**Supplementary Table 1**  The different optimized parameters (bond lengths and bond angles) of organometallic Schiff base ligand (H2L).

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| **Bond length (Å)** | | | | | | |
| C(1)-C(2) | | 1.4436 | C(14)-H(18) | | 1.0808 | |
| C(1)-C(5) | | 1.4429 | C(16)-C(17) | | 1.4350 | |
| C(1)-H(6) | | 1.0811 | C(16)-H(19) | | 1.0801 | |
| C(1)-Fe(11) | | 2.1180 | C(17)-H(20) | | 1.0808 | |
| C(2)-C(3) | | 1.4432 | C(21)-C(22) | | 1.5199 | |
| C(2)-H(7) | | 1.0811 | C(21)-N(26) | | 1.2979 | |
| C(2)-Fe(11) | | 2.1201 | C(22)-H(23) | | 1.0973 | |
| C(3)-C(4) | | 1.4427 | C(22)-H(24) | | 1.0983 | |
| C(3)-H(8) | | 1.0814 | C(22)-H(25) | | 1.0918 | |
| C(3)-Fe(11) | | 2.1213 | N(26)-C(27) | | 1.3978 | |
| C(4)-C(5) | | 1.4433 | C(27)-C(28) | | 1.4294 | |
| C(4)-H(9) | | 1.0810 | C(27)-C(29) | | 1.4104 | |
| C(4)-Fe(11) | | 2.1208 | C(28)-C(30) | | 1.3900 | |
| C(5)-H(10) | | 1.0810 | C(28)-H(31) | | 1.0853 | |
| C(5)-Fe(11) | | 2.1179 | C(29)-C(32) | | 1.4032 | |
| Fe(11)-C(12) | | 2.1155 | C(29)-H(33) | | 1.0845 | |
| Fe(11)-C(13) | | 2.1152 | C(30)-C(34) | | 1.4208 | |
| Fe(11)-C(14) | | 2.1286 | C(30)-H(35) | | 1.0853 | |
| Fe(11)-C(16) | | 2.1101 | C(32)-C(34) | | 1.4328 | |
| Fe(11)-C(17) | | 2.1285 | C(32)-O(40) | | 1.3768 | |
| C(12)-C(13) | | 1.4550 | C(34)-C(36) | | 1.4532 | |
| C(12)-C(14) | | 1.4353 | C(36)-O(37) | | 1.2673 | |
| C(12)-H(15) | | 1.0777 | C(36)-O(38) | | 1.3827 | |
| C(13)-C(16) | | 1.4553 | O(38)-H(39) | | 0.9836 | |
| C(13)-C(21) | | 1.4822 | O(40)-H(41) | | 0.9992 | |
| C(14)-C(17) | | 1.4431 |  | |  | |
| **Bond angle (°)** | | | | | |
| C(2)-C(1)-C(5) | 107.9887 | | | Fe(11)-C(12)-H(15) | 125.0209 |
| C(2)-C(1)-H(6) | 126.0294 | | | C(13)-C(12)-C(14) | 108.2354 |
| C(5)-C(1)-H(6) | 125.9788 | | | C(13)-C(12)-H(15) | 126.1827 |
| H(6)-C(1)-Fe(11) | 124.8504 | | | C(14)-C(12)-H(15) | 125.5818 |
| C(1)-C(2)-C(3) | 107.9242 | | | Fe(11)-C(13)-C(21) | 124.6037 |
| C(1)-C(2)-H(7) | 125.9641 | | | C(12)-C(13)-C(16) | 106.9621 |
| C(3)-C(2)-H(7) | 126.1113 | | | C(12)-C(13)-C(21) | 128.6351 |
| H(7)-C(2)-Fe(11) | 125.5637 | | | C(16)-C(13)-C(21) | 124.3930 |
| C(2)-C(3)-C(4) | 108.1122 | | | Fe(11)-C(14)-H(18) | 125.3070 |
| C(2)-C(3)-H(8) | 126.0340 | | | C(12)-C(14)-C(17) | 108.3387 |
| C(4)-C(3)-H(8) | 125.8511 | | | C(12)-C(14)-H(18) | 125.6395 |
| H(8)-C(3)-Fe(11) | 125.0177 | | | C(17)-C(14)-H(18) | 126.0185 |
| C(3)-C(4)-C(5) | 107.9048 | | | Fe(11)-C(16)-H(19) | 125.5060 |
| C(3)-C(4)-H(9) | 126.0210 | | | C(13)-C(16)-C(17) | 108.4307 |
| C(5)-C(4)-H(9) | 126.0727 | | | C(13)-C(16)-H(19) | 125.9283 |
| H(9)-C(4)-Fe(11) | 125.0672 | | | C(17)-C(16)-H(19) | 125.6358 |
| C(1)-C(5)-C(4) | 108.0701 | | | Fe(11)-C(17)-H(20) | 125.7768 |
| C(1)-C(5)-H(10) | 125.9879 | | | C(14)-C(17)-C(16) | 108.0302 |
