Crystal Structure, Hirshfeld Analysis and DFT Study of 6,7-Dimethoxy-3-[(5-Chloro-1,3,3-Trimethylindolin-2-Ylidene)Methyl]Isobenzofuran-1(3H)-One

(Struktur Hablur, Analisis Hirshfeld dan Kajian DFT 6,7- Dimetoksi -3-[5-Kloro-1,3, 3-Trimetilindolin-2- ilidena] Isobenzofuran-1(3h)-one)

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ABSTRACT

The coupling reaction of 5-chloro-1,3,3-methylenindoline with 6-formyl-2,3-dimethoxy benzoic acid under solvent-free microwave irradiation gave of 6,7-dimethoxy-3-[(5-chloro-1,3,3-trimethylindolin-2-ylidene)methyl]isobenzofuran-1(3H)-one, C_{22}H_{22}ClNO_4 in high yield. The molecular structure consists of indoline system (C1-C8/N1) and isobenzofuran system (C10-C17/O1) connected by the enamine carbon. The title compound is crystallised in P2_1/n space group, Z’=4 with two independent molecules in centrosymmetric dimer. By using Hirshfeld surface and two-dimensional fingerprint plots, the presence of short intermolecular interactions and π-π stacking in the crystal structure were analysed. The DFT study is performed by comparing the calculation data with the X-ray data and analysing the HOMO and LUMO value, which gave -0.26952 a.u. and -0.17760 a.u., respectively.

Keywords: Crystal; DFT; Hirshfeld analysis; phthalide; synthesis

INTRODUCTION

Phthalides is a versatile compound that can act as building blocks in the synthesis of biologically active compounds such as anthracenes, naphthalenes, and naphthacenes natural product as well as displays wide spectrum of biological activities such as anticancer, antibacterial, antihypertensive activity, antiplatelet and anti-inflammatory. Phthalides are basically derived from the combination of a γ-lactone with benzene. It is also called as 3H-isobenzofuran-1-ones. The existence of phthalides in natural products began in the 18th century although it has been known for 100 years. Among the first phthalide products that can be considered as natural products are phthalide isoquinolines, hydrastine, or noscapine (Inubushi et al. 1964; Karmakar et al. 2014). One of the most common methods used in the preparation of phthalide is to use a catalyst where it is more economical, easy to use and does not produce harmful waste (Horváth 2003; Landge et al. 2008). Another method that gain attention nowadays is microwave assisted organic synthesis (MAOS) because it has proven to accelerate the reaction and give high yield of products (Ahyak et al. 2016; Sheryn et al. 2018). Since many reactions capable to be conducted in solvent-free conditions, microwave irradiation could be also counted as a ‘green’ alternative in synthesising organic compound (Ling et al. 2018; Rohadi et al. 2014). Herein, we report the crystal structure of phthalide from the condensation of Fischer’s base with aldehyde to produce phthalide (Figure 1) by using solvent free microwave irradiation method.

FIGURE 1. 6,7-dimethoxy-3-[(5-chloro-1,3,3-trimethylindolin-2-ylidene)methyl]isobenzofuran-1(3H)-one
MATERIALS AND METHODS

SYNTHESIS OF 6,7-DIMETHOXY-3-[(5-CHLORO-1,3,3-TRIMETHYLINDOLIN-2-YLIDENE)METHYL]-ISOBENZOFURAN-1(3H)-ONE

This compound was prepared by the reaction of 5-chloro-1,3,3-methylenindoline with 6-formyl-2,3-dimethoxy benzoic acid under solvent-free microwave irradiation. All the reagents were purchased from Sigma Aldrich and Acros and used directly without further purification. The crude product was crystallised by slow evaporation from acetone to obtain pure crystal (68% yield). All the characterisation including \(^1\)H and \(^13\)C NMR, Infra-Red and Mass Spectrometry are in agreement with Sheryn et al. (2018).

