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Electrochemical Synthesis and X-Ray Molecular Structure of 3,4,5-Triaryl-2-Aryliminooxazolines

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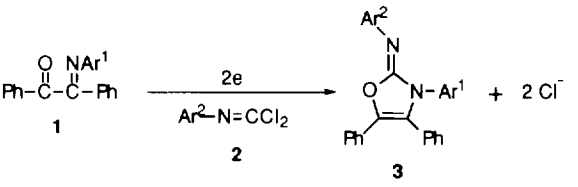
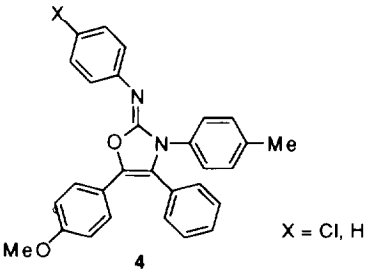
Abstract : *Selective cathodic reduction of benzilmonoimines in aprotic medium at a mercury pool cathode, under constant potential, in the presence of equimolecular amounts of N-arylcarbonimidoyl dichlorides, provides a new and very convenient method for the synthesis of 3,4,5-triaryl-2-aryliminooxazolines. Geometrical characteristics of this little known class of heterocycle were determined by X-ray crystallography of 2-(4-chlorophenylimino)-5-(4-methoxyphenyl)-3-(4-methylphenyl)-4-phenyloxazoline.*

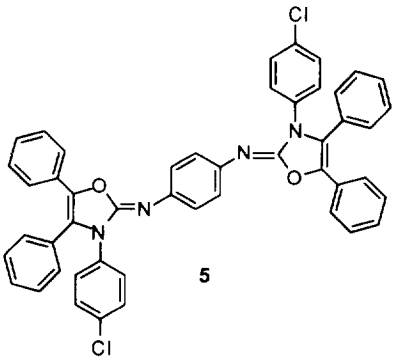
A few studies concerning the electrochemical reduction of monoimines of diaryl-1,2-diketones have been reported. Overall the reactions in protic media correspond to two-electron, two-proton processes, with primary formation of eneaminols, which later rearrange to α-aminoketones[1]. However, from a synthetic point of view, cathodic reductions in aprotic media in the presence of non-electroactive electrophilic reagents are of great interest. Thus, the attack of electrogenerated intermediates on the electrophilic reagents can take place instead of simple protonation. This type of electrolysis has been applied to diaryl-1,2-diketones, demonstrating substantial usefulness in synthesis[2-9]. Nevertheless, the electrochemical reduction in aprotic media of their imino derivatives has scarcely been investigated. Cathodic reduction of benzil monoanil has been carried out in the presence of methyl chloride and in the presence of methyl tosylate; the formation of a mixture of E and Z α-methoxy-α'-N-methylanilinostilbenes, and 2-hydroxy-2-phenylpropiophenoneanil has been reported[8]. The reduction leading to macrocyclic heterocycles has also been reported[9].

In order to explore the synthetic usefulness of N-arylcarbonimidoyl dichlorides **2** as agents to capture electrogenerated intermediates, the electrochemical reductions of monoimines **1** were carried out in the

presence of the reagents **2**.







Electrolysis of solutions of monoimines **1** in DMF-$LiClO_4$ at a mercury pool cathode, under constant potential were carried out in the presence of equimolecular amounts of carbonimidoyl dichlorides **2**. The electricity consumption was 2 F/mol of **1**. After electrolysis, crude solid reaction products were easily isolated by simple mixing of the catholyte solution with water. After crystallization the products were identified by IR, MS, high field NMR spectroscopy and microanalyses as the corresponding 3,4,5-triaryl-2-aryliminooxazolines **3**, which are a little-known class of oxazoline derivatives. Yields were high. The results of the electrochemical reductions are summarized in Table 1. The synthesis of product **3a** has been reported previously[10]. The rest of the entries correspond to new iminooxazolines.

