

Supplementary Information:

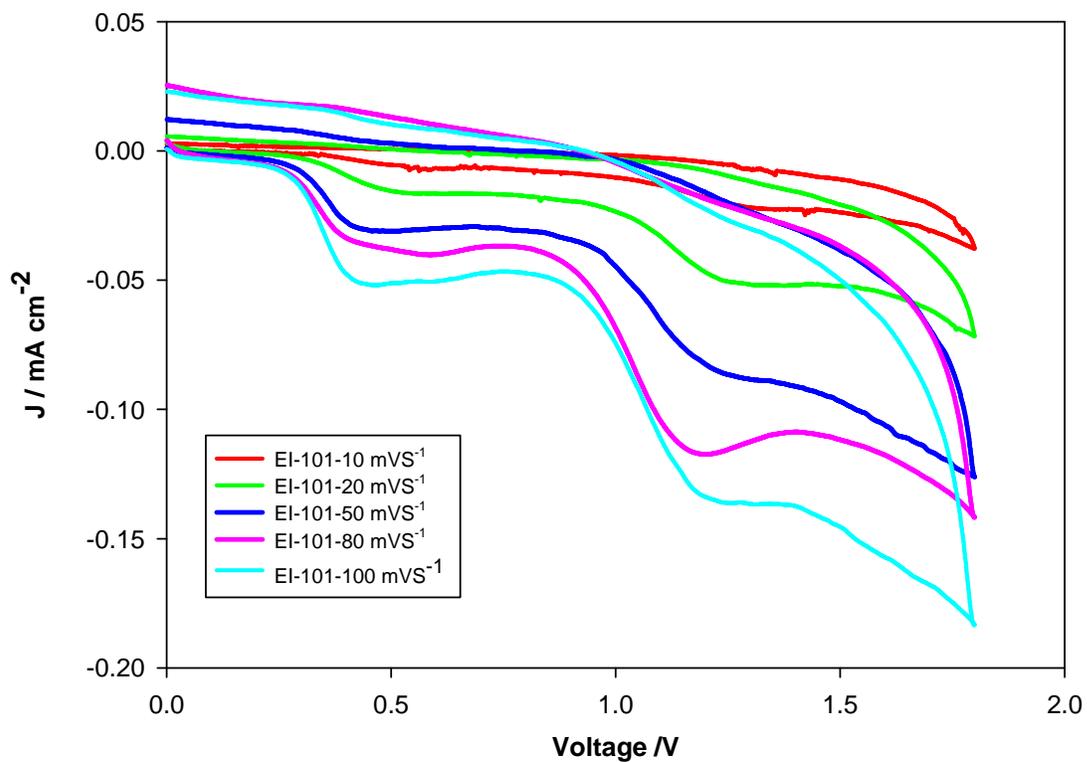


Fig S. 1. Cyclic voltammetry. Analyte: 1 mM of EI-101 (LiQ). Supporting electrolyte: tetrabutylammonium tetrafluoroborate., Working electrode: Pt., Reference electrode: Ag/AgCl.

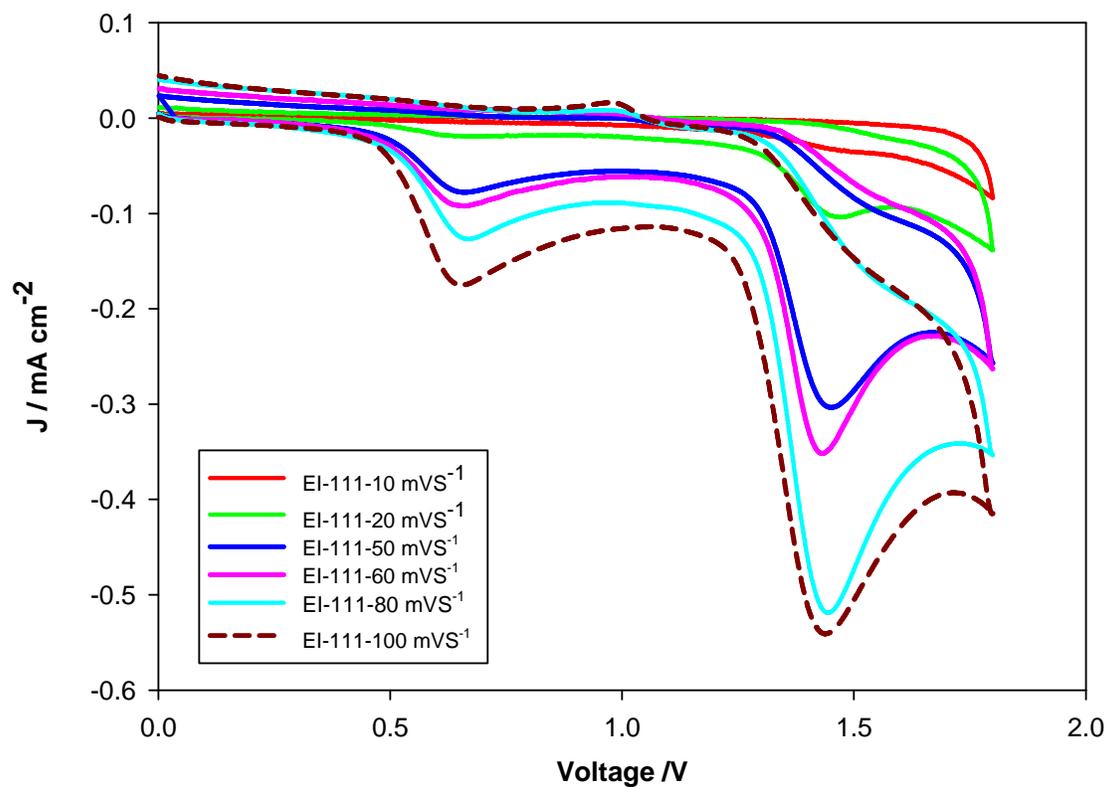


Fig. S.2. Cyclic voltammetry. Analyte: 1 mM of EI-111. Supporting electrolyte: tetrabutylammonium tetrafluoroborate., Working electrode: Pt., Reference electrode: Ag/AgCl.

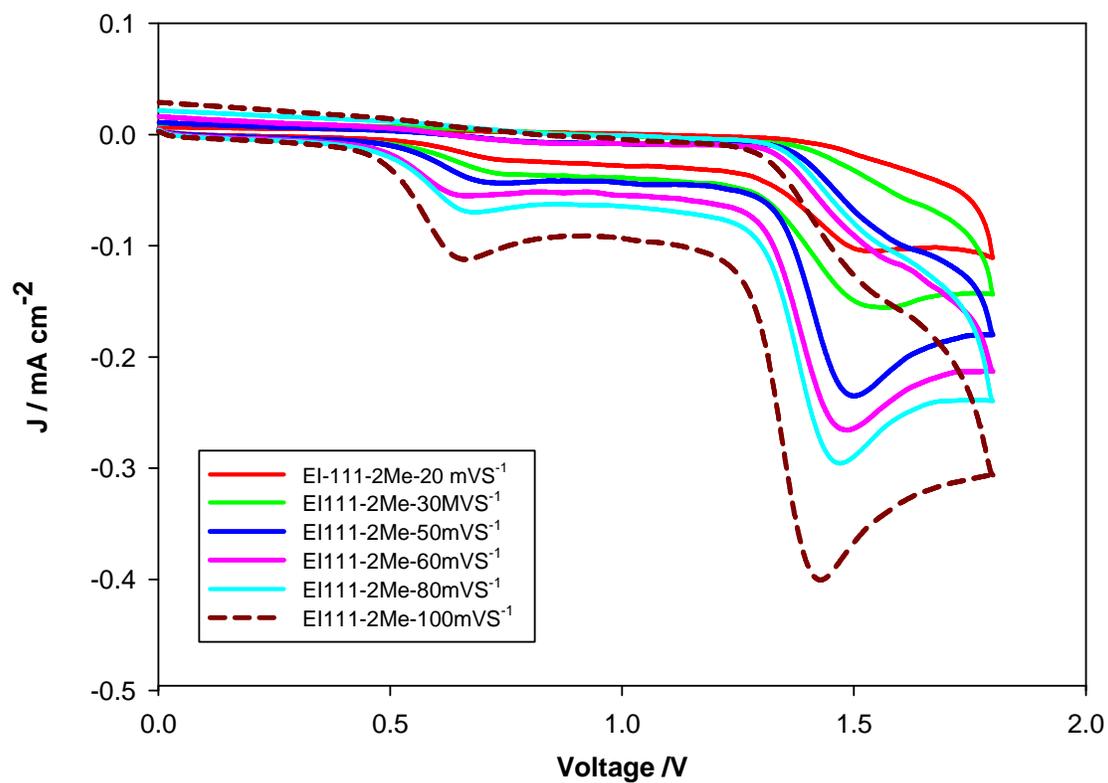


Fig. S.3. Cyclic voltammetry.. Analyte: 1 mM of EI-111-2 Me. Supporting electrolyte: tetrabutylammonium tetrafluoroborate., Working electrode: Pt., Reference electrode: Ag/AgCl.

