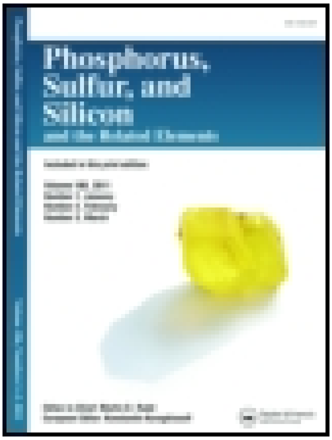

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NEW CHEMISTRY OF N,N´-BIS(ARYL)-ETHANE-l,2-DIYLIDENEDIAMINES TOWARDS CARBON DISULFIDE AND PHENYL ISOTHIOCYANATE

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NEW CHEMISTRY OF N,N′-BIS(ARYL)-ETHANE-1,2-DIYLIDENEDIAMINES TOWARDS CARBON DISULFIDE AND PHENYL ISOTHIOCYANATE

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N,N′-Bis(aryl)-ethane-1,2-diylidenediamine (**1a,b**) reacts with carbon disulfide to give 5-hydroxy-2-arylamino-3N-arylamino-4H-1, 3-thiazoline-2-thione (**2a,b**) in addition to N-aryl-thiourea derivatives **3–5**. The reaction of phenyl isothiocyanate with **1a,b** provides 1,2-diphenylamino-1,1,2,2-tetra-arylamino-ethane (**6a,b**), 1N-phenyl-3N-aryl-diazetidine-2,4-dithione (**7a,b**) and 1-toluidino-1,1 ,2,2-tetra-arylamino-ethane (**8a,b**).

Keywords: carbon disulfide; phenyl isothiocyanate; diimines

It has been reported that N,N′-bis(aryl)-ethane-1,2-diylidenediamine (**1**)[1,2] reacts with tetracyanoethylene (TCNE), 1,4-benzo- and naphtho-quinone derivatives to give imidazolidine, quinoline, quinoxaline, indole, carbazole and other interesting condensed heterocyclic derivatives[3,4]. On the other hand, 3,4,5,6-tetrachloro-1,2-benzoquinone (*o*-CHL) reacts with **1** to give a transient condensation product which underwent [4+2] cycloaddition reaction with another molecule of diimine[5]. Reaction of benzyne with diimines **1** exerts a different behavior as dienophile and it gives with **1**, bis-acridine derivatives via [2+2] cycloaddtion reactions[6].

In extension of this work, the chemistry of diimine **1a,b** was investigated towards some selected electron deficient compounds such as carbon disulfide and phenyl isothiocyanate (Scheme 1).

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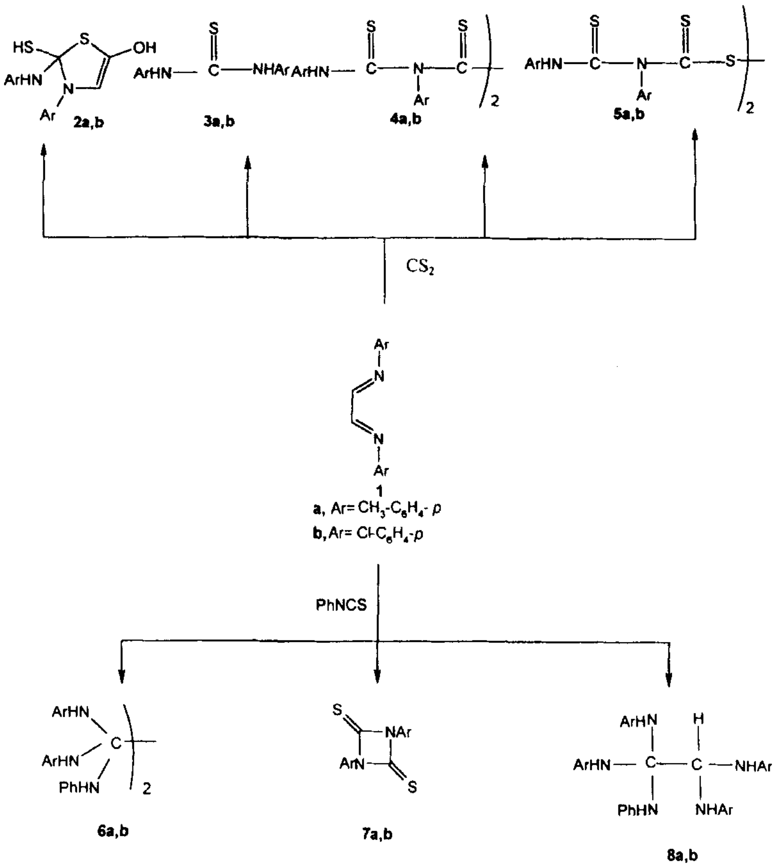