***Table 1**. Preparation of 3,4,5-triaryl-2-aryliminooxazolines **3**, by cathodic reduction of monoimines **1** in the presence of N-arylcarbonimidoyl dichlorides **2**.*

Entry	Ar^1	Ar^2	Yield [a] (%)
3a	C_6H_5	C_6H_5	70
3b	4-MeO-C_6H_4	C_6H_5	85
3c	4-Me-C_6H_4	C_6H_5	80
3d	4-MeO-C_6H_4	4-Cl-C_6H_4	91
3e	4-Cl-C_6H_4	2-Cl-4-Me-C_6H_3	74
3f	4-Me-C_6H_4	2-Cl-4-Me-C_6H_3	73
3g	4-Cl-C_6H_4	C_6H_5	75
3h	C_6H_5	2-Cl-4-Me-C_6H_3	60
3i	4-Me-C_6H_4	2,4-Cl_2-C_6H_3	78
3j	4-Cl-C_6H_4	2,4-Cl_2-C_6H_3	72
3k	C_6H_5	4-Cl-C_6H_4	93
3l	4-Me-C_6H_4	4-Cl-C_6H_4	96
3m	4-MeO-C_6H_4	2,4-Cl_2-C_6H_3	69

[a] Yields of crystallized products.

In order to determine a crystallographic X-ray structure representative of this class of compound, products **4** were also obtained. Product **4a** provided suitable single crystals. The molecular structure found is illustrated in Figure 1. Selected intramolecular distances (crystallographic numbering of atoms) and selected bond angles are given in Table 2. The oxazoline ring is planar (mean deviation 0.01 Å), with C2-O1 1.361(3), C2-N3 1.365(3), N3-C4 1.418(3), O1-C5 1.403(3) and C4-C5 1.341(3) Å.

4-Chlorophenyliminobenzil was also reduced using 1,4-phenylene-bis(carbonimidoyl dichloride) as electrophilic reagent (ratio 2:1). The electrolysis afforded 2-(1,4-phenylenediimino)-bis[3-(4-chlorophenyl)-4,5-diphenyloxazoline] **5**, a previously unknown compound.

Carbonimidoyl dichlorides **2** themselves undergo electrochemical reduction giving isocyanides in almost quantitative yields[11]. However, the reduction potentials of monoimines **1** and carbonimidoyl dichlorides are different enough to allow the selective reduction of compounds **1**. This implies that reagents **2**

participate as non-electroactive electrophilic agents exclusively, being attacked by the negatively charged electrogenerated intermediates. Thus, both chlorine atoms undergo nucleophilic displacement closing the oxazoline ring.

The synthesis of product **3a** was previously reported in moderate yield by reaction of benzoylphenylacetaldehyde with N-phenylhydroxylamine[10]. 2-benzoyl-N,2-diphenylvinylideneamine was identified as an intermediate of the reaction process, and has also been used as starting material for the synthesis[12,13]. Cyanogen bromide has been used as an efficient cyclizing reagent of α-alkylaminophenones yielding trisubstituted 2-iminooxazolines whith unsubstituted imino groups[14-16]. N-arylation of these products has not been reported.

In comparison to previously known syntheses of iminooxazolines of type **3**, the present electrochemical process is far more convenient. Good yields, easy availability of starting materials and simple experimental procedure are noteworthy features of the new method.

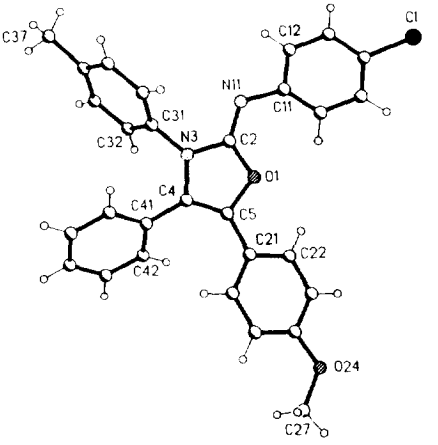


Figure 1. Molecular structure of **4a**, *showing the crystallographic numbering system used.*

Table 2. Selected Bond Lengths and Bond Angles in Crystal Structure of **4a**

Bond lenghts (Å)

Cl-C(14)	1.746(3)	O1-C(5)	1.403(3)
C(2)-N(3)	1.365(3)	N(3)-C(31)	1.437(3)
N(11)-C(11)	1.402(3)	O(24)-C(27)	1.424(3)
C(34)-C(37)	1.501(3)	O(1)-C(2)	1.361(3)
C(2)-N(11)	1.280(3)	N(3)-C(4)	1.418(3)
C(4)-C(5)	1.341(3)	C(5)-C(21)	1.457(3)
C(24)-O(24)	1.362(3)		