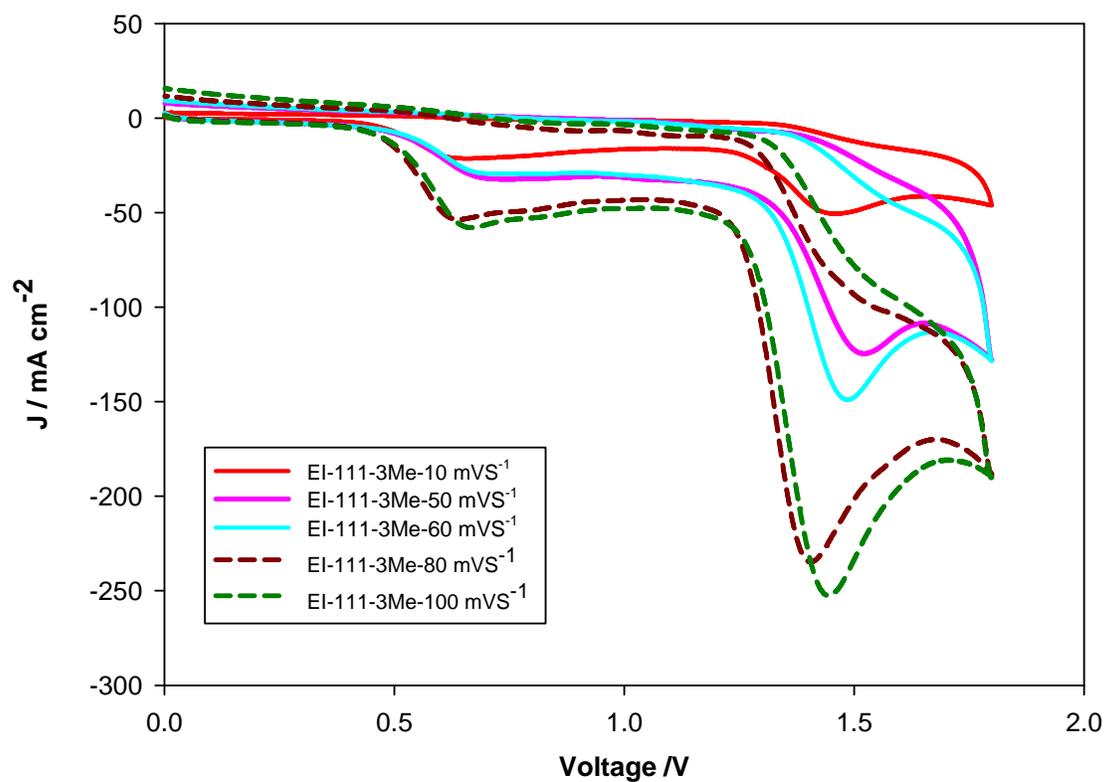


Fig . S4. Cyclic voltammetry . Analyte: 1 mM of EI-111-3 Me. Supporting electrolyte: tetrabutylammonium tetrafluoroborate., Working electrode: Pt., Reference electrode: Ag/AgCl.

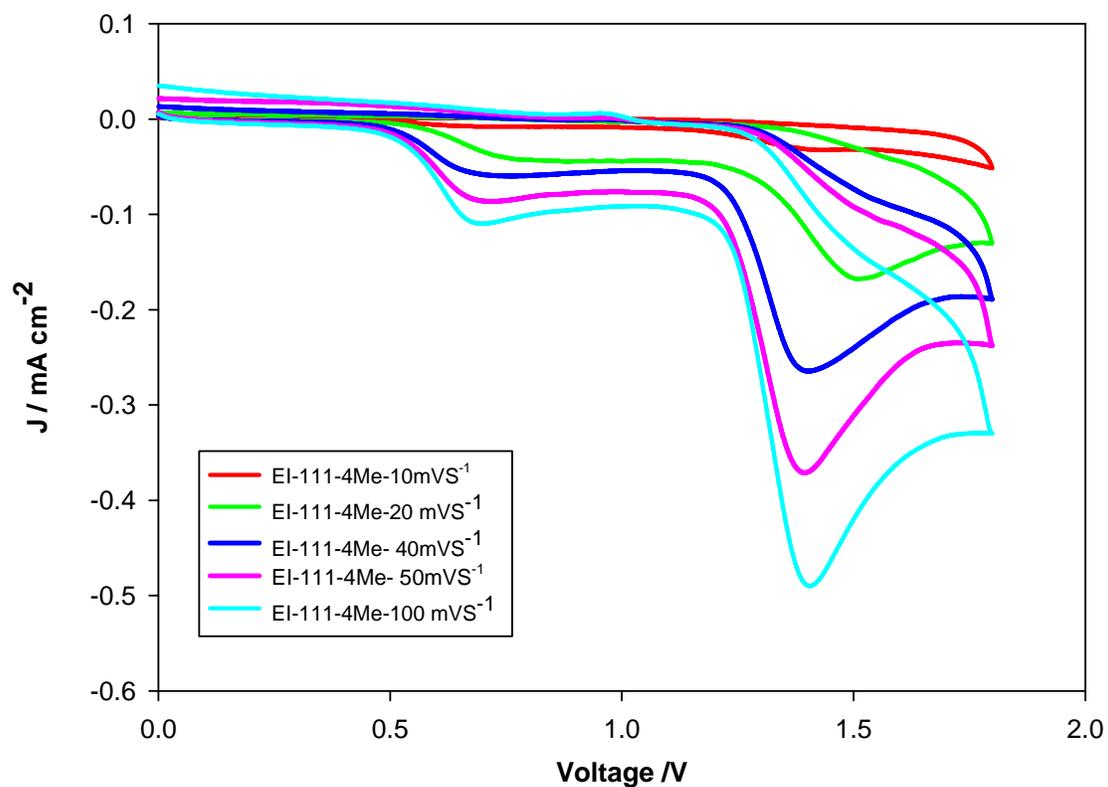


Fig. S 5. Cyclic voltammetry at 100 mVs^{-1} . Analyte: 1 mM of EI-101 (LiQ), EI-111, EI-111-2 Me, EI-111-3 Me, EI-111-4 Me. Supporting electrolyte: tetrabutylammonium tetrafluoroborate., Working electrode: Pt., Reference electrode: Ag/AgCl.

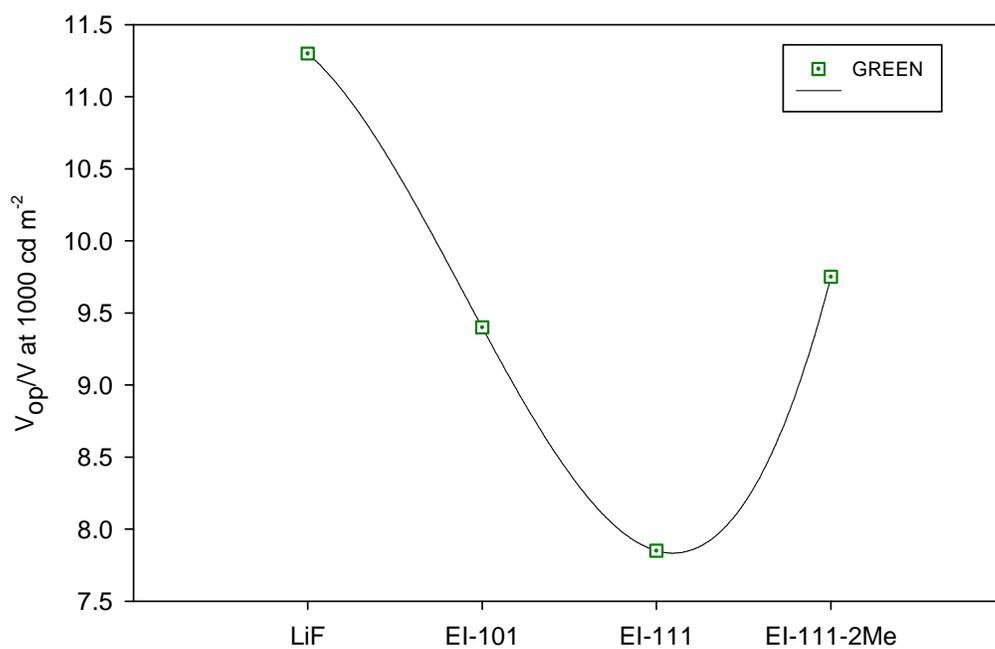


Fig S. 6. Operating voltage at 1000 cd m^{-2} for ITO/ZnTPTP (20 nm)/ α -NPB (50 nm)/Alq₃:DPQA (40: 0.1 nm)/Zr_q₄ (20 nm)/EI (1nm)/Al where EI: LiF, EI-101 (LiQ), EI-111, EI-111-2 Me.