SCHEME 1

Reaction of diimine **1** with carbon disulfide is essentially based on the reactivity of both moieties. The reactivity of carbon disulfide itself depended on its polar stability which enhances the *ease* of carbophilic and/or thiophilic attack[7–9], thus reactions of carbon disulfide with electron-rich olefins were described as due to dipoles[10,11].

Heating *1a* with excess carbon disulfide in toluene afforded 5-hydroxy-2(*p*-toluidino)-3N(*p*-tolyl)-1,3-thiazoline-2-thione (*2a*), N,N′-di(*p*-toluidino)thiourea (*3a*), bis-[N,N′-(4,4′-di-*p*-toluidino-N-thiono]thiourea (*4a*) and bis-[N,N′-(4,4′-toluidino-N-thiocarbamido]thiourea (*5a*) in 82% total yield. Similarly, the reaction of *1b* with carbon disulfide gave 5-hydroxy-2(*p*-chloroanilino)-3N(*p*-chloroanilino)-1,3-thiazoline-2-thione (*2b*), N,N′-di(*p*-chloroanilino)thiourea (*3b*), bis-[N,N′-(4,4′-di-*p*-chloroanilino-N-thiono]thiourea (*4b*) and bis-[N,N-(4,4′-chloroanilino-N-thiocarbamido]thiourea (*5b*) in 63% total yield.

Using phenyl isothiocyanate, *1a* yielded with prolonged heating 1,2-diphenylamino-1,1,2,2-tetra-*p*-toluidino-thiourea (*6a*), 1N-phenyl-3N-p-toluidino-diazetidine-2,4-dithione (*7a*) and 1-anilino-1,1,2,2-tetra-*p*-toluidino-ethane (*8a*) in 68% total yield. *1b* on reaction with phenyl isothiocyanate produced 1,2-diphenylamino-1,1,2,2-tetra-*p*-chloroanilio-thiourea (*6b*), 1N-phenyl-3N-*p*-chlorophenyl-diazetidine-2,4-dithione (*7b*) and 1-anilino-1,1,2,2-tetra-*p*-chloroanilino-ethane (*8b*) in 62% total yield.

Water from the solvent used and hydrogen from the diimines are involved in formation of some reaction products.

Prolonged heating is required due to the unreactive behavior of this type of reactions, in which the steric course may play an important role. The structural proof of 1 N-phenyl-3N-(*p*-substituted-aryl)-diazetidine-2,4-dithione (**7a,b**) is based on its MS, ^{1}H-NMR, ^{13}C-NMR and IR spectra as well as elemental analyses. For example compound **7a** had the molecular ion peak at $m/z = 284$. ^{1}H-NMR spectrum revealed two double-doublets, one at $\delta = 6.90–6.93$ (J= 10.81 Hz) for two protons and another at $\delta = 6.97–7.00$ (J = 10.80 Hz) also for two protons. The phenyl protons appear as a multiplet at $\delta = 6.40–6.85$. ^{13}C-NMR spectrum showed the two C = S signals at $\delta = 188.18$ and at $\delta = 188.60$. The aromatic carbons appeared as fifteen signals, confirming its unsymmetrical structure. Although compound **7a,b** is a four member ring with two nitrogen atoms, it is stable in this form, since compounds with structural relationship are also stable[18].

We hope by introducing such reactions between diimine **1a,b** with the aforementioned electron π-deficient compounds that we describe to some extent the chemistry of these diimines towards these compounds. Since, organosulfur compounds play an important role in modern organic synthesis, not only because they constitute a particularly useful class of synthons[20] but also because they are of great biological interest[21]. So, the expectation that the formed products have biological effect is also taken into consideration.