Bond angles (°)

C(2)-O(1)-C(5)	108.4(2)	N(11)-C(2)-N(3)	124.9(2)
C(2)-N(3)-C(4)	108.9(2)	C(4)-N(3)-C(31)	127.3(2)
C(5)-C(4)-C(41)	131.9(2)	C(4)-C(5)-O(1)	108.8(2)
O(1)-C(5)-C(21)	114.4(2)	C(42)-C(41)-C(4)	120.5(2)
C(22)-C(21)-C(5)	119.7(2)	C(32)-C(31)-N(3)	119.8(2)
N(11)-C(2)-O(1)	127.7(2)	C(2)-N(11)-C(11)	123.2(2)
O(1)-C(2)-N(3)	107.3(2)	C(2)-N(3)-C(31)	122.8(2)
C(5)-C(4)-N(3)	106.5(2)	N(3)-C(4)-C(41)	121.6(2)
C(4)-C(5)-C(21)	136.7(2)		

EXPERIMENTAL

Benzilmonoimines **1** were prepared by standard procedures[17,18], as were the carbonimidoyl dichlorides **2**[19]. DMF was taken from a freshly opened bottle and dried with molecular sieve. $LiClO_4$ was anhydrous. Both were purchased from Fluka, and were used directly without further purification. NMR spectra were determined on a 200 MHz spectrometer in $CDCl_3$ with TMS as internal reference. EI MS were obtained with a direct insertion probe and an ionizing voltage of 70 eV. All melting points were determined on a hot-plate melting point apparatus and are uncorrected. Electrochemical experiments were performed with a preparative potentiostat (200 V, 1 A) coupled to a current integrator.

X-Ray Crystallographic Analysis of 4a.

The crystals used for the X-ray study were obtained from acetonitrile - methylene dichloride. *Crystal data:* $C_{29}H_{23}ClN_2O_2$, Mr = 466.94, monoclinic, $P2_1/c$, a = 10.399 (2), b = 26.898 (4), c = 8.4583 (14) Å, β = 95.965 (12) °, V = 2354.2 $Å^3$, Z = 4, λ (Mo Kα) = 0.71073 Å, T = -130 °C. *Data collection:* Colourless plate 0.8 x 0.7 x 0.15 mm, Stoe STADI-4 diffractometer, 4745 intensities (4147 unique), $2\theta_{max}$ 50°. *Structure solution and refinement:* Direct methods, refined on F^2 (program SHELXL-93, G.M. Sheldrick, University of Göttingen), H atoms with riding model, wR (F^2) 0.134, R (F) 0.051, 310 parameters, S = 1.04, max. $\Delta\rho$ 0.58 e $Å^{-3}$.

Full details of the structure determination have been deposited at the Fachinformationszentrum Karlsruhe, Gesellschaft für Wissenschaftlich-technische Information mbH, D-76344 Eggenstein-Leopoldshafen, Germany. Any request for this material should quote a full literature citation and the reference number CDS - 401719.

General Electrolysis Procedure.

Preparative electrolyses were carried out under a constant cathodic potential in a concentric cylindrical cell with two compartments separated by a circular glass frit (medium) diaphragm. A mercury pool (diameter 5 cm) was used as the cathode and a platinum plate as the anode. The catholyte was magnetically stirred. The temperature was kept at approximately 18 °C by external cooling. The reductions were performed out in DMF-$LiClO_4$, 0.2 M. Approximately 35 mL and 15 mL of this solution were placed in the cathodic and the anodic compartments, respectively. Anhydrous sodium carbonate (3 g) was placed in the anode compartment to prevent accumulation of electrogenerated acid. Solutions of monoimines **1** (5 mmol) and carbonimidoyl dichlorides **2** (5 mmol) were electrolyzed under the following cathodic potentials: -1.05 V vs SCE (entries **3a**, **3c**, **3e**, **3g**, **3i**); -1.10 V (entries **3d**, **3f**, **3h**, **3m**); -1.15 V (entries **3j**, **3k**, **3l**); -1.20 V (entries **3b**, **4a**, **4b**). The electricity consumption was 2 F/mol in all cases. Cathodic reduction of N-(4-chlorophenyl)-4-methoxybenzoylbenzylideneamine (5 mmol) in the presence of 1,4-phenylene-bis(carbonimidoyl dichloride) (2.5 mmol) (-1.05 V) gave product **5**. All electrolysis products were isolated by transferring the catholyte solution dropwise into cold brine (200 mL) and filtering. The isolated solid were washed with 5mL of very cold acetonitrile and crystallized from acetonitrile or methanol. Product **5** was crystallyzed from boiling DMF.