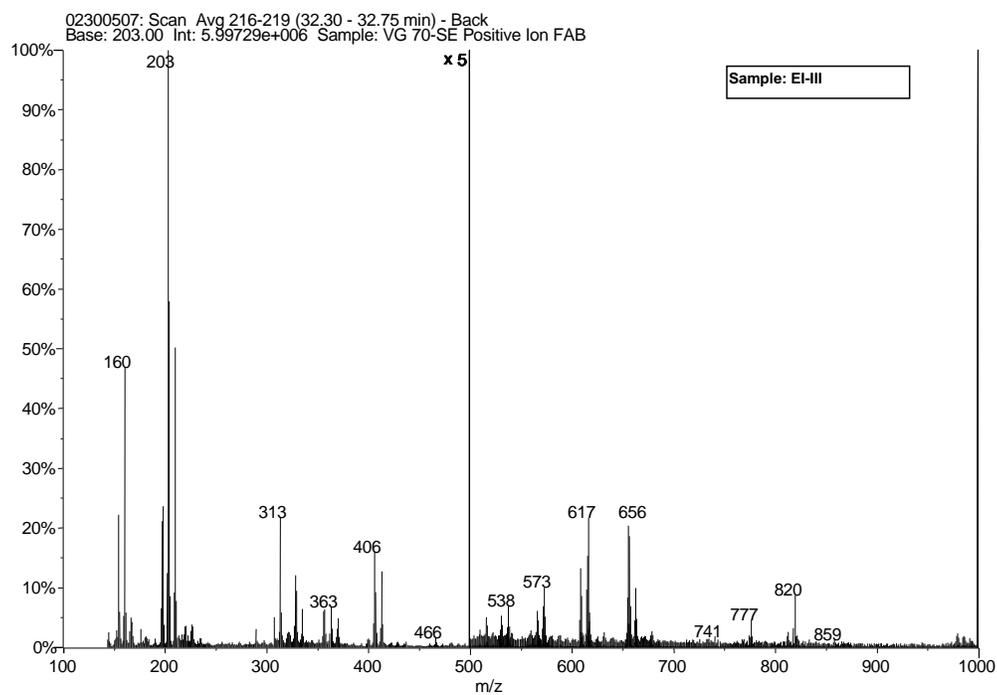


Fig. S7. FAB mass spectrometry of EI-111

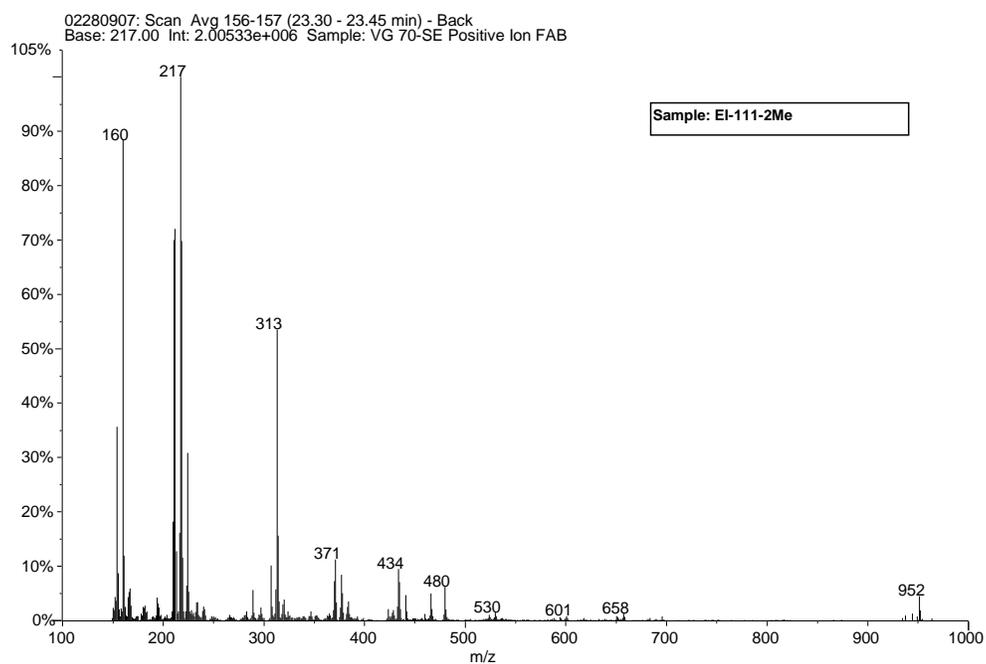


Fig. S 8. FAB mass spectrometry of EI-111-2Me

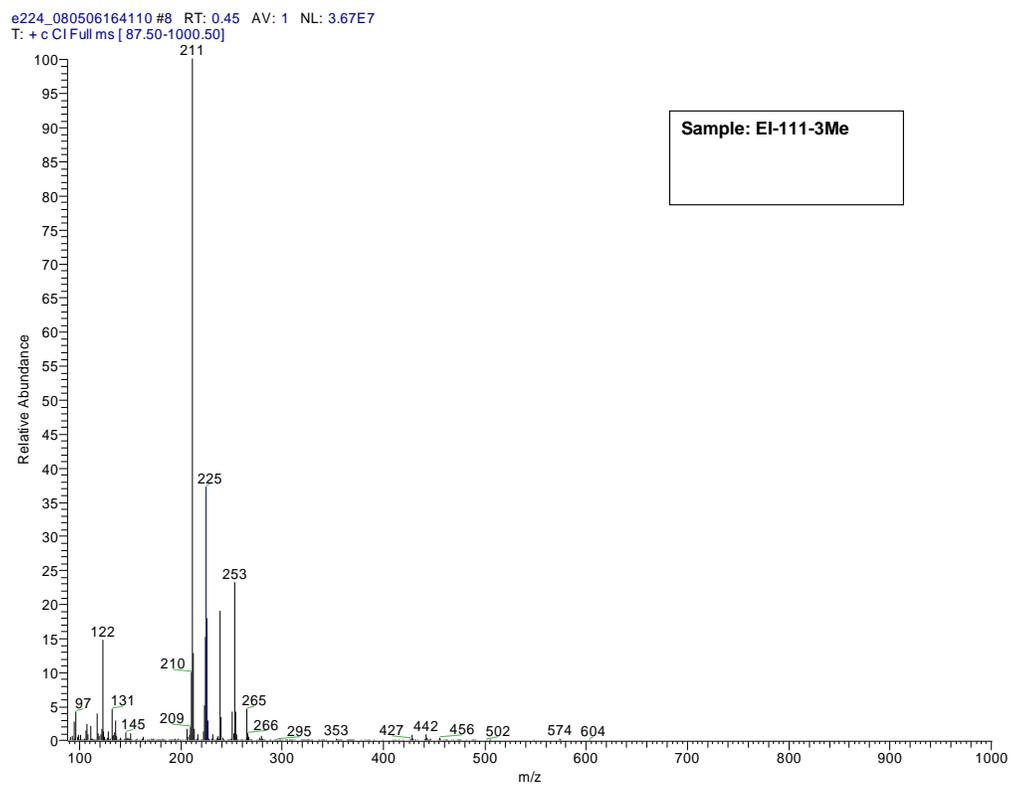


Fig. S 9. FAB mass spectrometry of EI-111-3Me

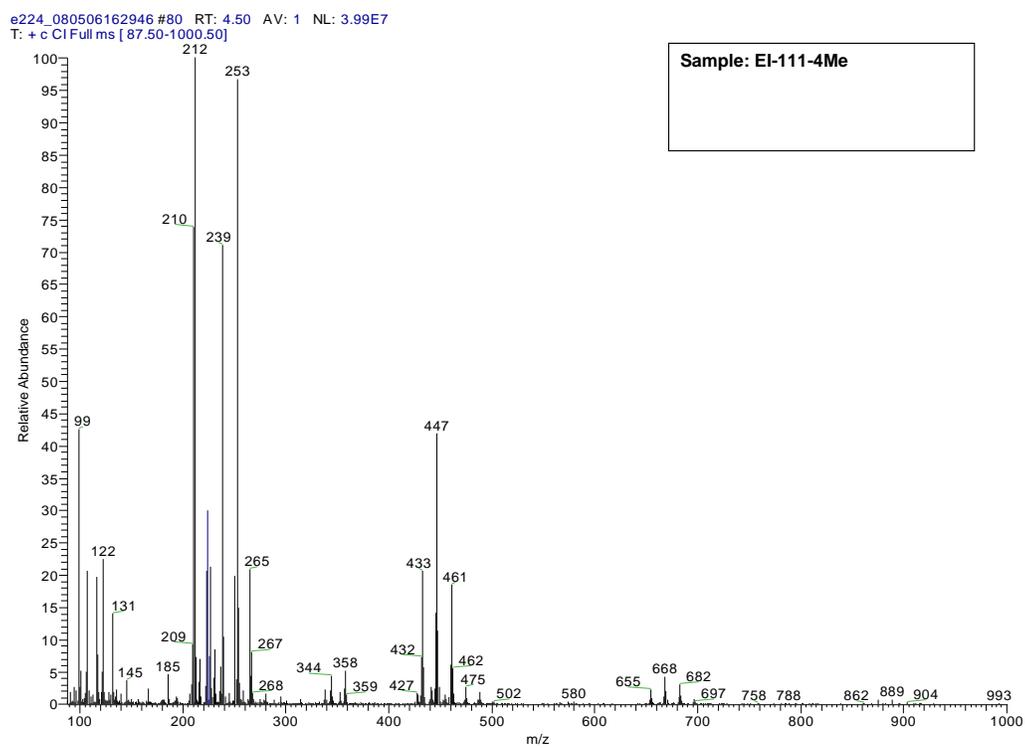


Fig. S 10. FAB mass spectrometry of EI-111-4Me

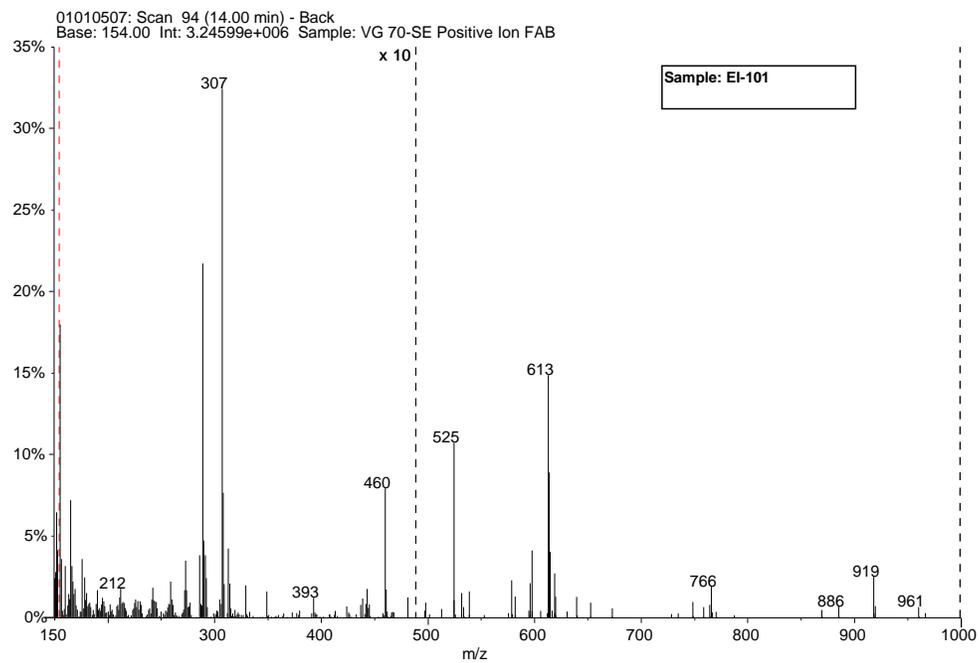


Fig. S 11. FAB mass spectrometry of EI-101

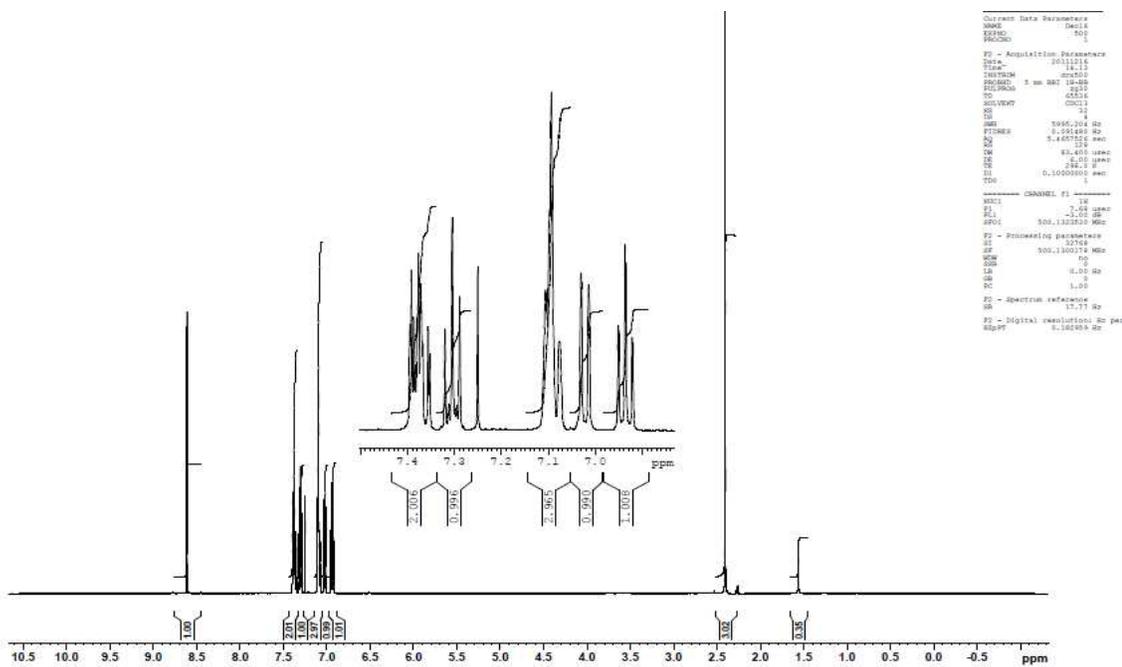


Fig. S14. ¹H NMR (500MHz, CDCl₃) of EI-111-3Me

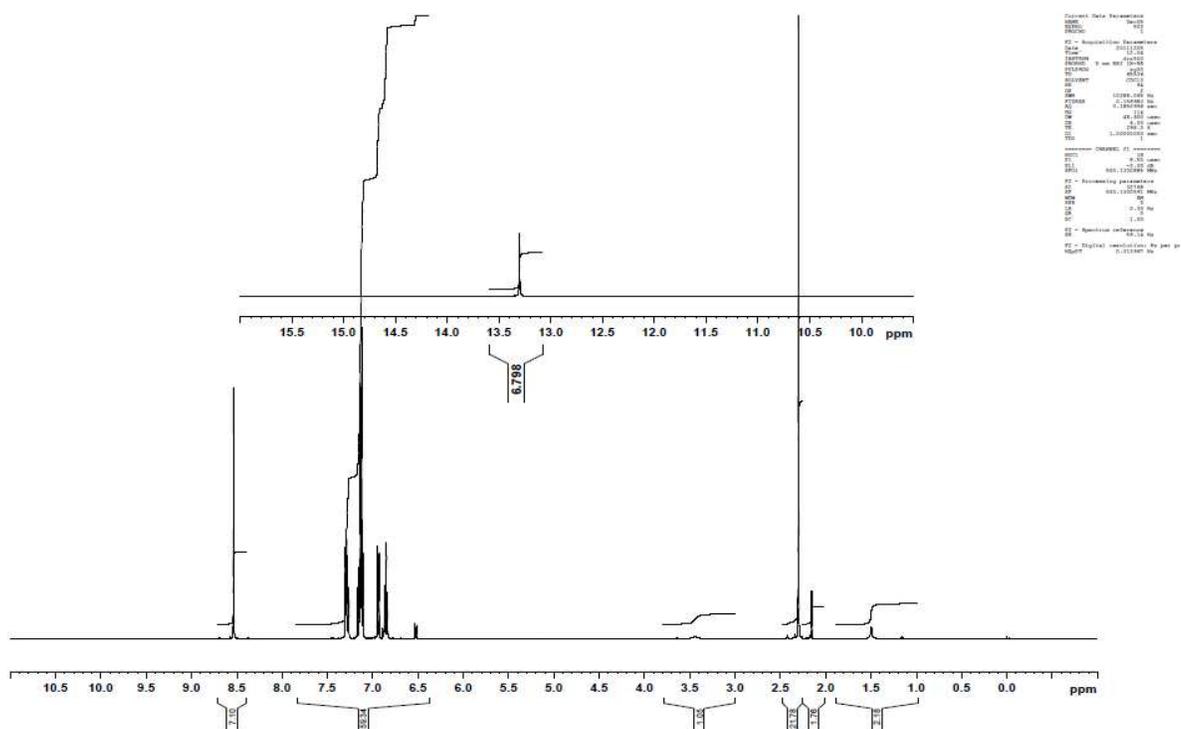


Fig. S 15. ¹H NMR (500MHz, CDCl₃) of EI-111-4Me

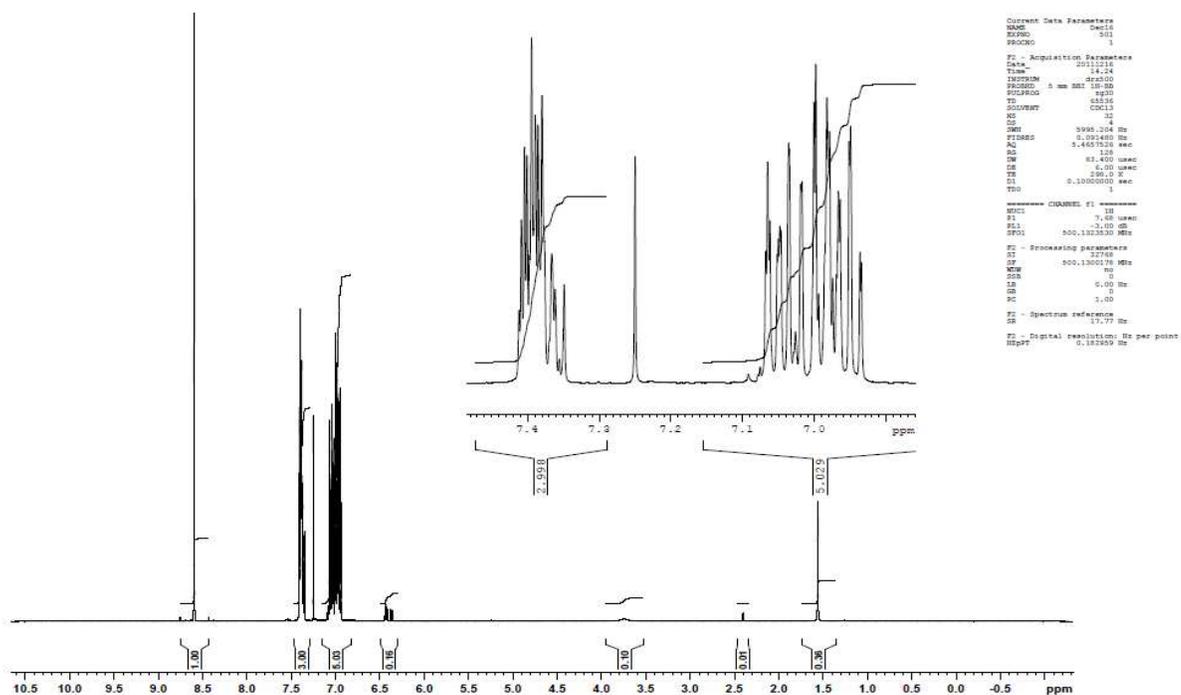


Fig. S 17. ¹H NMR (500MHz, CDCl₃) of EI-111-3F

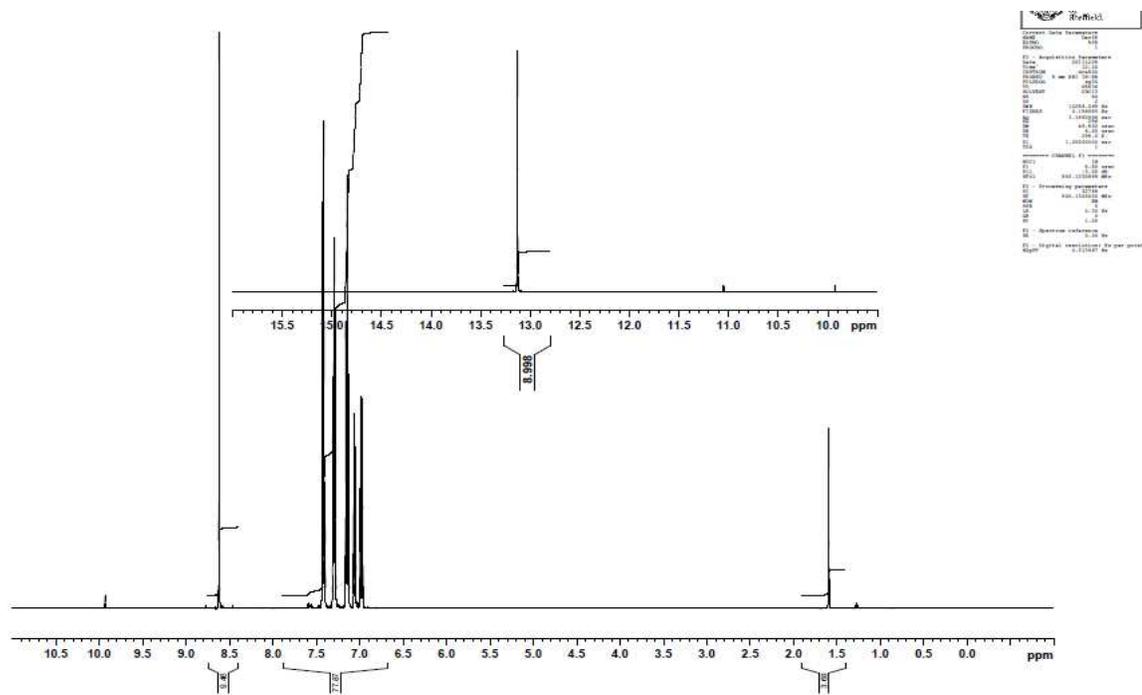


Fig. S 18. ¹H NMR (500MHz, CDCl₃) of EI-111-4F

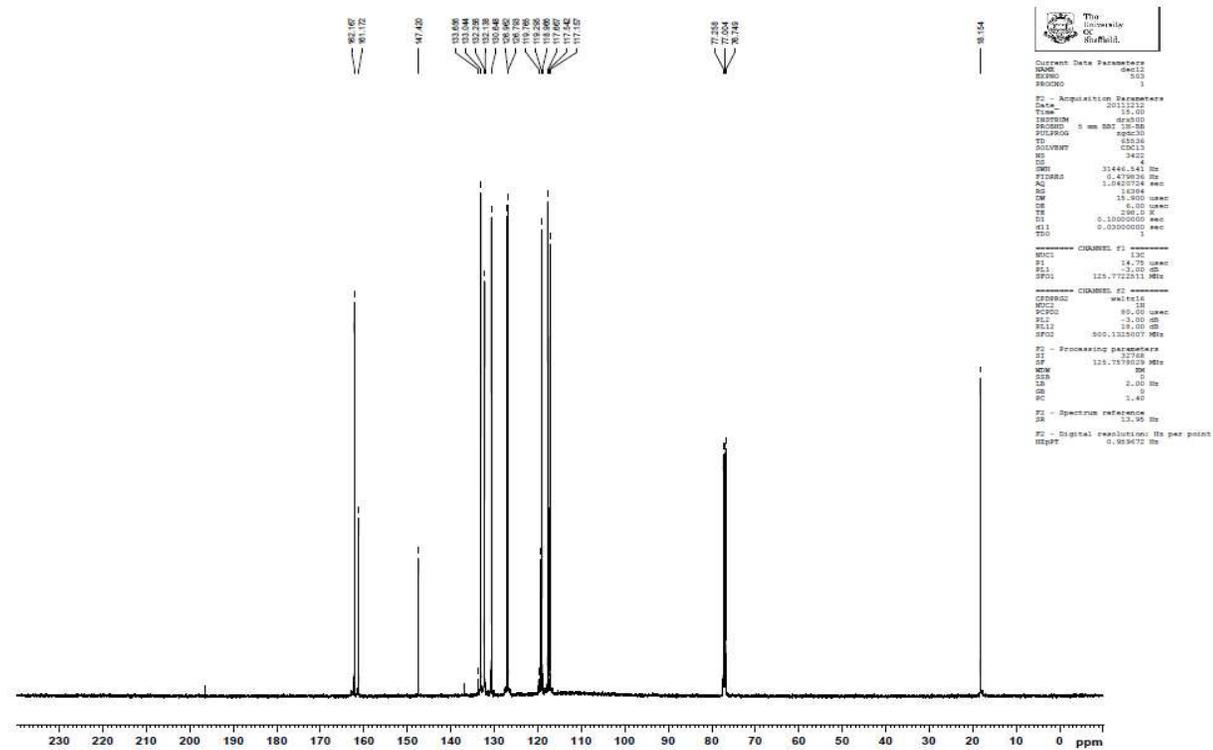


Fig. S 19. ¹³C NMR (500MHz, CDCl₃) of EI-111-2Me

Experimental Details

Synthesis of EI-111.

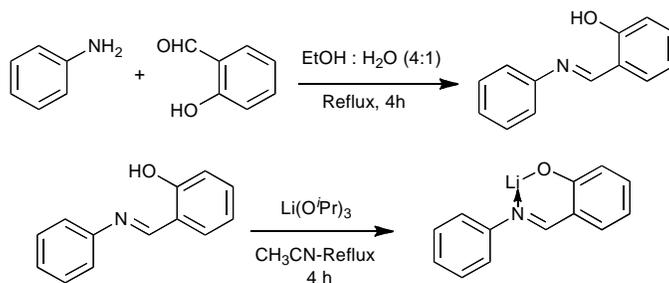
Synthesis of 2-((Phenylimino)methyl)Phenol

To a mixture of salicylaldehyde (40.0 mL, 45.8 g, 375.3 mmol) and aniline (22 mL, 31.7 g, 374.6 mmol), was added ethanol (60 mL) and water (10 mL). This reaction mixture was refluxed for 2 h and then allowed to cool to room temperature for