EXPERIMENTAL

Melting points are uncorrected. IR spectra were obtained on Shimadzu 470 spectrophotometer using potassium bromide pellets. ^{1}H-NMR

(400.134 MHz) and ^{13}C-NMR (100.164 MHz) spectra were measured on Bruker AM 400 with TMS as an internal standard, s = singlet, d = doublet, m = multiplet. Mass spectra were recorded on Finnigan MAT 8430 instrument at 70 eV. Elemental analysis were performed by the microanalytical unit at Cairo University. For preparative layer chromatography (PLC): air dried 1.00 mm thick layers of slurry applied silica gel Merck PF$_{254}$ on 48 cm × 20 cm glass plates were employed using the solvents list for development. Zones were detected by quenching of indicator fluorescence upon exposure to 254 nm light, extracted and eluted with either toluene or toluene/ethyl acetate mixture.

Materials

N,N′-Bis(aryl)-ethane-1,2-diylidenediamines 1a,b were prepared according to refs.[1 and 2].

Reaction of N,N′-bis(aryl)-ethane-1,2-diylidenediamines (1a,b) with carbon disulfide

General Procedure

20 Mmols of carbon disulfide were mixed with 472 mg (2 mmols) of **1a,b** in 100 ml toluene and refluxed for 12–18 h (12 h for **1a** and 18 h for **1b**). The red color solution was concentrated in vacuum and the residue was applied on TLC plates chromatography using toluene as eluent. Four zones were separated and purified several times chromatographically. Compound **3a,b** was the fastest migrating zone, followed successively by compounds **5a,b** and **2a,b,** whereas product **4a,b** was the slowest migrating one. All products were recrystallized from the proper solvents.

5-Hydroxy-2(*p*-toluidino)-3N(*p*-tolyl)-1,3-thiazoline-2-thione (2a)

Yield 250 mg (38%), m.p = 130–131°C, yellow crystals (toluene). ^{1}H NMR (CDCl$_3$): $\delta = 2.26$ (s, 3H, CH$_3$), 2.31 (s, 3H, CH$_3$), 6.22–7.68 (m, 12H). ^{13}C NMR (CDCl$_3$): $\delta = 19.88$, 20.88 (2CH$_3$-Ar) 130.68, 131.73, 132.52, 132.63, 133.87, 133.93, 134.09, 134.12, 136.64, 136.75, 138.06, 139.18 (Ar-C), 145.08 (C-4), 159.16 (C-5), 172.60 (C-2). IR (KBr): $\nu^{-} = 3545$–3230 cm^{-1} (OH, NH), 3030–3008 (Ar-CH), 2985 (Aliph.-CH), 1600 (C=C). MS (70 eV) m/z (%) 330 (M^+, 100), 280 (60), 265 (16), 256 (100, 248(22), 222 (10), 149 (50), 106 (64), 91 (30), 57 (18), 45 (24). Calcd. for $C_{17}H_{18}N_2OS_2$ (330.474): C, 61.79, H, 5.49, N, 8.48, S, 19.41. Found: C, 61.66, H, 5.40, N, 8.40, S, 19.32.

5-Hydroxy-2-*p*-chloroanilino-3N(*p*-chloroanilino)-1,3-thiazoline-2-thione (2b)

Yield 200 mg (27%), m.p. = 160–161°C, yellow crystals (toluene). ^{1}H NMR ($CDCl_3$) $\delta = 6.20–7.53$ (m, 12H). ^{13}C NMR ($CDCl_3$): $\delta = 130.45$, 131.62, 132.14, 132.41, 133.01, 133.17, 133.91, 134.03, 136.16, 136.43, 138.01, 139.04 (Ar-C), 144.16 (C-4), 159.00 (C-5), 170.44 (C-2). IR (KBr): ν^{-} = 3540–3232 cm^{-}1 (OH, NH), 3018–3000 (Ar-CH), 1600 (C=C). MS (70 eV) m/z (%) 373 (M^{+2}, 30), 372 (M^{+1}, 60), 371 (M^+, 100), 369 (M^{-1}, 58), 355 (26), 353 (26), 318 (32), 317 (30), 285 (34), 126 (38), 42 (20). Calcd. $C_{15}H_{12}Cl_2N_2OS_2$ (371.317): C, 48.52, H, 3.26, Cl, 19.10, N, 7.54, S, 17.27. Found: C, 48.40, H, 3.18, Cl 19.00, N, 7.43, S, 17.19.