3,4,5-Triphenyl-2-phenyliminooxazoline (**3a**)

White needles mp 181-183 °C. (Lit.[10], mp 179-180 °C). (Found: C, 83.82; H, 5.18; N, 7.18. $C_{27}H_{20}N_2O$ requires: C, 83.48; H, 5.19; N, 7.21); ^{1}H n.m.r. δ: 6.99 (m, 1H), 7.14 - 7.39 (m, 19 H); ^{13}C n.m.r. δ: 122.06, 123.48, 124.39, 124.57, 127.34, 127.50, 127.60, 127.90, 127.99, 128.48, 128.64, 128.79, 128.96, 129.34, 130.40, 134.90, 135.53, 146.76, 148.94; m.s., m/z (%): 388 (M^+, 88), 180 (47), 77 (100); i.r.(Nujol): 1707, 1688, 1657, 1591, 1499, 1372, 762, 710, 691 cm^{-1}.

3-(4-Methoxyphenyl)-4,5-diphenyl-2-phenyliminooxazoline (**3b**)

Pale yellow needles mp 172-174 °C. (Found: C, 80.57 ; H, 5.28; N, 6.70. $C_{28}H_{22}N_2O_2$ requires: C, 80.36; H, 5.30; N, 6.69); ^{1}H n.m.r. δ: 3.72 (s, 3H), 6.78 (d, 2H, J = 8.9 Hz), 6.98 (m, 1H), 7.13 - 7.25 (m, 7 H), 7.28 - 7.33 (m, 9H); ^{13}C n.m.r. δ: 55.36, 114.20, 121.97, 123.50, 124.46, 124.71, 127.52, 127.57, 128.07, 128.48, 128.63, 128.97, 129.22, 129.33, 130.45, 135.27, 146.90, 149.40, 158.60; m.s., m/z (%): 418 (M^+, 54), 210 (100), 77 (97); i.r.(Nujol): 1673, 1651, 1588, 1513, 1252, 987, 832, 770, 693 cm^{-1}.

3-(4-Methylphenyl)-4,5-diphenyl-2-phenyliminooxazoline (**3c**)

White needles mp 183-184 °C. (Found: C, 83.64; H, 5.50; N, 6.98. $C_{28}H_{22}N_2O$ requires: C, 83.56; H,

5.51; N, 6.96); ^{1}H n.m.r. δ: 2.26 (s, 3H), 7.03-7.30 (m, 19H); ^{13}C n.m.r. δ: 21.13, 121.93, 123.48, 124.48, 127.49, 127.55, 127.66, 128.04, 128.44, 128.57, 128.91, 129.28, 129.48, 130.40, 132.21, 135.33, 137.21, 146.90, 149.14; m.s., m/z (%): 402 (M^+, 58), 194 (100), 91 (57), 77 (40); i.r.(Nujol): 1705, 1671, 1593, 1516, 1377, 822, 762, 720, 700, 694 cm^{-1}.

2-(4-Chlorophenylimino)-3-(4-methoxyphenyl)-4,5-diphenyloxazoline (**3d**)

Pale yellow needles mp 170-172 °C. (Found: C, 73.99; H, 4.65; N, 6.19. $C_{28}H_{21}ClN_2O_2$ requires: C, 74.25; H, 4.67 N, 6.18); ^{1}H n.m.r. δ: 3.70 (s, 3H), 6.76 (d, 2H, J = 9.0 Hz), 7.09 - 7.31 (m, 16H); ^{13}C n.m.r. δ: 55.38, 114.29, 124.54, 124.77, 124.84, 126.73, 127.48, 127. 57, 127.68, 128.40, 128.57, 128.99, 129.23, 129.42, 130.50, 135.55, 145.76, 149.63, 158.83; m.s., m/z (%): 452 (M^+, 38), 210 (100), 77 (81); i.r.(Nujol): 1689, 1651, 1514, 1383, 1247, 1029, 835, 693 cm^{-1}.