| C(4)-C(5)-H(10) | 125.9391 | | | C(14)-C(17)-H(20) | 126.0745 |
| H(10)-C(5)-Fe(11) | 124.8516 | | | C(16)-C(17)-H(20) | 125.8952 |
| C(1)-Fe(11)-C(3) | 66.8218 | | | C(13)-C(21)-C(22) | 116.9237 |
| C(1)-Fe(11)-C(4) | 66.8846 | | | C(13)-C(21)-N(26) | 126.3818 |
| C(1)-Fe(11)-C(12) | 158.3470 | | | C(22)-C(21)-N(26) | 116.6922 |
| C(1)-Fe(11)-C(13) | 159.8498 | | | C(21)-C(22)-H(23) | 110.9765 |
| C(1)-Fe(11)-C(14) | 123.4913 | | | C(21)-C(22)-H(24) | 110.7208 |
| C(1)-Fe(11)-C(16) | 123.9779 | | | C(21)-C(22)-H(25) | 109.2524 |
| C(1)-Fe(11)-C(17) | 108.8960 | | | H(23)-C(22)-H(24) | 107.5778 |
| C(2)-Fe(11)-C(4) | 66.8607 | | | H(23)-C(22)-H(25) | 109.4182 |
| C(2)-Fe(11)-C(5) | 66.8692 | | | H(24)-C(22)-H(25) | 108.8511 |
| C(2)-Fe(11)-C(12) | 160.2095 | | | N(26)-C(27)-C(28) | 119.9179 |
| C(2)-Fe(11)-C(13) | 124.7591 | | | N(26)-C(27)-C(29) | 120.0711 |
| C(2)-Fe(11)-C(14) | 159.2140 | | | C(28)-C(27)-C(29) | 119.7091 |
| C(2)-Fe(11)-C(16) | 109.9255 | | | C(27)-C(28)-C(30) | 119.8234 |
| C(2)-Fe(11)-C(17) | 124.5365 | | | C(27)-C(28)-H(31) | 119.1204 |
| C(3)-Fe(11)-C(5) | 66.7917 | | | C(30)-C(28)-H(31) | 121.0546 |
| C(3)-Fe(11)-C(12) | 124.5295 | | | C(27)-C(29)-C(32) | 120.3808 |
| C(3)-Fe(11)-C(13) | 109.8357 | | | C(27)-C(29)-H(33) | 120.7534 |
| C(3)-Fe(11)-C(16) | 125.3604 | | | C(32)-C(29)-H(33) | 118.8609 |
| C(3)-Fe(11)-C(17) | 160.2581 | | | C(28)-C(30)-C(34) | 121.2400 |
| C(4)-Fe(11)-C(12) | 108.8506 | | | C(28)-C(30)-H(35) | 120.4425 |
| C(4)-Fe(11)-C(13) | 124.2894 | | | C(34)-C(30)-H(35) | 118.3175 |
| C(4)-Fe(11)-C(14) | 123.4105 | | | C(29)-C(32)-C(34) | 120.2248 |
| C(4)-Fe(11)-C(16) | 160.4953 | | | C(29)-C(32)-O(40) | 117.5504 |
| C(5)-Fe(11)-C(12) | 123.1982 | | | C(34)-C(32)-O(40) | 122.2244 |
| C(5)-Fe(11)-C(13) | 159.1654 | | | C(30)-C(34)-C(32) | 118.6139 |
| C(5)-Fe(11)-C(14) | 108.3310 | | | C(30)-C(34)-C(36) | 122.3206 |
| C(5)-Fe(11)-C(17) | 123.2579 | | | C(32)-C(34)-C(36) | 119.0634 |
| C(12)-Fe(11)-C(16) | 67.2135 | | | C(34)-C(36)-O(37) | 124.7097 |
| C(12)-Fe(11)-C(17) | 66.7152 | | | C(34)-C(36)-O(38) | 115.3791 |
| C(13)-Fe(11)-C(14) | 66.9824 | | | O(37)-C(36)-O(38) | 119.9112 |
| C(13)-Fe(11)-C(17) | 67.0772 | | | C(36)-O(38)-H(39) | 110.0013 |
| C(14)-Fe(11)-C(16) | 66.6542 | | | C(32)-O(40)-H(41) | 110.0769 |

**Supplementary Table 3** Main calculated optical transitions with composite ion in terms of molecular orbitals.

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| --- | --- | --- | --- | --- | --- |
| **Compound** | **Transition** | **Excitation energy (eV)** | **λmax calc.**  **nm (eV)** | **λmax exp.**  **nm (eV)** | **Oscillator strength** |
| **Ligand (H2L)** | HOMO-4 LUMO (68%) | 5.04 | 277 (4.48) | 271 (4.57) | 0.005 |
| HOMO LUMO+3 (48%)  HOMO-2 LUMO (38%) | 5.32  4.40 | 312 (3.98) | 314 (3.95) | 0.3211 |