**O-hydroxy Schiff bases derived from 2-hydroxy-4-methoxy benzaldehyde: Synthesis, X-ray studies and Hydrogen Bonding Attributes**

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$^1$H NMR of N'-(Z)-(2-hydroxy-4-methoxyphenyl)methylidcarbohydrazide

![NMR Spectrum](image)

**NMR-isoniazid**
$^{13}$C NMR of $N'-(Z)-(2$-hydroxy-4-methoxyphenyl)methylidene]pyridine-4-carbohydrazide
$^1$H NMR of N’-[(Z)-(2-hydroxy-4-methoxyphenyl)methylidene]pyridine-3-carbohydrazide
$^{13}$C NMR of $N'\text{-[}(Z)\text{-}(2\text{-hydroxy-4-methoxyphenyl)methylidene]pyridine-3-}
\text{carbohydrazide}$
Mass spectrum of \( N' - [(Z) - (2\text{-hydroxy-4\text{-methoxyphenyl})\text{methylidene}}]\text{pyridine-4\text{-carbohydrazide}} \)
Mass spectrum of \( N'\)-(Z)-(2-hydroxy-4-methoxyphenyl)methylidene]pyridine-3-carbohydrazide
Table 1: Bond lengths, r (Å) and bond angles, A (°) of the molecule in ground state.

| r/A                | 1  | 2  |
|--------------------|----|----|
|                    | XRD| B3LYP/ cc-pVDZ | XRD| B3LYP/ cc-pVDZ |
| R(1-2)             | 1.352| 1.339 | 1.331| 1.341 |
| R(1-6)             | 1.33 | 1.342 | 1.334 | 1.337 |
| R(2-3)             | 1.376| 1.398 | 1.363 | 1.398 |
| R(3-4)             | 1.385| 1.403 | 1.379 | 1.395 |
| R(4-5)             | 1.391| 1.402 | 1.376 | 1.402 |
| R(4-7)             | 1.5 | 1.503 | 1.389 | 1.407 |
| R(5-6)             | 1.375| 1.397 | 1.492 | 1.498 |
| R(7-8)             | 1.211| 1.223 | 1.227 | 1.224 |
| R(7-9)             | 1.368| 1.38 | 1.35 | 1.381 |
| R(9-10)            | 1.366| 1.359 | 1.379 | 1.357 |
| R(10-11)           | 1.285| 1.294 | 1.286 | 1.293 |
| R(11-12)           | 1.453| 1.447 | 1.444 | 1.448 |
| R(12-13)           | 1.41 | 1.414 | 1.405 | 1.413 |
| R(12-17)           | 1.405| 1.426 | 1.399 | 1.425 |
| R(13-14)           | 1.372| 1.382 | 1.368 | 1.382 |
| R(14-15)           | 1.4 | 1.414 | 1.388 | 1.413 |
| R(15-16)           | 1.389| 1.397 | 1.385 | 1.398 |
| R(16-17)           | 1.403| 1.405 | 1.386 | 1.403 |
| R(OH)              | 1.355| 1.341 | 1.357 | 1.345 |
| R(OCH3)            | 1.363| 1.358 | 1.358 | 1.357 |
| R(O-CH3)           | 1.422| 1.422 | 1.419 | 1.422 |
| R(O18-H---N10)     | 1.884| 1.746 | 1.884 | 1.69 |
| R(O34-H---O12)     | 2.016| 1.86  | 1.875 | 1.93 |
| R(O34-H---N10)     | --  | --    | --    | --    |
| A(2-1-6)           | 115.8| 116.8 | 116.3 | 117.2 |
| A(1-2-3)           | 124 | 123.9 | 123.6 | 123.7 |
| A(1-6-5)           | 124.2| 124.2 | 124.6 | 124.1 |
| A(2-3-4)           | 119.3| 118.7 | 119.3 | 118.5 |
| A(3-4-5)           | 117 | 118 | 118.9 | 118.8 |
| A(3-4-7)           | 125.2| 123.4 | 117.2 | 117.7 |
| A(5-4-7)           | 117.8| 118.6 | 125.8 | 124.9 |
| A(4-5-6)           | 119.6| 118.4 | 117 | 117.4 |
| A(4-7-8)           | 122.1| 123 | 120.6 | 122.4 |
| A(4-7-9)           | 114.9| 114.9 | 117.2 | 115.3 |
| A(8-7-9)           | 123 | 122.1 | 122.2 | 122.3 |
| A(7-9-10)          | 117.9| 119.3 | 118.7 | 119.3 |
| A(9-10-11)         | 118.1| 119.4 | 116.3 | 119.8 |
| A(10-11-12)        | 120.6| 120.8 | 121.4 | 120.5 |
| A(11-12-13)        | 120.2| 120.5 | 119.4 | 120.6 |
| A(11-12-17)        | 121.3| 121.4 | 123.2 | 121.2 |
| A(13-12-17)        | 118.4| 118.1 | 117.4 | 118.2 |
Table 2. Assignment of electronic excitations for the molecules. Only selected transitions with enough oscillator strength around the main peak are included (H stands for HOMO and L for LUMO).