3-(4-Chlorophenyl)-2-(2-chloro-4-methylphenylimino)-4,5-diphenyloxazoline (**3e**)

White needles mp 193-195 °C. (Found: C, 71.57; H, 4.28; N, 5.95. $C_{28}H_{20}Cl_2N_2O$ requires: C, 71.34; H, 4.28; N, 5.94.); ^{1}H n.m.r. δ: 2.30 (s, 3H), 7.02 (dd, 1H, J = 8.1 Hz, J = 1.5 Hz), 7.19-7.39 (m, 16H); ^{13}C n.m.r. δ: 20.68, 123.78, 124.00, 124.77, 127.08, 127.64, 127.80, 128.51, 128.78, 129.03, 129.22, 129.67, 130.01, 130.42, 132.95, 133.00, 136.06, 141.16, 148.91; m.s., m/z (%): 470 (M^+, 2) 216 (34), 214 (100); i.r.(Nujol): 1701, 1666, 1493, 1378, 1242, 1065, 820, 692 cm^{-1}.

2-(2-Chloro-4-methylphenylimino)-3-(4-methylphenyl)-4,5-diphenyloxazoline (**3f**)

White needles mp 174-175 °C. (Found: C, 76.98; H, 5.12; N, 6.22. $C_{29}H_{23}ClN_2O$ requires: C, 77.24; H, 5.14; N, 6.21); ^{1}H n.m.r. δ: 2.26 (s, 3H), 2.28 (s, 3H), 6.97 - 7.33 (m, 17H); ^{13}C n.m.r. δ: 20.61, 21.12, 123.89, 124.55, 124.61, 127.44, 127.54, 127.64, 127.82, 127.93, 128.39, 128.92, 129.32, 129.48, 129.87, 130.43, 132.05, 132.46, 135.56, 137.24, 141.82, 149.41; m.s., m/z (%): 450 (M^+, 94), 194 (100), 91 (54), 77 (33); i.r.(Nujol): 1705, 1668, 1516, 1489, 1377, 1242, 1061, 818, 762, 693 cm^{-1}.

3-(4-Chlorophenyl)-4,5-diphenyl-2-phenyliminooxazoline (**3g**)

White plates mp 194-196 °C. (Found: C, 76.90; H, 4.52; N, 6.65. $C_{27}H_{19}ClN_2O$ requires: C, 76.68; H, 4.53; N, 6.62); ^{1}H n.m.r. δ: 7.01 (m, 1H), 7.16 - 7.27 (m, 10H), 7.29 - 7.37 (m, 8H); ^{13}C n.m.r. δ: 122.32, 123.41, 123.91, 124.62, 127.15, 127.73, 127.81, 128.51, 128.70, 128.97, 129.00, 129.17, 129.58, 130.34, 132.93, 133.38, 135.81, 146.26, 148.60; m.s., m/z (%): 422 (M^+, 100), 121 (33), 214 (16), 77 (22); i.r.(Nujol): 1683, 1650, 1588, 1492, 1381, 1056, 988, 768, 725, 691 cm^{-1}.

2-(2-Chloro-4-methylphenylimino)-3,4,5-triphenyloxazoline (**3h**)

White needles mp 162-163 °C. (Found: C, 77.21; H, 4.86; N, 6.40. $C_{28}H_{21}ClN_2O$ requires: C, 76.97; H, 4.84; N, 6.41); ^{1}H n.m.r. δ: 2.28 (s, 3H), 7.00 (dd, 1H, J = 8.1 Hz, J = 1.5 Hz), 7.16 - 7.35 (m, 17H); ^{13}C n.m.r. δ: 20.62, 123.86, 124.42, 124.68, 127.34, 127.37, 127.64, 127.68, 127.85, 128.42, 128.78, 128.95, 129.38, 129.91, 130.42, 132.60, 134.70, 135.73, 141.65, 149.20; m.s., m/z (%): 436 (M^+, 100), 435 (45), 180 (25), 77 (43); i.r.(Nujol): 1693, 1659, 1595, 1493, 1387, 1249, 758, 689 cm^{-1}.

