**Supporting Information**

***Frontier molecular orbitals (FMOs) and chemical activity***

The excitation energy of a molecule involves the difference between the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) of a neutral system. HOMO-LUMO gap is generally the lowest energy electronic excitation that is possible in a molecule and helps to characterize the chemical reactivity and the kinetic stability of the molecule. A molecule with a small frontier orbital gap is generally associated with a high chemical reactivity, low kinetic stability and is also termed as soft molecule [39]. The chemical activity (global descriptors) calculated from the FMOs energies (EHOMO and ELUMO). The smaller energy gap and global hardness values mean the more reactive molecule; electrophilicity index in terms of the electronic chemical potential and global hardness represents the capability of the molecule to accept the electrons from a donor environment.

By using HOMO and LUMO energy values for a molecule, electronegativity and chemical hardness can be calculated as follow:

(electronegativity) (8)

(chemical potential) (9)

(chemical hardness) (10)

= (chemical softness) (11)

(electrophilicity index) (12)

Where I and A are ionization potential and electron affinity; I = − EHOMO and A= − ELUMO respectively. The (HOMO) and (LUMO) energies are -5.348 eV and -1.969 eV, respectively. According to DFT calculation, the frontier orbital energy gap was found to be 3.37 eV.

The values of electronegativity, chemical potential, chemical hardness, softness, and electrophilicity indexes are 3.658 eV, -3.658 eV, 1.685 eV, 0.296 eV, and 3.970 eV in gas phase, respectively. Lower values of hardness and Mulliken electronegativity are supported that the title molecule was soft and more reactive molecule. Hence, the energy gap of compound was lower for metal-complexes [40,41]. The smaller energy gap of HOMO–LUMO explains the eventual charge transfer occurs within the molecule, which influences its high polarizability/hyperpolarizability values and biological activities.

***Net charges (MPA and NPA)***

The knowledge of charge distribution of the molecule is very much essential to understand the chemical reactivity, molecular electrostatic potential and the electrostatic interactions [42, 43]. The MPA and NPA charges are of the simplest ways to display the distribution of electrons between atoms forming molecule that shows which atom accepts electron and has negative charge and which atom loses electron and has positive charge. To explore the electrostatic potential (electrophilic/nucleophilic nature) of the molecule, here, we reported two kinds of charges, which were obtained from MPA [44, 45] and NPA [46]. The total atomic charge values were obtained by MPA and natural charges are obtained by NBO although both methods predict the same tendencies. The total atomic charge values for title compound were listed in Table S2.

[Table S2]

The most negative charges were mainly on Ni, N1, N2, O1, O2, O3 and O4 atoms. As can be seen in Table S2, the other electrophilic regions were located on C1, C5, C6, C7, C8, C10, C11, C12, C13, C15 and C16 atoms. Besides, the atomic charges explained the presence of hydrogen bonding of N-H…O, O-H…O and C-H…*π* interactions. All hydrogen atoms and C2, C3, C4, C9 and C14 atoms were carrying high positive charges. Title compound was shown the higher electrophilic nature. The results of MEP, NPA and MPA could be used for interpreting and predicting the reactive behavior of a wide variety of chemical systems in both electrophilic and nucleophilic reactions.

***Molecular electrostatic potential (MEP)***

Molecular electrostatic potential (MEP) is related to the electronic density and is a very useful descriptor in understanding sites for electrophilic attack and nucleophilic reactions as well as hydrogen bonding interactions [47]. Different values of the electrostatic potential are represented by various colours; red represented the regions of the most negative electrostatic potential and blue represents the region of zero potential. Negative (red colour) and positive (blue colour) regions of MEP are related to electrophilic and nucleophilic reactivity. The potential increases in the order red < orange < yellow < green < blue.