CHEMICAL AND INSTRUMENTATION

Analytical grade quality of reagent and solvents were used without further purification. The single crystal of the complex was mounted on glass fibre and measured on a Bruker SMART APEX CCD (Bruker 2002) area detector at 296K using graphite monochromated MoKα (\(\lambda = 0.717073 \, \text{Å} \)). The structure was solved by a direct method and refined by full-matrix least square on F2 using the SHELXTL program (Sheldrick 2008). All non-hydrogen atoms were refined anisotropically. Structural parameters calculation and graphic were determined by ORTEPIII (Burnett & Johnson 1996) and PLATON (Spek 2009).

THE HIRSHFELD SURFACE ANALYSIS

The various intermolecular interaction of the title compound was subjected to a Hirshfeld surface analysis and their fingerprint (FP) plots were generated using CrystalExplorer17 with standard surface resolution with the three-dimensional \(d_{\text{norm}}\) surfaces mapped over a fixed colour scale of -0.031 (red) to 2.139 (blue) a.u. (McKinnon et al. 2007). The contributions from various contacts, listed in Table 2, were selected by the partial analysis of the FP plots. The Hirshfeld surfaces and FP plots for the title compounds are discussed at appropriate points in the Results and Discussion section.

COMPUTATIONAL METHODS

The geometry of the compound has been fully optimised at the DFT level of theory. The DFT calculations were carried out with the density functional of Becke three-parameter Lee-Yang-Parr (B3LYP) using the standard 6-31G (d,p) basis set as implemented in GAUSSIAN09 (Becke 1993).

RESULTS AND DISCUSSION

MOLECULAR CONFORMATIONS

The title compound is crystallised in monoclinic space group P2\(_1\)/n, \(Z'=4\) with two independent molecules in centrosymmetric dimer. The bond lengths and angles are in the normal ranges. The dihedral angle between the indoline system (C1-C8/N1) and isobenzofuran system (C10-C17/O1) groups is 88.88° (8). The torsion angle along the atom C8-C9-C10-O1 is 110.80° (3). The five and six membered ring in the isobenzofuran and indoline system gave dihedral angle of 4.43° (13) and 1.37° (13), respectively (Figure 2). The present of dimethoxy group (O3-C22) and (O4-C21) at isobenzofuran system gave distorted of planarity with value -0.045° (2) and 0.083° (2), respectively.

The interaction between linking adjacent molecules formed a chain propagating along the \(b\)-axis direction with graph set analysis C(6) motif (Figure 3). The isobenzofuran in the molecule participates in \(\pi-\pi\) stacking interaction (Figure 4(a)) through the \(c\)-axis direction give centroid-centroid distance of 3.693 (3) Å, it can be shows with electrostatic potential diagram in the blue-red triangle shape (Figure 4(b)).

FIGURE 2. The molecular structure of the title compound with displacement ellipsoids drawn at the 50% probability level
The compound crystallised in P2₃/n space group, Z′=4 with two independent molecules in centrosymmetric dimer. Crystal data, data collection and structure refinement details are summarised in Table 1.

Computer programs: APEX3 and SAINT (Bruker 2016), SHELXT (Sheldrick 2015), SHELXL (Sheldrick 2015), SHELXTL (Sheldrick 2008), PLATON (Spek 2009), OLEX2 (Dolomanov et al. 2009) and pubICIF (Westrip 2010), Crystal Explorer (Turner et al. 2017), Mercury (Macrae et al. 2008).