over 20 h. The crude orange coloured solid was filtered off and then washed with ethanol to give a yellow solid. The solid was purified by recrystallization from ethanol to give bright yellow fluorescent product, which was dried in a vacuum oven at 50 °C for 12 h to provide *N*-salicylidene-aniline (32.0 g, 58%).

δ_{H} (500 MHz, CDCl_3): 6.99 (1H, d, Ar-H, J 7.4Hz), 7.07(1H, d, Ar-H, J 8.1Hz), 7.32(3H, m, Ar-H), 7.44(4H, m, Ar-H), 8.65(1H, s, CH=N), 13.30(1H,s, OH). IR (KBr disc) $\gamma_{\text{max}}/\text{cm}^{-1}$: 1614, 1589, 1564 (C=N, C=C), 1273, 1185, 1149 and 1073.

Synthesis of Lithium 2-((Phenylimino)methyl)Phenolate

To a stirred solution of *N*-salicylidene-aniline (25.0 g, 126.7 mmol) in dry acetonitrile (100 mL), was added lithium isopropoxide (1 M solution in Hexane) (139.0 mL, 139.0 mmol). The mixture was refluxed under nitrogen atmosphere at 80 °C for 4 h and then allowed to cool to room temperature. The white precipitate was filtered, washed with Petroleum Ether (40-60 mixtures) and dried in a vacuum oven to give the crude product of the desired Li-complex EI-111 (15g, 58.3% yield). The crude product was purified by sublimation (at 260 °C; 10^{-6} Torr) to give an analytically pure EI-111 as a yellow solid. m.p 301 °C (DSC, Onset); Anal. Calcd