[Fig. S1]

In Fig. S1a demonstrated that Ni, N and O atoms displayed relatively negative potential, while benzene group protons carry the most positive potential. Negative potential region was given information about inter and intra molecular hydrogen bonds. According to above mentioned conditions, in the case of the chemical reactions or intermolecular interactions for chemical active moieties of the complex can play important roles. The MEP surface, local (Fukui function analysis) and global chemical descriptors (, , , and parameters) and atomic charge distribution results are supported each other for identifying the electrophilic and nucleophilic attacks. In addition, the MEP contour plot is a two dimensional display of the regions where the values of the relative electron density lie within a specific range. The electrostatic potential contour map for positive and negative potentials was shown in Fig. S1b, for Ni(II) complex. These results provided information concerning the region where the compound could be interact intra molecularly.

**Table S1.** Second order perturbation theory analysis of Fock matrix in NBO for Ni(II) complex.

|  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **Donor (i)**  **(occupancy)** | **Type** | **EDA,%**  **EDB,%** | **Acceptor (j)**  **(occupancy)** | **Type** | **EDA,%**  **EDB,%** | **E(2)a**  **(kcal/mol)** | **Ej-Eib (a.u.)** | **F(ij)c (a.u.)** |
| BD C16-H16c  (0.99488) | σ | 62.20  37.80 | RY\*C16  (0.00023) | - | - | 6.90 | 1.56 | 0.131 |
| BD C16-H16c  (0.99488) | σ | 62.20  37.80 | RY\*C13  (0.00130) | - | - | 9.44 | 0.86 | 0.114 |
| BD C16-H16c  (0.99488) | σ | 62.20  37.80 | BD\*C12-C13  (0.15428) | π\* | 52.46  47.54 | 17.06 | 0.29 | 0.096 |
| BD C16-H16b  (0.99024) | σ | 62.06  37.94 | RY\*C16  (0.00023) | - | - | 13.23 | 1.59 | 0.184 |
| BD C16-H16b  (0.99024) | σ | 62.06  37.94 | RY\*C14  (0.00023) | - | - | 5.20 | 1.87 | 0.125 |
| BD C16-H16b  (0.99024) | σ | 62.06  37.94 | RY\*C13  (0.00130) | - | - | 19.52 | 0.89 | 0.167 |
| BD C16-H16b  (0.99024) | σ | 62.06  37.94 | RY\*H16c  (0.00008) | - | - | 7.34 | 0.75 | 0.094 |
| BD C16-H16b  (0.99024) | σ | 62.06  37.94 | BD\*C12-C13  (0.15428) | π\* | 52.46  47.54 | 32.51 | 0.32 | 0.139 |
| BD C16-H16b  (0.99024) | σ | 62.06  37.94 | BD\*C11-H11  (0.00849) | σ \* | 37.69  62.31 | 8.76 | 0.85 | 0.109 |
| BD C14-C15  (0.98969) | σ | 51.30  48.70 | RY\*C14  (0.00013) | - | - | 247.08 | 0.09 | 0.184 |
| BD C14-C15  (0.98969) | σ | 51.30  48.70 | BD\*C12-C13  (0.15428) | π\* | 52.46  47.54 | 7.36 | 0.48 | 0.081 |
| BD C15-N2  (0.99160) | σ | 39.21  60.79 | RY\*C16  (0.00022) | - | - | 9.12 | 1.56 | 0.149 |
| BD C15-N2  (0.99160) | σ | 39.21  60.79 | RY\*C16  (0.00023) | - | - | 59.55 | 1.83 | 0.418 |
| BD C15-N2  (0.99160) | σ | 39.21  60.79 | RY\*C14  (0.00015) | - | - | 17.99 | 2.59 | 0.274 |
| BD C15-N2  (0.99160) | σ | 39.21  60.79 | RY\*C14  (0.00013) | - | - | 1785.35 | 0.17 | 0.701 |
| BD C15-N2  (0.99160) | σ | 39.21  60.79 | RY\*C14  (0.00002) | - | - | 21.05 | 2.11 | 0.267 |
| BD C15-N2  (0.99160) | σ | 39.21  60.79 | RY\*C15  (0.00006) | - | - | 15.57 | 2.23 | 0.236 |
| BD C15-N2  (0.99160) | σ | 39.21  60.79 | RY\*C13  (0.00130) | - | - | 71.47 | 1.13 | 0.360 |
| BD C15-N2  (0.99160) | σ | 39.21  60.79 | RY\*C11  (0.00023) | - | - | 16.73 | 1.86 | 0.224 |
| BD C15-N2  (0.99160) | σ | 39.21  60.79 | RY\*H16c  (0.00008) | - | - | 25.72 | 0.99 | 0.202 |
| BD C15-N2  (0.99160) | σ | 39.21  60.79 | RY\*N2  (0.00091) | - | - | 33.50 | 1.78 | 0.309 |