2-(2,4-Dichlorophenylimino)-3-(4-methylphenyl)-4,5-diphenyloxazoline (**3i**)

Pale yellow needles mp 169-171 °C. (Found: C, 71.29; H, 4.29; N, 5.92. $C_{28}H_{20}Cl_2N_2O$ requires: C, 71.34; H, 4.28; N, 5.94); ^{1}H n.m.r. δ: 2.27 (s, 3H), 7.06-7.38 (m, 17H); ^{13}C n.m.r. δ: 21.19, 124. 65, 124.66, 124.91, 126.91, 127.11, 127.21, 127.46, 127.75, 127.82, 128.54, 129.02, 129.18, 129.50, 129.61, 130.45, 131.81, 135.83, 137.61, 143.52, 149.78; m.s., m/z (%): 470 (M^+, 6), 194 (66), 165 (27), 105 (33), 91 (98), 77 (87), 65 (100); i.r.(Nujol) 1692, 1660, 1377, 1051, 985, 818, 785, 694 cm^{-1}.

3-(4-Chlorophenyl)-2-(2,4-dichlorophenylimino)-4,5-diphenyloxazoline (**3j**)

Pale yellow needles mp 198-200 °C. (Found: C, 65.99; H 3.47; N, 5.71. $C_{27}H_{17}Cl_3N_2O$ requires: C, 65.94; H, 3.48; N, 5.70); ^{1}H n.m.r. δ: 7.15-7.40 (m, 17H); ^{13}C n.m.r. δ: 124.07, 124.72, 124.77, 126.87, 127.22, 127.27, 127.46, 128.10, 128.61, 128.78, 129.06, 129.12, 129.28, 129.30, 129.81, 130.41, 133.05, 133.28, 136.26, 142.96, 149.23; m.s., m/z (%): 492 (M^+ + 2, 21), 490 (M^+, 27), 214 (72), 165 (42), 111 (62), 77 (100), 75 (57); i.r.(Nujol): 1699, 1666, 1496, 1471, 1378, 1094, 1063, 829, 756, 694 cm^{-1}.

2-(4-Chlorophenylimino)-3,4,5-triphenyloxazoline (**3k**)

Pale yellow needles mp 177-179 °C. (Found: C, 76.43; H 4.52; N, 6.64. $C_{27}H_{19}ClN_2O$ requires: C, 76.68; H, 4.53; N, 6.62); ^{1}H n.m.r. δ: 7.19-7.33 (m, 19H); ^{13}C n.m.r. δ: 124.41, 124.51, 124.71, 126.76, 127.23, 127.49, 127.73, 127.78, 127.85, 128.52, 128.56, 128.82, 128.97, 129.41, 130.32, 134.63, 135.63, 145.44, 149.12; m.s., m/z (%): 422 (M^+, 2), 180 (28), 77 (100), 51 (36); i.r.(Nujol): 1681, 1651, 1580, 1499, 1486, 1448, 1376, 761, 724, 685 cm^{-1}.

2-(4-Chlorophenylimino)-3-(4-methylphenyl)-4,5-diphenyloxazoline (**3l**)

White needles mp 168-169 °C. (Found: C, 76.63; H 4.85; N, 6.39. $C_{28}H_{21}ClN_2O$ requires: C, 76.97; H, 4.84; N, 6.41); ^{1}H n.m.r. δ: 2.27 (s, 3H), 7.08 (br s, 4H), 7.20-7.72 (m, 14H) ; ^{13}C n.m.r. δ: 21.17, 124.53,

124.63, 124.78, 126.72, 127.40, 127.71, 127.94, 128.58, 129.01, 129.43, 129.59, 130.44, 132.06, 135.56, 137.50, 145.66, 149.50 ; m.s., m/z (%): 438 (M^+ + 2, 20), 436 (M^+, 47), 194 (100), 165 (25), 91 (84), 77 (43), 65 (52) i.r.(Nujol): 1703, 1668, 1514, 1487, 1376, 1243, 1057, 820, 767, 698 cm^{-1}.