A search of the Cambridge Structural database (Groom et al. 2016) showed 10 structures of 6,7-Dimethoxy-3-[(5-chloro-1,3,3-trimethylindolin-2-ylidene)methyl]isobenzofuran-1(3H)-one derivatives. Compound (Z)-3-(4-hydroxyphenyl)-2-thiaoxy-5-((E)-(E)-(1,3,3-trimethylindolin-2-ylidene)methyl)diazenyphenylbenzylidene)thiazolin-4-one isopropanol solvate hemihydrate has potential as NLO chromophores (Ashraf et al. 2012). Pyrrolidinium 2,2-difluoro-4-[2-(pyrrolidin-1-yl)-1-propenyl]-6-[3-(1,3,3-trimethylindolin-2-ylidene)-1-propenyl]-1,3,2-(2H)-dioxaborine-5-carboxylate effectively as agent of fluorescent amine probes (Gerasov et al. 2008). Compound 3-Ethoxy-4-(1,3,3-trimethylindolin-2-ylidenemethyl)cyclobut-3-ene-1,2-dione are chromophore in dye application (Yagi et al. 2002). Compound 1,7-bis(1,3,3-trimethylindolin-2-yl)-4-(3-(1,3,3-trimethylindolin-2-ylidene)-propenyl)heptamethinium tetrafluoroborate methylene chloride solvate (Allmann et al. 1982). Another more derivatives has been reported N-(1,3,3-trimethylindolin-2-ylidene)acetamide (Mekhai et al. 2002), 4-tert-buty1-2-[2-(1,3,3-trimethylindolin-2-ylidene)ethylidene]cyclohexanone (Gainsford et al. 2011), (1-Ethoxy-3-phenyl-4-(1,3,3-trimethylindolin-2-ylidene)but-2-enylidene)-pentacarbonyl-tungsten, 3-Phenyl-2-(1,3,3-trimethylindolin-2-ylidene)but-3-enal and (1-Ethoxy-2-(1-phenyl-2-methylprop-1-eny)-4-
(1,3,3-trimethylindolin-2-ylidene)but-2-en-1-ylidene)pentacarbonyltungsten (Göttker-Schnetmann et al. 2001) and 5-((1,3,3-Trimethylindolin-2-ylidene)methylimino)-5,6-dihydronaphtho(1'2':4,5)imidazo(1,2-a)pyrimidin-6-one hydrate (Laréginie et al. 1996). X-ray database show no report crystal of R indolen ring isobenzofuran system.

THE HIRSHFELD SURFACE ANALYSIS

The dnorm maps that shows interaction in fingerprint style were generated by internal (di) and external (de) (Figure 5) distances to the nearest nucleus calculation (Spackman & Jayatilaka 2009). The intense red spots near H14 Figure 6(a) and O2 Figure 6(b) which is represent the donor and acceptor of intermolecular interaction for C-H-O. This contribute to the 21.0% of O-H/H-O interaction in the 2D fingerprint plot with forceps like tips at de + di = 2.4 Å. The diminutive red spot at the H21A and C22 Figure 4a shows the short contact of C-H/H-C interaction which contribute 12.4% with di + dH = 2.2 Å. The fingerprint plot for the H-H contact make the largest contribution to the Hirshfeld surface with 47.4% and di + dH = 2.2 Å. Intermolecular interaction of Cl-H/H-Cl are characterised as the pair of spike with their tips at dH + dCl = 3.0 Å with percentage of contribution 8.2%, as for C-Cl/C1-C interaction with 5.2% contribution and fingerprint pointy style with di + dH = 1.8 Å. There is also presence of minor contact that contributed to the assembly of the title compound, summarized in Table 2.

TABLE 1. Crystallographic details

| Crystal data          |                  |
|-----------------------|------------------|
| Chemical formula      | C22H22ClNO4      |
| M                    | 399.85           |
| Crystal system, space group | Monoclinic, P2/n |
| Temperature (K)       | 299              |
| a, b, c (Å)           | 13.595 (7), 8.029 (4), 18.722 (11) |
| β (°)                 | 105.39 (2)       |
| V (Å³)                | 4                |
| Radiation type        | Mo Kα            |
| μ (mm⁻¹)              | 0.22             |
| Crystal size (mm)     | 0.55 × 0.38 × 0.30 |

Data collection

|                  |                  |
|------------------|------------------|
| Diffractometer   | Bruker PHOTON 100 CMOS |
| Absorption correction | Multi-scan, SADABS (Bruker 2016) |
| Tmin, Tmax       | 0.587, 0.746     |
| No. of measured, independent and observed [I > 2σ(I)] reflections | 60633, 4910, 3269 |
| Rint             | 0.100            |
| (sin θ/λ)max (Å⁻¹) | 0.668           |

Refinement

|                  |                  |
|------------------|------------------|
| R(F² > 2σ(F²)), wR(F²), S | 0.068, 0.174, 1.07 |
| No. of reflections | 4910            |
| No. of parameters  | 258             |
| H-atom treatment  | H-atom parameters constrained |
| Δρmax, Δρmin (e Å⁻³) | 0.23, −0.38 |

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DFT STUDY

DFT structure optimisation of title compound was compared with experimental value (Table 3). From the DFT study, the HOMO and LUMO value is -0.26952 a.u. and -0.17760 a.u., respectively, in which gave 0.09192 a.u. energy gap (Figure 7).