| Molecule     | Peak | Transition energy/eV (state) | Oscillator strength | Wave function (excitation amplitude) |
|--------------|------|------------------------------|---------------------|--------------------------------------|
| A(12-13-14)  | 1    | 3.272 (1)                    | 0.714               | H → L (0.99)                         |
| A(12-17-16)  | 2    | 4.232 (3)                    | 0.390               | H → L + 1 (0.95)                     |
| A(12-17-18)  | 2    | 3.376 (1)                    | 0.820               | H → L (0.99)                         |
| A(13-14-15)  | 2    | 4.186 (3)                    | 0.284               | H - 1 → L (0.71), H → L + 1 (0.66)   |
| A(14-15-16)  | 2    | 5.046 (10)                   | 0.198               | H - 3 → L (0.77), H - 4 → L (0.36), H - 5 → L (0.37) |
Figure 1. Electronic spectra of compound 1 & 2
Fig. 2. Molecular electrostatic potential of the compounds. 1 and 2
### Table 3: Crystal data and structure refinement Table

| Parameter                                           | Value                                    | Value                                    |
|-----------------------------------------------------|------------------------------------------|------------------------------------------|
|                                                     | **Compound 1**                           | **Compound 2**                           |
| CCDC deposit No.                                     | 1046722                                  | 1046723                                  |
| Empirical formula                                   | C_{14}H_{15}N_{3}O_{4}                   | C_{14}H_{15}N_{3}O_{4}                   |
| Formula weight                                       | 289.29                                   | 289.29                                   |
| Temperature                                          | 293(2) K                                 | 289.29                                   |
| Wavelength                                          | 1.54178 Å                                | 1.54178 Å                                |
| Crystal system, space group                         | Monoclinic, *P*2₁/C                      | Triclinic, *P*–1                        |
| Unit cell dimensions                                 |                                          |                                          |
|                                                     | *a* = 7.2054(2) Å                        | *a* = 6.4749(11) Å                      |
|                                                     | *b* = 12.4742(3) Å                       | *b* = 7.8787(14) Å                      |
|                                                     | *c* = 14.8390(3) Å                       | *c* = 14.395(3) Å                       |
|                                                     | *β* = 96.6560(10)°                      | *α* = 93.757(9)°                       |
|                                                     |                                          | *β* = 96.6560(10)°                      |
|                                                     |                                          | *γ* = 103.982(8)°                      |
| Volume                                              | 1324.76(6) Å                            | 689.8(2) 689.8(2)                       |
| Z, Calculated density                                | 4, 1.450 Mg/m³                           | 2, 1.393 Mg/m³                           |
| Absorption correction                               | Multi–scan                               | Multi–scan                               |
| Absorption coefficient                              | 0.907 mm⁻¹                               | 0.871 mm⁻¹                               |
| *F*(000)                                            | 608                                      | 304                                      |
| Crystal size                                        | 0.25 x 0.25 x 0.25 mm                    | 0.26 x 0.26 x 0.26 mm                    |
| Theta range for data collection | 4.64° to 64.27° | 3.17° to 64.30° |
| Limiting indices | $-8 \leq h \leq 6$ | $-8 \leq h \leq 7$ |
|                  | $-13 \leq k \leq 14$ | $-8 \leq k \leq 9$ |
|                  | $-16 \leq l \leq 16$ | $-16 \leq l \leq 16$ |
| Reflections collected / unique | 10721 / 2139 [R(int) = 0.0319] | 6324 / 2224 [R(int) = 0.0342] |
| Refinement method | Full-matrix least-squares on $F^2$ | Full-matrix least-squares on $F^2$ |
| Data / restraints / parameters | 2139 / 0 / 199 | 2224 / 0 / 199 |
| Goodness-of-fit on $F^2$ | 1.072 | 1.043 |
| Final R indices [$I>2\sigma(I)$] | $R_I = 0.0331$, $wR^2 = 0.0815$ | $R_I = 0.0431$, $wR^2 = 0.1217$ |
| R indices (all data) | $R_I = 0.0341$, $wR^2 = 0.0823$ | $R_I = 0.0499$, $wR^2 = 0.1283$ |
| Largest diff. peak and hole | 0.151 and $-0.195$ e. Å$^{-3}$ | 0.182 and $-0.162$ e. Å$^{-3}$ |
| Atoms    | Length     | Atoms    | Length     |
|----------|------------|----------|------------|
| N1–C2    | 1.3411(19) | C1–N6    | 1.331(2)   |
| N1–C6    | 1.3415(19) | C1–C2    | 1.363(2)   |
| C2–C3    | 1.3813(19) | C2–C3    | 1.379(2)   |
| C3–C4    | 1.3927(19) | C3–C4    | 1.376(2)   |
| C4–C5    | 1.3911(19) | C4–C5    | 1.389(2)   |
| C4–C7    | 1.5013(18) | C4–C7    | 1.492(2)   |
| C5–C6    | 1.384(2)   | C5–N6    | 1.334(2)   |
| C7–O8    | 1.2320(17) | C7–O8    | 1.2273(19) |
| C7–N9    | 1.3479(17) | C7–N9    | 1.350(2)   |
| N9–N10   | 1.3812(15) | N9–N10   | 1.3791(18) |
| N10–C11  | 1.2868(17) | N10–C11  | 1.286(2)   |
| C11–C12  | 1.4441(18) | C11–C12  | 1.444(2)   |
| C12–C13  | 1.4025(19) | C12–C17  | 1.399(2)   |
| C12–C17  | 1.4088(19) | C12–C13  | 1.405(2)   |
| C13–C14  | 1.3724(19) | C13–C14  | 1.368(2)   |
| C14–C15  | 1.3939(19) | C14–C15  | 1.388(2)   |
| C15–O19  | 1.3631(16) | C15–O19  | 1.3576(19) |
|        |        |        |        |
|--------|--------|--------|--------|
| C15–C16 | 1.3871(19) |        | C15–C16 | 1.385(2) |
| C16–C17 | 1.3903(19) | C16–C17 | 1.386(2) |
| C17–O18 | 1.3509(16) | C17–O18 | 1.3566(18) |
| O19–C20 | 1.4361(16) | O19–C20 | 1.419(2) |