N,N′-Di(*p*-toluidino) thiourea (3a)

Yield 80 mg (16%), m.p.= 183°C, colorless crystals, (Ref. [12], 182–184°C).

N,N′-Di(*p*-chloroanilino) thiourea (3b)

Yield 70 mg (12%), m.p.= 166–167°C, colorless crystals, (Ref. [12] 165°C).

Bis-[N,N-(4,4′-di-*p*-toluidino-N-thiono] thiourea (4a)

Yield 150 mg (12%), m.p. = 132°C, red crystals (benzene), 1 H NMR ($CDCl_3$): $\delta = 2.29$ (s, 6H, $2CH_3$), 2.31 (s, 6H, $2CH_3$), 6.20–7.42 (m, 18H, 4Ar-H, 2NH). ^{13}C NMR ($CDCl_3$): δ_c= 19.93, 20.16 ($2CH_3$-Ar) 126.16, 127.29, 128.17, 128.42, 128.98, 129.68, 130.27, 131.53, 132.27, 134.09, 138.41, 138.54 (Ar-H), 180.40, 180.99 (C=S). IR (KBr): ν^{-}= 3331–3210 cm^{-1} (NH), 3035–3008 (Ar-CH), 2993–2891 (Aliph.-CH), 1603 (C=C), 1491(C=S). MS (70 eV) m/z (%) 599 (M^{+1}, 12), 598 (M^+, 18), 583 (56), 526 (8), 488 (12), 487 (22), 486 (54), 398 (34), 397 (100), 381 (86), 91 (48), 57 (30). Calcd. for $C_{32}H_{30}N_4S_4$ (598.881): C, 64.18, H, 5.05, N, 9.36, S, 21.42. Found: C, 64.09, H, 5.00, N, 9.28, S, 21.32.

Bis-[N,N-(4,4′-di-*p*-chloroanilino-N-thiono] thiourea (4b)

Yield 140 mg (11%), m.p. = 148-150°C, red crystals (benzene). ^1H NMR ($CDCl_3$): 6 = 6.18–7.35 (m, 18H, 4Ar-H, 2NH). ^{13}C NMR ($CDCl_3$): δ = 126.16, 127.24, 128.03, 128.40, 128.91, 129.50, 130.18, 131.44, 132.20, 134.02, 138.12, 138.32 (Ar-C), 180.00, 181.18 (C=S). IR (KBr): v^{-} = 3320–3208 cm^{-1}(NH), 3030–3001 (Ar-CH), 1600 (C=C), 1510 (C=S). MS (70 eV) m/z (%) 684 (M^{+4}, 12), 683 (M^{+3}, 20), 682 (M^{+2}, 42), 681 (M^{+1}, 60), 680 (M^+, 100), 679 (M^{-1}, 28), 645 (20), 644 (32), 643 (22), 555(13), 554 (18), 553 (10), 511 (40), 510 (10), 342 (68), 214 (30), 213 (32), 212 (46), 128 (16). Calcd. $C_{28}H_{18}Cl_4N_4S_4$ (680.566): C, 49.42, H, 2.67, Cl, 20.84, N, 8.23, S, 18.85. Found: C, 49.30, H, 2.61, Cl 20.78, N, 8.20, S, 18.78.

Bis-[N,N-(4,4′-*p*-toluidino-N-thiocarbamido] thiourea (5a)