Freistas et al. (2016) and Srivastava et al. (2012) reported that their synthesised phthalide derivatives had an energy gap from 0.158-0.278 a.u. Meanwhile, the title compound has much smaller energy gap, 0.09192 a.u. compared to the reported phthalide derivatives. Thus, suggested that our compound has better or higher biological activity, as it has been reported that small energy gap can lead to high anti-oxidant activity and good inhibition corrosion efficiency (Bentiss et al. 2003; Sarkar et al. 2012).

As the energy gap is smaller, the condition of compound is actively in favour to add electrons to a high-lying LUMO and to remove electrons from a low-lying HOMO. Hence, it causes low kinetic stability and high chemical reactivity, and thus facile formation of activated complexes for any potential reactions (Aihara 1999).

LUMO sites were scattered over phthalide ring implying the susceptibility of the ring toward nucleophilic attack. On the other hand, the HOMO sites were distributed over the indoline group enabling the group to donate electrons and form bonds.
Figure 5. 2D fingerprint plot of the title compound

Figure 6. Hirshfeld surface interaction

Table 2. Percentage contribution of interatomic contacts to the Hirshfeld surface analysis of the title compound

| Contact   | Percentage contribution (%) |
|-----------|-----------------------------|
| H-H       | 47.4                        |
| O-H/H-O   | 21.0                        |
| C-H/H-C   | 12.4                        |
| Cl-H/H-Cl | 8.2                         |
| C-Cl/Cl-C | 5.2                         |
| C-O/O-C   | 1.3                         |
| N-H/H-N   | 1.1                         |
| C-C       | 2.7                         |
| C-N/N-C   | 0.3                         |
| Cl-Cl     | 0.3                         |
| O-O       | 0.1                         |

Conclusion

Compound 6,7-dimethoxy-3-[(5-chloro-1,3,3-trimethylindolin-2-ylidene) methyl] isobenzofuran-1(3H)-one exists as two independent molecules in centrosymmetric dimer in one unit cell. The theoretical and the crystallographic data has confirmed the importance of π-π interaction that relatively short [Cg-Cg = 3.693(3) Å] for stabilising the unit cell with chain propagating at motif of C(6). Hirshfeld analysis highlighted the intermolecular interaction of C-H-O, in which the highest contact contribution is O-H/H-O (21.0%), \(d_e + d_i = 2.4\, \text{Å}\) and C-H/H-C (12.4%), \(d_e + d_i = 2.2\, \text{Å}\). From DFT study, the HOMO and LUMO value is -0.26952 a.u. and -0.17760 a.u., respectively, in which gave 0.09192 a.u. energy gap.
Table 3. Comparison of selected observed (X-ray data) and calculated (DFT) geometric parameter (Å)

| Bond/Angle | DFT   | X-ray  |
|------------|-------|--------|
| C1-C3      | 1.764 | 1.741 (3) |
| O1-C10     | 1.477 | 1.477 (3) |
| O1-C11     | 1.364 | 1.356 (3) |
| O2-C11     | 1.214 | 1.200 (3) |
| O3-C17     | 1.360 | 1.361 (3) |
| O3-C22     | 1.435 | 1.431 (4) |
| O4-C16     | 1.360 | 1.369 (3) |
| O4-C21     | 1.419 | 1.420 (4) |
| N1-C6      | 1.397 | 1.391 (3) |
| N1-C8      | 1.391 | 1.391 (3) |
| N1-C18     | 1.450 | 1.437 (3) |
| C11-O1-C10 | 111.88 | 111.16 (18) |
| C17-O3-C22 | 116.4 | 115.3 (2) |
| C16-O4-C21 | 118.3 | 117.3 (2) |
| C6-N1-C8   | 111.5 | 111.29 (19) |

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