for $\text{C}_{13}\text{H}_{10}\text{LiNO}$: C, 76.85; H, 4.96; N, 6.89. Found: C, 76.91, H, 4.81, N, 6.86 %. MS (FAB⁺) m/z 203 (M^+); HRMS (FAB⁺) Calcd. For $\text{C}_{13}\text{H}_{10}\text{LiNO}$: 203.2718; found: 203.2713



Synthesis of EI-111-2Me.

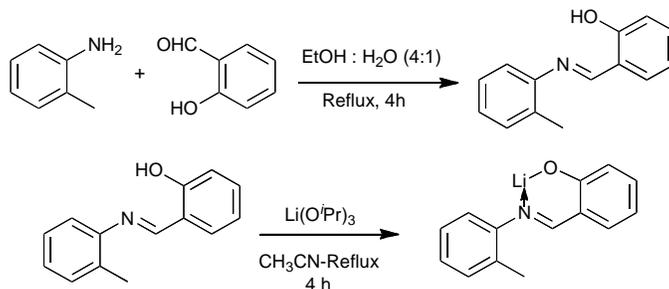
Synthesis of 2-((*o*-tolylimino)methyl)Phenol

To a mixture of salicylaldehyde (15.0 mL, 17.1 g, 140 mmol) and *o*-toluidine (15.0 mL, 15.0 g, 140 mmol), was added ethanol (50 mL) and water (10 mL). This reaction mixture was refluxed for 2 h and left vigorously stirred at room temperature over night. After cooled to room temperature, the crude orange coloured solid was filtered off and then washed with ethanol to give a yellow solid. The resulting yellow solid was purified by recrystallization from ethanol and then dried in a vacuum oven at 30 °C for 12 h to provide *N*-salicylidene-2-methylaniline (17.2 g, 47%). M.p 48 °C (DSC, onset)

δ_{H} (500 MHz, CDCl_3): 2.45 (3H, s, CH_3), 6.99(1H, t, Ar-H, J 6.6Hz), 7.10(1H, d, Ar-H, J 8.1Hz), 7.15(1H, d, Ar-H, J 7.5Hz), 7.25(1H, d, Ar-H, J 8.2Hz), 7.30(2H, m, Ar-H), 7.44(2H, m, Ar-H), 8.61 (1H, s, $\text{CH}=\text{N}$), 13.50 (1H, s, OH). ^{13}C NMR δ_{c} (500 MHz, CDCl_3): 162.17(C=N), 161.17(C-OH), 147.41, 