minimum amount of H_2O was added slowly to maintain the temperature below 10 °C. The solution turned red-brown, and most of the suspended aniline was dissolved. The solution was decanted away from any undissolved solid and warmed to room temperature while being stirred for 15 min. Solid $NaPF_6$ (1.82 g, 0.016 mol) was added in small portions. A solid began to precipitate immediately. Stirring was continued for an additional 10 min, and the white solid was collected on a filter and dried under vacuum: yield, 3.8 g (90%); mp 108–110 °C dec; NMR (^1H, 360 MHz, CD_3CN) δ 8.25 (2 H, d), 8.30 (1 H, s), 8.60 (2 H, d). Integration of the spectrum showed the solid to be ~99% diazonium salt. The reaction proceeds best with freshly prepared (dicyanovinyl)aniline and careful control of the temperature during nitrite addition.

(Tricyanovinyl)benzenediazonium hexafluorophosphate was prepared by a similar procedure from (tricyanovinyl)aniline.[7] In this reaction, HCl can be used instead of H_2SO_4 to give a quantitative yield of diazonium salt (>98% active): mp 119–121 °C dec; NMR (^1H, 360 MHz, CD_3CN) δ 8.30 (2 H, d), 8.60 (2 H, d).

Azo Coupling Reactions. [[4-[[4-(Dibutylamino)-phenyl]azo]phenyl]methylene]propanedinitrile. (Tricyanovinyl)benzenediazonium hexafluorophosphate (0.84 g, 0.0024 mol) was dissolved in a mixture of 15 mL of glacial AcOH and 15 mL of H_2O at 0 °C. Dibutylaniline (0.5 g, 0.0024 mol) was added dropwise, and the solution was stirred for 15 min at 0 °C. Sodium acetate (0.3 g, 0.0037 mol) was added in two portions with continued stirring at 0 °C for 30 min. The solution was warmed

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to room temperature and stirred overnight. A dark blue-green solid was collected and purified by column chromatography on silica gel (hexane–CH_2Cl_2) to give the azo dye: yield 0.5 g (49%); mp 137–140 °C; NMR (^1H, 360 MHz, $CDCl_3$) δ 1.0 (6 H, t), 1.5 (4 H, m), 1.7 (4 H, m), 3.4 (4 H, t), 6.7 (2 H, d), 7.9 (2 H, d), 8.0 (2 H, d), 8.2 (2 H, d); UV–vis (CH_2Cl_2) λ_{max} 417, 625 nm. Anal. Calcd for $C_{25}H_{26}N_6$: C, 73.14; H, 6.38; N, 20.47. Found: C, 72.54; H, 6.40; N, 20.26.

***p*-Nitro-*N*,*N*-dioctadecylaniline.** *p*-Fluoronitrobenzene (15.2 g, 0.11 mol), dioctadecylamine (57.2 g, 0.11 mol), and triethylamine (15.3 mL, 0.11 mol) were dissolved in 20 mL of 1-methyl-2-pyrrolidinone (NMP) and heated at 100 °C overnight with stirring. A yellow solid that precipitated during the heating was collected on a Buchner funnel, triturated, and washed with warm EtOH. A second fraction was isolated by mixing the NMP filtrate with hot EtOH and filtering the resulting solid: yield 45 g (64%); mp 85–88 °C; NMR (^1H, 360 MHz, $CDCl_3$) δ 0.9 (6 H, t), 1.25 (60 H, m), 1.6 (4 H, m), 2.7 (4 H, t), 6.5 (2 H, d), 8.05 (2 H, d).

***p*-Amino-*N*,*N*-dioctadecylaniline.** *p*-Nitro-*N*,*N*-dioctadecylaniline (3 g, 0.0046 mol), $SnCl_2{\cdot}2H_2O$[12] (4.7 g, 0.021 mol), and Sn (0.55 g, 0.0046 mol) were added to a mixture of concentrated HCl (15 mL), toluene (15 mL), and EtOH (45 mL), which was refluxed overnight with stirring. After cooling, petroleum ether (50 mL) was added to the reaction mixture, and this solution was poured into H_2O (100 mL). The aqueous layer was brought to pH 8 with saturated Na_2CO_3 solution. The petroleum ether layer was separated, and the aqueous layer washed with two 50-mL portions of petroleum ether. The combined petroleum ether fractions were dried over anhydrous Na_2CO_3, and the solvent was removed on a rotary evaporator to give a light brown oil used without further purification: yield 2.5 g (89%); NMR (^1H, 360 MHz, $CDCl_3$) δ 0.9 (6 H, t), 1.25 (60 H, m), 1.5 (4 H, m), 3.1 (4 H, t), 6.6 (4 H, AB quartet).

***N*,*N*-Dioctadecylaniline.** *p*-Amino-*N*,*N*-dioctadecylaniline (2.5 g, 0.004 mol) was dissolved in a mixture of H_3PO_2[13] (10 mL) and EtOH (10 mL) with stirring at 0 °C. Sodium nitrite (0.4 g, 0.006 mol) was added in small portions over 5 min while the temperature was maintained below 5 °C. The solution turned red, and the stirring was continued for 30 min at 0 °C and then overnight at room temperature. Aqueous NaOH (5 g in 60 mL of H_2O) was added, and the solution was extracted with two portions (100 mL) of petroleum ether. The organic layer was dried over anhydrous K_2CO_3 and concentrated on a rotary evaporator. The brown oil was chromatographed on 50 g of neutral alumina with petroleum ether as the eluent to give a light tan wax, which was used without further purification: yield 1.5 g (63%); mp 40–41 °C; NMR (^1H, 360 MHz, $CDCl_3$) δ 0.9 (6 H, t), 1.3 (60 H, m), 1.55 (4 H, m), 3.2 (4 H, t), 6.6 (3 H, m), 7.2 (2 H, m); MS, *m/e* (relative intensity) 598 (4, M^+), 358 (56), 120 (100), 43 (66). Anal. Calcd for $C_{42}H_{79}N$: C, 84.34; H, 13.31; N, 2.34. Found: C, 84.04; H, 13.34; N, 2.39.