Yield 210 mg (16%), m.p. = 135°C, red crystals (benzene). ^1H NMR ($CDCl_3$): δ = 2.28 (s, 6H, $2CH_3$), 2.30 (s, 6H, $2CH_3$), 6.18–7.45 (m, 18H, 4Ar-H, 2NH). ^{13}C NMR ($CDCl_3$): δ = 19.93, 20.18 ($2CH_3$-Ar). 128.09, 128.65, 129.19, 128.42, 129.94, 130.64, 130.93, 131.28, 132.66, 134.28, 138.53, 138.60 (Ar-H), 180.16, 182.91 (C=S). IR (KBr): v^{-} = 3331–3208 cm^{-1} (NH), 3035–3010 (Ar-CH), 2990–2890 (Aliph.-CH), 1608 (C=C), 1491(C=S). MS (70 eV) m/z (%) 663 (M^+, 14), 662 (M^{-1}, 14), 647 (16), 532 (8), 531 (10) 515 (12), 472 (6), 413 (20), 397 (14), 380 (24), 371 (8), 354 (42), 341 (14), 329 (100), 328 (38), 279 (8), 256 (10), 22 (12), 195 (14), 149 (16), 135 (12), 106 (80), 91(62). Calcd. for $C_{32}H_{30}N_4S_6$ (663.013): C, 57.97, H, 4.56, N, 8.45, S, 29.02. Found: C, 57.78, H, 4.49, N, 8.38, S, 28.93.

Bis-[N,N-(4,4′-di-*p*-chloroanilino-N-thiocarbamido] thiourea (5b)

Yield 190 mg (13%), m.p. = 154–156°C, red crystals (benzene). ^1H NMR ($CDCl_3$): δ = 6.20–7.38 (m, 18H, 4Ar-H, 2NH). ^{13}C NMR ($CDCl_3$):

δ = 128.11, 128.30, 129.02, 129.10, 129.80, 130.53, 130.82, 131.20, 132.54, 134.03, 138.20, 138.42 (Ar-C), 182.08, 183.80 (C=S). IR (KBr): $\bar{\nu}$: 3280–3210 cm^{-1} (NH), 3010–3001 (Ar-CH), 1608 (C=C), 1510 (C=S). MS (70 eV) m/z (%) 748 (M^{+4}, 10), 747 (M^{+3}, 24), 746 (M^{+2}, 38), 745 (M^{+1}, 60), 744 (M^{+}, 100), 743 (M^{-1}, 58), 713 (12), 712 (12), 711 (16), 710 (26), 669 (40), 668 (42), 667 (38), 545 (30), 544 (38), 543 (16), 500 (52), 400 (62), 498 (44), 373 (10), 372 (28), 371 (14), 341 (30), 340 (38), 239 (22), 183 (10), 182 (19), 145 (20), 128 (16). Calcd. $C_{28}H_{18}Cl_4N_4S_6$ (744.698): C, 45.16, H, 2.44, Cl, 19.04, N, 7.52, S, 25.84. Found: C, 45.00, H, 2.40, Cl 18.89, N, 7.48, S, 25.75.

2-Reaction of N,N′-bis(aryl)-ethane-1,2-diylidenediamines (1a,b) with phenyl isothiocynate

General Procedure

A mixture of 540 mg (4 mmols) of phenyl isothiocyanate and 472 mg (2 mmols) of **1a,b** was heated under reflux in 50 ml toluene for 72 hs. Toluene was evaporated and the brownish-red solution was concentrated. The residue was applied on TLC chromatography using toluene/ethyl acetate (10:1). Three zones were well separated in which compound **8a,b** was separated as the fastest migrating zone, followed by **7a,b** and finally compound **6a,b** is the slowest migrating one.

1,2-Diphenylamino-1,1,2,2-tetra-*p*-toluidino-thiourea (6a)

Yield 300 mg (24%), m.p. = 193–195°C, red crystals (benzene). ^{1}H NMR ($CDCl_3$): δ = 2.28–2.30 (m, 12H, 4CH_3), 6.18–7.45 (m, 32H, 4Ar-H, 2Ph-H, 6NH). ^{13}C NMR ($CDCl_3$): δ = 20.09, 20.18 (2CH_3-Ar) 40.18 (sym. ethane-C), 129.22, 129.30, 129.42, 129.65, 129.87, 130.30, 130.39, 131.44, 131.65, 131.98, 132.09, 132.15, 133.38, 133.42, 133.65, 133.87, 134.38, 134.50 (Ar-C, Ph-C). IR (KBr): $\bar{\nu}$ = 3340–3200 cm^{-1}(NH), 3080–3030 (Ar-CH), 2998–2890 (Aliph.-CH), 1610 (C=C). MS (70 eV) m/z (%) 632 (M^{+}, 48), 631 (M^{-1}, 100), 617 (74), 603 (42), 589 (24), 514 (22), 496 (10), 413 (14), 408 (12), 390 (10), 247 (18), 233 (14), 208 (6), 162 (8), 130 (8), 118 (22), 106 (10), 91 (30). Calcd. for $C_{42}H_{44}N_6$ (632.852): C, 79.71, H, 7.01, N, 13.28. Found: C, 79.60, H, 6.98, N,13.18.