133.04, 132.26, 132.14, 130.65, 126.96, 126.79, 119.29, 118.97, 117.67, 117.16 (11, Ar-C), 18.15(CH_3). IR (KBr disc) $\gamma_{\text{max}}/\text{cm}^{-1}$: 1616, 1595, 1569 (C=N, C=C), 1278, 1208, 1178, 1151 and 1112.

Synthesis of Lithium 2-((*o*-tolylimino)methyl)Phenolate

To a stirred solution of *N*-salicylidene-2-methylaniline (11.0 g, 52.1 mmol) in dry acetonitrile (60 mL), was added lithium isopropoxide (1 M solution in Hexane), (57.0 mL, 57.3 mmol), The mixture was refluxed under nitrogen atmosphere at 80 °C for 4 h and then allowed to cool to room temperature. The white precipitate was then filtered and washed with acetonitrile followed by Pet. Ether (40-60 mixtures) and dried in a vacuum oven to give the crude product of Li-complex EI-111-2Me (9.5 g, 84% yield). The product was dried under vacuum before purified by sublimation (at 190 °C; 10^{-6} Torr) to give an analytically pure EI-111-2Me as a pale yellow solid. m.p 249 °C (DSC, Onset); Anal. Calcd for $\text{C}_{14}\text{H}_{12}\text{LiNO}$: C, 77.42; H, 5.57; N, 6.45. Found: C, 77.35, H, 5.49, N, 6.42. MS (FAB⁺) m/z 217 (M^+); HRMS (FAB⁺) Calcd. For $\text{C}_{14}\text{H}_{12}\text{LiNO}$: 217.1836; found: 217.1842



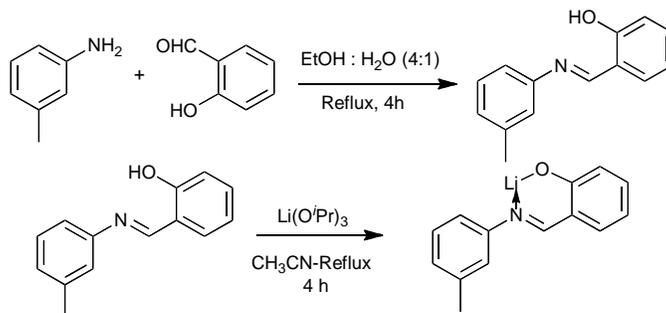
Synthesis of EI-111-3Me.