[[4-[[4-(Dioctadecylamino)phenyl]azo]phenyl]methylene]propanedinitrile. Dioctadecylaniline (0.37 g, 0.6 mmol) was dissolved in 30 mL of glacial AcOH at room temperature. (Dicyanovinyl)benzenediazonium hexafluorophosphate (0.25 g (80% diazonium salt), 0.6 mmol) was added with vigorous stirring. After 15 min, NaOAc (0.098 g, 1.2 mmol) was added in two portions, and the solution was then stirred overnight. A dark red solid was collected and purified by column chromatography on silica gel (20 g) with hexane–EtOAc to give the desired product: yield 0.4 g, (85%); mp 59–60 °C; NMR (^1H, 360 MHz, $CDCl_3$) δ 0.85 (6 H, t), 1.25 (60 H, m), 1.6 (4 H, m), 3.35 (4 H, t), 6.67 (2 H, d), 7.75 (1 H, s), 7.86 (2 H, d), 7.90 (2 H, d), 8.0 (2 H, d). Anal. Calcd for $C_{52}H_{83}N_5$: C, 80.25; H, 10.75; N, 9.00. Found: C, 79.65; H, 10.68; N, 8.54.

[4-[[4-(Dioctadecylamino)phenyl]azo]phenyl]ethenetricarbonitrile. Via a similar procedure to that described above, dioctadecylaniline was reacted with (tricyanovinyl)benzenediazonium hexafluorophosphate in glacial acetic acid/solution acetate. After being stirred overnight, the AcOH solution was poured into H_2O and stirred for 10 min. The blue-black solid was collected on a Buchner funnel and purified by column chromatography on silica gel with hexane/EtOAc: yield 0.51 g (80%); mp 35–37 °C; NMR (^1H, 360 MHz, $CDCl_3$) δ 0.85 (6 H, t), 1.25 (60 H, m), 1.6 (4 H, m), 3.35 (4 H, t), 6.7 (2 H, d), 7.85 (2 H, d), 7.95 (2 H, d), 8.1 (2 H, d). Anal. Calcd for $C_{53}H_{82}N_6$: C, 79.25; H, 10.29; N, 10.46. Found: C, 79.31; H, 10.18; N, 10.02.

Coupling of *p*-(Dicyanovinyl)benzenediazonium Hexafluorophosphate to Copolymers of (*N*-Ethylanilino)ethyl Methacrylate and Methyl Methacrylate. The copolymer was dissolved in AcOH (20 mL/g) and stirred overnight with 1.25 equiv of the diazonium salt and 2 equiv of NaOAc. The solids were allowed to settle, and the supernatant liquid was decanted into 5 volumes of stirred water. The residual solids were dissolved in MEK with stirring and added to the aqueous mixture. The precipitated solids were collected and dried in air. An impurity absorbing at 440 nm was removed by reprecipitation from MEK–CCl_4, and an ionic impurity was removed by reprecipitation from MEK–dilute aqueous NaOAc. The yield was 60–90%: NMR ($CDCl_3$, back-bone omitted) δ 3.6 (OCH_3 and NCH_2), 4.1 (OCH_2), 6.8 and 7.9 (Ar H and vinyl H); UV max (film) 510, (Me_2SO) 510 nm. Coupling with *p*-(tricyanovinyl)benzenediazonium hexafluorophosphate was accomplished in a similar manner, and its absorption maximum was at 580 nm (film).

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Registry No. 4-(Dicyanovinyl)aniline, 17082-32-5; [[4-[[4-(dibutylamino)phenyl]azo]phenyl]methylene]propanedinitrile, 116350-27-7; dibutylaniline, 613-29-6; *p*-nitro-*N*,*N*-dioctadecylaniline, 116350-26-6; *p*-fluoronitrobenzene, 350-46-9; dioctadecylamine, 112-99-2; *p*-amino-*N*,*N*-dioctadecylaniline, 85074-67-5; [[4-[[4-(dioctadecylamino)phenyl]azo]phenyl]methylene]propanedinitrile, 116350-28-8; dioctadecylaniline, 72072-19-6; [4-[[4-(dioctadecylamino)phenyl]azo]phenyl]ethenetricarbonitrile, 116350-29-9.

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Lewis Acid Induced α-Alkoxyalkylation of 1,3-Dicarbonyl Compounds

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The α-alkoxyalkylation of carbonyl compounds represents one of the most important C–C bond forming reactions.[1] In the last few years an impressive series of papers appeared in the literature, showing that Lewis acid induced α-hydroxy- and α-alkoxyalkylation of carbonyl compounds could be achieved via the corresponding enol esters, alkyl enol ethers, or, more conveniently, silyl enol ethers.[2] Furthermore, high enantioselectivity or stereoselectivity is possible by using suitable Lewis acids.[3,4] In particular, the employment of trimethylsilyl trifluoromethanesulfonate (TMSOTf), an efficient catalyst in the aldol-type condensation of enol silyl ethers with acetals or ketals, has allowed the formation of adducts with a high to moderate degree of stereoselectivity.[3]

On the other hand, successful α-alkoxyalkylations of active methylene compounds of type 1[5] are often precluded

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