1,2-Diphenylamino-1,1,2,2-tetra-*p*-chloroanilino-thiourea (6b)

Yield 340 mg (24%), m.p. = 210–212°C, red crystals (benzene). ^{1}H NMR ($CDCl_3$): $\delta = 6.20$–7.68 (m, 32H, 4Ar-H, 2Ph-H, 6NH). ^{13}C NMR ($CDCl_3$): $\delta = 42.50$ (sym. ethane-C), 129.03, 129.18, 129.22, 129.31, 129.40, 129.60, 130.02, 130.18, 131.31, 131.64, 131.85, 132.09, 133.18,133.30, 133.41, 133.60, 134.12, 134.44 (Ar-C, Ph-C). IR (KBr): $\nu^- = 3300$–3200 cm^{-1}(NH), 3065–3018 (Ar-CH), 1600 (C=C). MS (70 eV) m/z (%) 718 (M^{+4}, 18), 717 (M^{+3}, 32), 716 (M^{+2}, 60), 715 (M^{+1}, 78), 714 (M^+, 100), 713 (M^{-1}, 58), 580 (30), 579 (34), 578 (28), 453 (48), 452 (50), 451 (46), 429 (38), 428 (44), 426 (40), 359 (22), 358 (28), 357 (26), 316 (22), 230 (18),146(16), 128 (58). Calcd. for $C_{38}H_{32}Cl_4N_6$ (714.537): C, 63.88, H, 4.51, Cl, 19.85, N, 11.76. Found: C, 63.69, H, 4.40, Cl, 19.73, N, 11.65.

1 N-Phenyl-3N-*p*-toluidino-diazetidine-2,4-dithione (7a)

Yield 100 mg (18%), m.p. = 98–100°C green crystals (cyclohexane). ^{1}H NMR ($CDCl_3$): $\delta = 2.28$ (s, 3H, CH_3), 6.40–6.85 (m, 5H), 6.90–6.93 (dd, 2H, J= 10.18 Hz), 6.97–7.00 (dd, 2H, J= 10.80 Hz). ^{13}C NMR ($CDCl_3$): $\delta = 20.18$ (CH_3-Ar) 129.88, 130.09, 130.18, 130.37, 131.09, 131.42, 131.66, 131.94 132.28, 133.33, 134.46, 135.18 (Ar-C, Ph-C), 188.18, 188.60 (C=S). IR (KBr): $\nu^- = 3020$–3009 cm^{-1}(Ar-CH), 2990–2895 (Aliph.-CH), 1610 (C=C), 1580 (C=S). MS (70 eV) *m/z* (%) 284 (M^+, 88), 270 (100), 256 (20), 223 (12), 167 (10), 150 (60), 149 (62), 136 (38), 124 (16), 118 (50), 107 (43), 93 (22), 91 (50), 77 (42),71 (14). Calcd. for $C_{15}H_{12}N_2S_2$ (284.406): C, 63.35, H, 4.25, N, 9.85, S, 22.25. Found: C, 63.21, H, 4.21, N, 9.83, S, 22.13.

1N-Phenyl-3N-*p*-chlorophenyl-diazetidine-2,4-dithione (7b)

Yield 90 mg (16%),), m.p. = 120-121°C, dark green crystals (cyclohexane). ^{1}H NMR ($CDCl_3$): $\delta = 6.45$–6.94 (m, 5H), 7.00–7.03 (dd, 2H, J= 10.93 Hz), 7.05–7.08 (dd, 2H, J= 11.00 Hz). ^{13}C NMR ($CDCl_3$): $\delta = 130.11$, 130.51, 130.70, 131.28, 131.54, 132.18, 132.44, 132.52, 133.00, 134.60, 134.81, 136.00 (Ar-C, Ph-C). 190.89, 191.22 (C=S). IR (KBr): $\nu^- = 3030$–3010 cm^{-1}(Ar-CH), 1605 (C=C), 1585 (C=S). MS (70 eV) m/z (%) 305 (M^{+1}, 40), 304 (M^+ 100), 303 (M^{-1}, 38), 269 (48), 268

(52), 225 (26), 224 (24), 193 (18), 180 (20), 112 (14). Calcd. for $C_{14}H_9$ Cl N_2S_2 (304.827): C, 55.16, H, 2.98, Cl, 11.63, N, 9.19, S, 21.04. Found: C, 55.00, H, 2.90, Cl, 11.53, N, 9.09, S, 21.00.