**Table 5. Bond angles (°) Compounds 1 and 2**

|        |        |        |        |
|--------|--------|--------|--------|
| Compound 1 |        | Compound 2 |        |
| **Atoms** | **Angle** | **Atoms** | **Angle** |
| C2–N1–C6 | 117.01(12) | N6–C1–C2 | 123.65(16) |
| N1–C2–C3 | 123.83(13) | C1–C2–C3 | 119.32(16) |
| C2–C3–C4 | 118.61(13) | C4–C3–C2 | 118.93(14) |
| C5–C4–C3 | 118.13(12) | C3–C4–C5 | 117.18(15) |
| C5–C4–C7 | 118.32(12) | C3–C4–C7 | 125.80(13) |
| C3–C4–C7 | 123.52(12) | C5–C4–C7 | 117.01(14) |
| C6–C5–C4 | 119.04(13) | N6–C5–C4 | 124.60(16) |
| N1–C6–C5 | 123.29(13) | C1–N6–C5 | 116.32(14) |
| O8–C7–N9 | 123.93(12) | O8–C7–N9 | 122.19(15) |
| O8–C7–C4 | 121.52(12) | O8–C7–C4 | 120.61(14) |
| N9–C7–C4 | 114.48(11) | N9–C7–C4 | 117.20(13) |
| Bond             | Distance(Å) | Bond             | Distance(Å) |
|------------------|-------------|------------------|-------------|
| C7-N9-N10        | 119.36(11)  | C7-N9-N10        | 118.73(13)  |
| C11-N10-N9      | 115.62(11)  | C11-N10-N9      | 116.34(13)  |
| N10-C11-C12     | 121.97(12)  | N10-C11-C12     | 121.37(14)  |
| C13-C12-C17     | 117.68(12)  | C17-C12-C13     | 117.37(15)  |
| C13-C12-C11     | 119.50(12)  | C17-C12-C11     | 123.20(14)  |
| C17-C12-C11     | 122.81(12)  | C13-C12-C11     | 119.39(14)  |
| C14-C13-C12     | 122.07(13)  | C14-C13-C12     | 122.02(15)  |
| C13-C14-C15     | 119.03(12)  | C13-C14-C15     | 119.46(15)  |
| O19-C15-C16     | 124.20(12)  | O19-C15-C16     | 124.12(14)  |
| O19-C15-C14     | 114.82(12)  | O19-C15-C14     | 115.57(13)  |
| C16-C15-C14     | 120.97(12)  | C16-C15-C14     | 120.30(15)  |
| C15-C16-C17     | 119.39(12)  | C15-C16-C17     | 119.84(15)  |
| O18-C17-C16     | 117.50(12)  | O18-C17-C16     | 117.06(14)  |
| O18-C17-C12     | 121.64(12)  | O18-C17-C12     | 121.93(14)  |
| C16-C17-C12     | 120.86(12)  | C16-C17-C12     | 121.00(14)  |
| C15-O19-C20     | 117.68(10)  | C15-O19-C20     | 118.63(12)  |