Synthesis of 2-((*m*-tolylimino)methyl)Phenol

To a mixture of salicylaldehyde (15.0 mL, 15.0 g, 140 mmol) and *m*-toluidine (16.3 mL, 18.7 g, 154 mmol), was added ethanol (45 mL) and water (10 mL). This reaction mixture was refluxed for 2 h and left vigorously stirred at room temperature overnight. After stirred at room temperature for over 20 h, the crude orange coloured solid was filtered off and washed with ethanol to give a yellow solid. The yellow solid was purified by recrystallization from ethanol and dried in a vacuum oven at 40 °C for 12 h to provide *N*-salicylidene-3-methylaniline (13.1 g, 44%). M.p 39 °C (DSC, onset). δ_{H} (500 MHz, CDCl_3):2.40 (3H, s, CH_3), 6.94(1H, dt, Ar-H, J 1.1 and 7.5Hz), 7.02(1H,d, Ar-H, J8.5Hz), 7.09(3H, m, Ar-H), 7.30(1H, t, Ar-H, J7.85Hz), 7.37(2H, m, Ar-H), 8.61 (1H,s, CH=N), not appeared (1H, s, OH). IR (KBr disc) $\gamma_{\text{max}}/\text{cm}^{-1}$: 1612, 1593, 1574 (C=N, C=C), 1278, 1209, 1151 and 1116.

Synthesis of Lithium 2-((*m*-tolylimino)methyl)Phenolate

To a stirred solution of *N*-salicylidene-3-methylaniline (7.0 g, 33.1 mmol) in dry acetonitrile (30 mL), was added lithium isopropoxide, 1 M solution in Hexane (36.0 mL, 36.4 mmol). The mixture was refluxed under nitrogen atmosphere at 80 °C for 4 h and then allowed to cool to room temperature. The white precipitate was then filtered, washed with acetonitrile followed by Pet. Ether (40-60 mixtures) and then dried under vacuum to give the crude product of desired Li-complex EI-111-3Me (6.3 g, 87% yield). The product was dried in vacuum before purified by sublimation (at 220 °C; 10^{-6} Torr) to give an analytically pure EI-111-3Me as a pale yellow solid. m.p 222 °C (DSC, Onset); Anal. Calcd for $\text{C}_{14}\text{H}_{12}\text{LiNO}$: C, 77.42; H, 5.57; N, 6.45. Found: C, 77.36, H, 5.49, N, 6.42. MS (CI) m/z 217 (M^+), 211, 225, 253.



Synthesis of EI-111-4Me.

Synthesis of 2-((*p*-tolylimino)methyl)Phenol

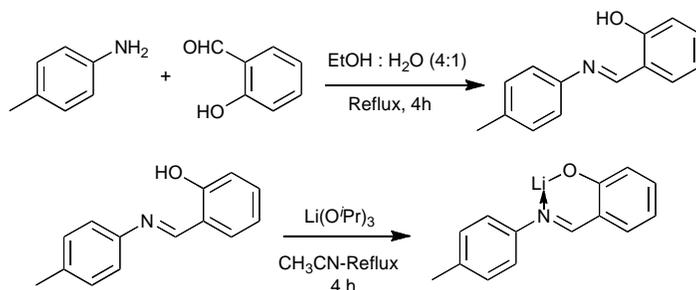
To a mixture of salicylaldehyde (15.1 mL, 15.0 g, 140 mmol) and *p*-toluidine (16.5 mL, 18.9 g, 155 mmol), was added ethanol (50 mL) and water (10 mL). This reaction mixture was refluxed for 2 h and cooled to room temperature for over 20 h. The crude orange coloured solid was filtered off and then washed with ethanol to give a yellow solid. The yellow solid was purified by recrystallization from ethanol and then dried in vacuum oven at 40 °C for 12 h to provide *N*-salicylidene-4-methylaniline (13.2 g, 45%). M.p 37 °C (DSC, onset).

δ_{H} (500 MHz, CDCl_3): 2.30(3H, s, CH_3), 6.85(1H, t, Ar-H, $J_{7.5\text{Hz}}$), 6.94(1H, d, Ar-H, $J_{8.2\text{Hz}}$), 7.14(4H, m, Ar-H), 7.28(2H, m, Ar-H), 8.53(1H, s, $\text{CH}=\text{N}$) and 13.30(1H, s, OH).

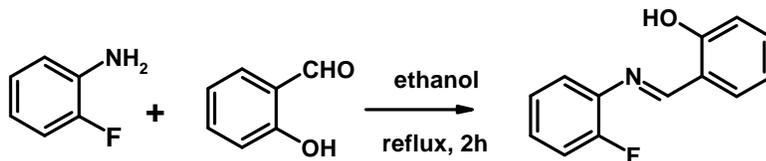
IR (KBr disc) $\gamma_{\text{max}}/\text{cm}^{-1}$: 1614, 1600, 1567 (C=N, C=C), 1364, 1281, 1179 and 1152.

Synthesis of Lithium 2-((*p*-tolylimino)methyl)Phenolate

To a stirred solution of *N*-salicylidene-4-methylaniline (13.0 g, 66.6 mmol) in dry acetonitrile (80 mL), was added lithium isopropoxide, 1 M solution in Hexane). (68.0 mL, 67.7 mmol). The mixture was refluxed under nitrogen atmosphere at 80 °C for 4 h and then allowed to cool to room temperature. The white precipitate was filtered, washed with acetonitrile followed by Pet. Ether (40-60 mixtures) and dried under vacuum to give the crude product of desired Li-complex EI-111-4Me (10.0 g, 75% yield). The crude product was dried in vacuum before purified by sublimation (at 230 °C; 10^{-6} Torr) to give an analytically pure EI-111-2Me as a pale yellow solid. m.p 318.5 °C (DSC, Onset); Anal. Calcd for $\text{C}_{14}\text{H}_{12}\text{LiNO}$: C, 77.42; H, 5.57; N, 6.45. Found: C, 76.73, H, 5.30, N, 6.44. MS (FAB^+) m/z 217 (M^+), 210, 212, 239, 253.



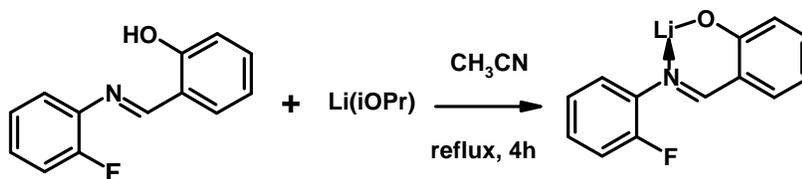
Synthesis of 2-[(2-Fluoro-phenylimino)-methyl]phenol



To 2-Fluoroaniline (10.0 g; 0.09 mole) in ethanol (60 ml) was added salicylaldehyde (8.8 g; 0.072 mole). The solution was magnetically stirred and refluxed under nitrogen for 2h. To the cooled reaction mixture small amounts of water added and on cooling a yellow crystalline solid was obtained. The product was filtered off and dried under vacuum at 40 °C to give 10.8g, (56 %) of the product. M.p 68 °C (DSC, onset).

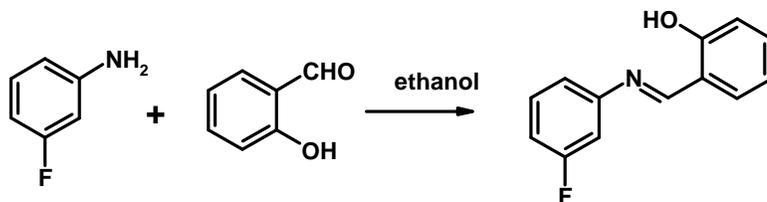
δ_{H} (500 MHz, CDCl_3): 6.98 (1H,t,Ar-H, J 7.5Hz), 7.07(1H, d, Ar-H, J 8.3Hz), 7.26(4H, m, Ar-H), 7.42(2H, m, Ar-H), 8.74(1H, s, CH=N), 13.14(1H,s, OH). IR (KBr disc) $\gamma_{\text{max}}/\text{cm}^{-1}$: 1620, 1597, 1571 (C=N, C=C), 1491, 1281, 1227, 1178 and 1105.