2-(2,4-Dichlorophenylimino)-3-(4-methoxyphenyl)-4,5-diphenyloxazoline (**3m**)

White needles mp 142-143 °C. (Found: C, 68.93; H 4.15; N, 5.78. $C_{28}H_{20}Cl_2N_2O_2$ requires: C, 69.00; H, 4.14; N, 5.75); ^1H n.m.r. δ: 3.73 (s, 3H), 6.80 (d, 2H, J = 8.9 Hz), 7.17-7.38 (m, 15H); ^{13}C n.m.r. δ: 55.40, 114.29, 124.62, 124.82, 124.91, 126.91, 127.11, 127.19, 127.77, 127.79, 128.53, 128.98, 129.03, 129.19, 129.51, 130.48, 135.76, 143.56, 149.96, 158.80; m.s., m/z (%): 488 (M^+ + 2, 13), 486 (M^+, 21), 210 (100), 165 (28), 105 (16), 92 (38), 77 (100); i.r.(Nujol): 1698, 1666, 1514, 1504, 1250, 1027, 830, 756, 698 cm^{-1}.

2-(4-Chlorophenylimino)-5-(4-methoxyphenyl)-3-(4-methylphenyl)-4-phenyloxazoline (**4a**)

(86%), colourless prisms mp 179-181 °C. (Found: C, 74.59; H 4.97; N, 5.98. $C_{29}H_{23}ClN_2O_2$ requires: C, 74.59; H, 4.96; N, 6.00); ^1H n.m.r. δ: 2.26 (s, 3H), 3.73 (s, 3H), 6.76 (d, 2H, J = 8.9 Hz), 7.07 (br s, 4H), 7.20-7.31 (m, 11H); ^{13}C n.m.r. δ: 21.14, 55.25, 114.08, 120.55, 122.93, 124.76, 126.19, 126.54, 127.54, 127.63, 128.53, 128.91, 129.17, 129.53, 130.39, 132.21, 137.70, 137.32, 145.77, 149.58, 159.23; m.s., m/z (%): 466 (M^+, 41), 194 (100), 91 (63), 65 (46); i.r.(Nujol): 1674, 1647, 1586, 1512, 1487, 1379, 1258, 1028, 830 cm^{-1}.

5-(4-Methoxyphenyl)-3-(4-methylphenyl)-4-phenyl-2-phenyliminooxazoline (**4b**)

(68%), white needles mp 197-198 °C. (Found: C, 80.79; H, 5.60; N, 6.49. $C_{29}H_{24}N_2O_2$ requires: C, 80.53; H, 5.59; N, 6.48); ^1H n.m.r. δ: 2.27 (s, 3H), 3.73 (s, 3H), 6.75 (d, 2H, J = 8.9 Hz), 6.97 (s, 1H) 7.07 (d, 2H, J = 8.5 Hz), 7.09 (d, 2H. J = 8.5), 7.21-7.31 (m, 13H); ^{13}C n.m.r. δ: 21.16, 55.26, 114.03, 120.78, 121.84, 122.90, 123.53, 126.21, 127.66, 127.81, 128.59, 128.89, 129.06, 129.48, 130.45, 132.47, 135.53, 137.09, 147.10, 149.30, 159.14; m.s., m/z (%): 432 (M^+, 5), 194 (53), 91 (100), 77 (66), 65 (72); i.r.(Nujol): 1682, 1650, 1587, 1514, 1379, 1258, 1026, 702, 692 cm^{-1}.

2-(1,4-Phenylenediimino)-bis[3-(4-chlorophenyl)-4,5-diphenyloxazoline] (**5**)

(83%), white needles mp > 320 °C. (Found: C, 75.35 ; H, 4.19; N, 7.33. $C_{48}H_{32}Cl_2N_4O_2$ requires: C, 75.10; H, 4.20; N, 7.30); n.m.r. spectra could not be recorded due to its extremely low solubility in the usual spectroscopic solvents; EI MS: M^+ absent, FAB M^+ = 766; i.r.(Nujol): 1689, 1658, 1590, 1495, 1374, 1065, 827, 693 cm^{-1}.

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