1-Anilino-1,1,2,2-tetra-*p*-toluidino-ethane (8a)

Yield 250 mg (26%), m.p. = 171–172°C, red crystals (petroleum ether). ^{1}H NMR ($CDCl_3$): $\delta = 2.28$–2.33 (m, 12H, $4CH_3$), 5.52 (s, 1H, CH-ethane), 6.49–7.42 (m, 26H, 4Ar-H, Ph-H, 5NH). ^{13}C NMR ($CDCl_3$): $\delta = 20.00$, 20.09, 20.18, 20.30 ($4CH_3$-Ar), 45.09 (CH-ethane-C), 50.12 (C-ethane), 128.12, 128.31, 128.54, 128.63, 128.73, 128.85, 129.00, 129.09, 129.14, 129.32, 129.65, 129.72, 130.22, 130.38, 131.42, 131.65, 132.21, 132.34, 132.50, 132.53, 132.64, 132.88, 133.64, 133.71, 133.82, 133.93, 134.00, 134.18, 138.45, 138.60 (Ar-C, Ph-C). IR (KBr): v^{-} = 3335–3220 cm^{-1}(NH), 3065–3025 (Ar-CH), 2995–2890 (Aliph.-CH), 1604 (C=C). MS (70 eV) m/z (%) 541 (M^+ 100), 527 (20), 458 (16), 456 (12), 426 (20), 396 (18), 381 (26), 298 (38), 297 (62), 255 (14), 240 (42), 239 (46), 214 (18), 213 (56), 202 (48), 172 (78), 171 (24), 157 (16), 143 (14). Calcd. for $C_{36}H_{39}N_5$ (541.739): C, 79.82, H, 7.26, N, 12.93. Found: C, 79.64, H, 7.18, N, 12.80.

1-Anilino-1,1,2,2-tetra-*p*-chloroanilino-ethane (8b)

Yield 280 g (22%), m.p. = 193–195°C red crystals (petroleum ether). ^{1}H NMR ($CDCl_3$): $\delta = 5.42$ (s, 1H, CH-ethane), 6.35–7.38 (m, 26H, 4Ar-H, Ph-H, 5NH). ^{13}C NMR ($CDCl_3$): $\delta = 48.12$ (CH-ethane-C), 52.12 (C-ethane), 128.08, 128.36, 129.04, 129.18, 130.11, 130.38, 130.41, 130.63, 130.82, 130.94, 131.28, 131.42, 131.65, 131.84, 131.92, 131.65, 132.01, 132.08, 132.18, 132.24, 132.31, 132.43, 132.65, 133.01, 133.09, 133.12, 13332.00, 134.00, 134.18, 136.98 (Ar-C, Ph-C). IR (KBr): v^{-} = 3330–3210 cm^{-1}(NH), 3065–3025 (Ar-CH), 2995–2966 (Aliph.-CH), 1601 (C=C). MS (70 eV) m/z (%) 627 (M^4, 18), 626 (M^{+3}, 38), 625 (M^{+2}, 60), 624 (M^{+1}, 82), 623 (M^+, 100), 622 (M^{-1}, 62), 621 (M^{-2}, 32), 589 (32), 588 (36), 587 (30), 497 (22), 496 (28), 495 (28), 406 (26), 405 (18), 404 (22), 403 (18), 393 (62), 392 (48), 231 (44), 230 (48), 229 (42), 194 (18), 192 (22). Calcd. for $C_{32}H_{27}CI_4N_5$ (623.424): C, 61.65, H, 4.37, CI, 22.75, N, 11.23. Found: C, 61.48, H, 4.28, Cl, 22.68, N, 11.18.

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