| BD C15-N2  (0.99160) | σ | 39.21  60.79 | BD\*C14-C15  (0.01410) | σ \* | 48.70  51.30 | 37.47 | 1.20 | 0.268 |
| BD C15-N2  (0.99160) | σ | 39.21  60.79 | BD\*C12-C13  (0.15428) | π\* | 52.46  47.54 | 90.31 | 0.56 | 0.308 |
| BD C15-N2  (0.99160) | σ | 39.21  60.79 | BD\*C11-H11  (0.00849) | σ \* | 37.69  62.31 | 22.75 | 1.10 | 0.199 |
| BD C12-C13  (0.82366) | π | 47.54  52.46 | BD\*C12-C13  (0.15428) | π\* | 52.46  47.54 | 19.74 | 0.01 | 0.021 |
| BD C11-C12  (0.99172) | σ | 50.55 49.45 | RY\*C16  (0.00022) | - | - | 29.14 | 1.89 | 0.298 |
| BD C11-C12  (0.99172) | σ | 50.55 49.45 | RY\*C14  (0.00013) | - | - | 657.45 | 0.23 | 0.496 |
| BD C11-C12  (0.99172) | σ | 50.55 49.45 | RY\*C13  (0.00130) | - | - | 23.68 | 1.19 | 0.213 |
| BD C11-C12  (0.99172) | σ | 50.55 49.45 | BD\*C12-C13  (0.15428) | π\* | 52.46  47.54 | 40.61 | 0.62 | 0.217 |
| BD C11-C12  (0.99172) | σ | 50.55 49.45 | BD\*C11-H11  (0.00849) | σ \* | 37.69  62.31 | 22.48 | 1.16 | 0.204 |
| BD C11-C12  (0.99172) | σ | 50.55 49.45 | BD\*C11-N2  (0.01061) | σ \* | 60.87  39.13 | 14.41 | 1.26 | 0.170 |
| BD C11-N2  (0.99225) | σ | 39.13  60.87 | RY\*C14  (0.00013) | - | - | 556.59 | 0.20 | 0.419 |
| BD C11-N2  (0.99225) | σ | 39.13  60.87 | RY\*C13  (0.00130) | - | - | 25.51 | 1.16 | 0.218 |
| BD C11-N2  (0.99225) | σ | 39.13  60.87 | BD\*C11-H11  (0.01061) | σ \* | 60.87  39.13 | 8.86 | 1.12 | 0.126 |
| BD C11-N2  (0.87579) | π\* | 36.90  63.10 | BD\*C12-C13  (0.15428) | π\* | 52.46  47.54 | 28.08 | 0.07 | 0.056 |
| CR N2  (0.99973) | - | - | RY\*C16  (0.00023) | - | - | 56.63 | 15.21 | 1.172 |
| CR N2  (0.99973) | - | - | RY\*C14  (0.00013) | - | - | 177.96 | 13.56 | 1.961 |
| CR N2  (0.99973) | - | - | BD\*C15-N2  (0.00954) | σ \* | 60.79  39.21 | 220.63 | 12.99 | 2.147 |
| BD C10-H10  (0.99548) | σ | 61.17  38.83 | RY\*C13  (0.00130) | - | - | 8.01 | 0.86 | 0.105 |
| BD C1-C9  (0.98575) | σ | 47.26  52.74 | RY\*C14  (0.00013) | - | - | 284.81 | 0.02 | 0.097 |
| BD C9-O3  (0.99680) | σ | 34.82  65.18 | RY\*C16  (0.00022) | - | - | 58.82 | 1.9 | 0.432 |
| BD C9-O3  (0.99680) | σ | 34.82  65.18 | RY\*C14  (0.00013) | - | - | 1004.1 | 0.33 | 0.724 |
| BD C9-O2  (0.99699) | σ | 34.38  65.62 | RY\*C14  (0.00013) | - | - | 79.70 | 0.30 | 0.195 |
| BD C8-N1  (0.99274) | σ | 38.66  61.34 | RY\*C14  (0.00013) | - | - | 22.69 | 0.15 | 0.073 |
| BD C7-H7  (0.99017) | σ | 62.01  37.99 | RY\*C14  (0.00013) | - | - | 1451.15 | 0.28 | 0.808 |
| BD\* C3-C4  (0.25825) | π\* | 48.04  51.96 | RY\*C14  (0.00013) | - | - | 295.51 | 4.49 | 2.860 |
| BD C9-O3  (0.99680) | σ | 34.82  65.18 | BD\* C8-N1  (0.01152) | σ \* | 61.34  38.66 | 21.51 | 1.33 | 0.214 |
| CR O3  (0.99990) | - | - | RY C16  (0.00023) | - | - | 43.17 | 19.88 | 1.169 |
| BD N1-H1  (0.99444) | σ | 26.79  73.21 | BD\* C3-C8  (0.01315) | σ \* | 50.25  49.75 | 0.90 | 1.26 | 0.043 |
| LP O3  (0.97734) | n | - | BD\* C6-C7  (0.15514) | π\* | 49.79  50.21 | 1.26 | 3.24 | 0.087 |
| LP O3  (0.92910) | n | - | BD\* C1-C9  (0.03521) | σ \* | 52.74  47.26 | 8.11 | 0.80 | 0.104 |
| LP O3  (0.92910) | n | - | BD\* C9-O2  (0.02746) | σ \* | 65.62  34.38 | 6.62 | 0.81 | 0.094 |
| LP N1  (0.80622) | n | - | RY\* O1  (0.00001) | - | - | 0.05 | 1.05 | 0.010 |
| BD O1-H1a  (0.99723) | σ | 23.86  76.14 | RY\* Ni  (0.00060) | - | - | 33.97 | 0.73 | 0.198 |
| LP Ni  (0.96992) | n | - | BD\* C11-N2  (0.22120) | π\* | 63.10  36.90 | 1.54 | 0.15 | 0.021 |
| LP Ni  (0.12875) | n | - | BD\* C6-C7  (0.15514) | π\* | 49.79  50.21 | 0.09 | 2.54 | 0.035 |