Synthesis of Lithium 2-[(2-Fluoro-phenylimino)-methyl]phenolate



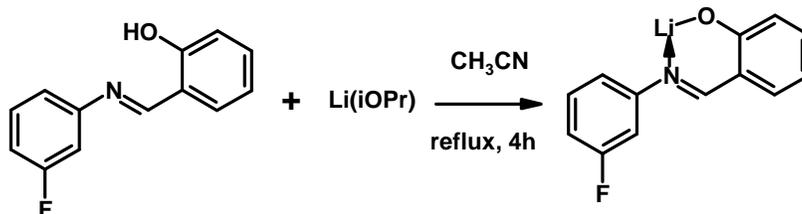
To a stirred solution of 2-[(2-Fluoro-phenylimino)-methyl]phenol (8.0 g, 37.2 mmol) in dry acetonitrile (50 mL), was added lithium isopropoxide, 1 M solution in Hexane (41.0 mL, 40.9 mmol). The mixture was refluxed under nitrogen atmosphere at 80 °C for 4 h and then allowed to cool to room temperature. The pale yellow precipitate was then filtered, washed with acetonitrile followed by Pet. Ether (40-60 mixtures) and then dried under vacuum to give the crude product of desired Li-complex EI-111-2F (6.2 g, 76% yield). The product was dried in vacuum before purified by sublimation (at 220 °C; 10^{-6} Torr) to give an analytically pure EI-111-2F as a yellow solid. m.p 276 °C (DSC, Onset); Anal: Found, C 70.41, H 3.94, N 6.43, F 8.09. $\text{C}_{13}\text{H}_9\text{FLiNO}$ requires, C 70.60, H 4.10, N 6.33 and F 8.59 %.

Synthesis of 2-[(3-Fluoro-phenylimino)-methyl]phenol



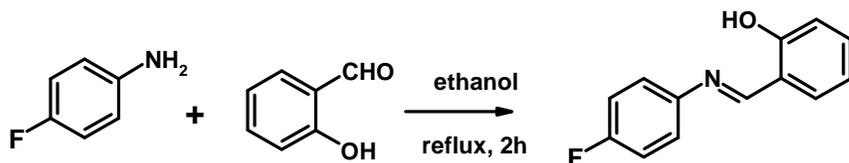
To 3-Fluoroaniline (5.0 g; 0.045 mole) in ethanol (50 ml) was added salicylaldehyde (5.2 g; 0.043 mole). The solution was magnetically stirred and refluxed under nitrogen for 2h. To the cooled reaction mixture small amounts of water added and on cooling a yellow crystalline solid separated out. The product was filtered off and dried under vacuum at 40 °C to give 6.2g, (62 %) of the product. M.p 41 °C (DSC, onset). δ_{H} (500 MHz, CDCl_3): 7.00 (5H,m,Ar-H), 7.38(3H, m, Ar-H), 8.59(1H, s, CH=N), Not appeared(1H,s, OH). IR (KBr disc) $\gamma_{\text{max}}/\text{cm}^{-1}$: 1622, 1603, 1590, 1576 (C=N, C=C), 1499, 1476, 1461, 1410, 1372, 1282, 1259, 1219, 1202, 1151 and 1129.

Synthesis of Lithium 2-[(3-Fluoro-phenylimino)-methyl]phenolate



To a stirred solution of 2-[(3-Fluoro-phenylimino)-methyl]phenol (5.0 g, 23.3 mmol) in dry acetonitrile (30 mL), was added lithium isopropoxide, 1 M solution in Hexane (26.0 mL, 25.6 mmol). The mixture was refluxed under nitrogen atmosphere at 80 °C for 4 h and then allowed to cool to room temperature. The pale yellow precipitate was then filtered, washed with acetonitrile followed by Pet. Ether (40-60 mixtures) and then dried under vacuum to give the crude product of desired Li-complex EI-111-3F (4.2 g, 82% yield). The product was dried in vacuum before purified by sublimation (at 220 °C; 10^{-6} Torr) to give an analytically pure EI-111-3F as a pale yellow solid. m.p 314 °C (DSC, Onset); Anal: Found, C 70.46, H 4.03, N 6.34. $\text{C}_{13}\text{H}_9\text{FLiNO}$ requires, C 70.60, H 4.10 and N 6.33 %.

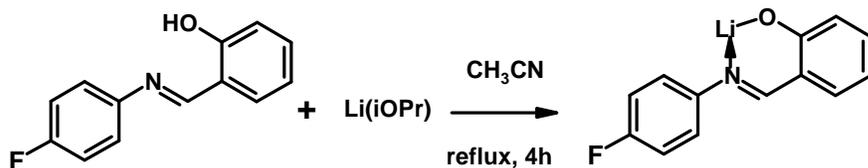
Synthesis of 2-[(4-Fluoro-phenylimino)-methyl]phenol



To 2-Fluoroaniline (10.0 g; 0.09 mole) in ethanol (60 ml) was added salicylaldehyde (8.8 g; 0.072 mole). The solution was magnetically stirred and refluxed under nitrogen for 2h. To the cooled reaction mixture small amounts of water added and on cooling a yellow crystalline solid was obtained. The product was filtered off and dried under vacuum at 40 °C to give 15g, (77 %) of the product. M.p 82 °C (DSC, onset).

δ_{H} (500 MHz, CDCl_3): 6.98 (1H,t,Ar-H, J 7.5Hz), 7.05(1H, d, Ar-H, J 8.4Hz), 7.14(2H, t, Ar-H, J8.6Hz), 7.29(2H, m, Ar-H), 7.41(2H, m, Ar-H), 8.62(1H,s, CH=N) and 13.13 (1H, s, OH). IR (KBr disc) $\gamma_{\text{max}}/\text{cm}^{-1}$: 1615, 1591, 1570 (C=N, C=C), 1504, 1491, 1457, 1272, 1229, 1182 and 1150.

Synthesis of Lithium 2-[(4-Fluoro-phenylimino)-methyl]phenolate



To a stirred solution of 2-[(4-Fluoro-phenylimino)-methyl]phenol (10.0 g, 46.5 mmol) in dry acetonitrile (60 mL), was added lithium isopropoxide, 1 M solution in Hexane (52.0 mL, 51.2 mmol). The mixture was refluxed under nitrogen atmosphere at 80 °C for 4 h and then allowed to cool to room temperature. The yellow solid was then filtered, washed with acetonitrile followed by Pet. Ether (40-60 °C) and then dried under vacuum to give the crude product of desired Li-complex EI-111-4F (7.4 g, 72% yield). The product was dried in vacuum before purified by sublimation (at 220 °C; 10^{-6} Torr) to give an analytically pure EI-111-4F as a yellow solid. m.p 279 °C (DSC, Onset); Anal: Found, C 70.44, H 3.86, N 6.42,. $\text{C}_{13}\text{H}_9\text{FLiNO}$ requires, C 70.60, H 4.10, and N 6.33 %.

Diffraction data were acquired at 150 K, as ω scans on a Bruker SMART1000 CCD area detector diffractometer, using graphite-monochromated Mo- K_{α} radiation ($\lambda = 0.71073 \text{ \AA}$). Data were corrected for Lorentz and polarization effects. The structures were solved by direct methods using *SHELXS97*^{X1}, developed by iterative cycles of least squares refinement and difference Fourier synthesis, and refined by full-matrix least-squares on F^2 using *SHELXL97*^{X1}. All non-hydrogen atoms were refined anisotropically. Hydrogen atoms were introduced at geometrically calculated positions and refined using a riding model with $U_{\text{iso}}(\text{H}) = 1.2U_{\text{eq}}(\text{C})$. The Crystallographic Information File was prepared using *enCIFer*^{X2} and validated using *PLATON*^{X3}.

Crystal structure determination references:

X1. G.M. Sheldrick, *Acta Crystallogr., Sect. A*, 2008, **64**, 112-122.

X2. F.H. Allen, O. Johnson, G. P. Shields, B. R. Smith & M. Towler, *J. Appl. Crystallogr.*, 2004, **37**, 335-338.

X3. A.L. Spek, *Acta Crystallogr., Sect. D*, 2009, **65**, 148-155.