a *E*(2) means energy of hyperconjucative interactions (stabilization energy).

b Energy difference between donor (*i*) and acceptor (*j*) NBO orbitals.

c *F*(*i*,*j*) is the Fock matrix element between *i* and *j* NBO orbital.

Percentage electron density over bonded atoms (EDA,B,%).

**Table S2.** Calculated charges by MPA and NPA for Ni(II) complex.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| **Atom** | **MPA** | **NPA** | **Atom** | **MPA** | **NPA** |
| C1 | -0.065143 | -0.55084 | C13 | -0.524180 | -0.512689 |
| C2 | 0.101184 | 0.01188 | C14 | 0.339448 | 0.334313 |
| C3 | 1.570398 | 0.27715 | C15 | -0.011795 | -0.09198 |
| C4 | 0.325013 | 0.08765 | C16 | -0.771777 | -0.72160 |
| C5 | -0.425935 | -0.29194 | O1 | -0.468096 | -0.70963 |
| C6 | -0.560095 | -0.28235 | O2 | -0.159587 | -0.75278 |
| C7 | -0.304596 | -0.28712 | O3 | -0.234612 | -0.55379 |
| C8 | -1.509865 | -0.90655 | O4 | -0.234612 | -0.55379 |
| C9 | 0.089538 | 0.72160 | N1 | -0.096572 | -0.30239 |
| C10 | -0.330275 | -0.30649 | N2 | -0.530305 | -0.55084 |
| C11 | -0.147708 | -0.13312 | Ni | -1.147859 | -0.22341 |
| C12 | -0.491367 | -0.25766 |  |  |  |

**Figure S1.** MEP surface and MEP contour for positive and negative potentials was imposed along the [111] direction of Ni(II) complex.

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