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Synthesis, structural determination, *Hirshfeld* surface analysis, 3D energy frameworks, electronic and (static, dynamic) NLO properties of *o*-Nitroacetanilide (*o*-NAA): A combined experimental and quantum chemical study

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ABSTRACT

o-Nitroacetanilide (***o*-NAA**), an organic crystal, was synthesized and fully characterized by means of single-crystal XRD, FTIR, UV–visible, ^{1}H and ^{13}C NMR spectroscopic techniques. Its molecular structure was optimized, the subsequent electronic properties, the spectroscopic spectra were quantified using quantum chemical computations by density functional theory calculations and compared with the experimental results. Furthermore, the Mulliken atomic charges were estimated andthe molecular electrostatic potential map was plotted in order to identify the chemical reactive sites. To study the nonlinear optical (NLO) activity of ***o*-NAA**, the electric dipole moment, the static and dynamic polarizability and hyperpolarizabilities were computed indicating that it could be considered as a good candidate for NLO applications. The hydrogen bonding and the non-classical intermolecular interactions were investigated by performing a *Hirshfeld* surface analysis and their contributions were analyzed *via* the 2D-Fingerprint plots, thus revealing that the crystal structure of the studied molecule is mainly built up of H…H, H…O/O…H, H…C/C…H and C…O/O…C close contacts. From the 3D-molecular energy frameworks analysis, the *lattice energy* of the compound was found to be − 95.15 kJ.mol^{-1}, and has exhibited stabilizing strong C—H…O, N—H…C, *lp*…*lp*, *lp*…π and π…π interactions.

1. Introduction

Acetanilide derivatives are considered as an important class of materials in both chemistry and pharmaceutical industry owing to their large biological applications as analgesic as well as antipyretic agents [1] for the reason that they are mostly metabolized in the human body into paracetamol [2]. Furthermore, their applications are mainly related to thermodynamic properties [3],peculiar spectroscopy and quantum vibrational [4], herbicide properties [5] and their corrosion inhibition properties for 304L stainless steel [6]. An increasing interest of organic materials uses as nonlinear optical NLO devices has been noticed during the last years [7–8] due to their very large second-order electric susceptibilities [9] and their potential applications in electro-optic modulation [10,11], frequency conversion [12,13] and THz wave generation [14].Thus, nitroacetanilide derivatives, namely m- and p-substituted ones were investigated both experimentally and theoretically for their interesting NLO properties [15–17]. It is worth to be noted that a comprehensive and detailed structural literature survey of *o*-nitroacetanilide (***o*-NAA**) in the structural database (CSD, Version 5.39 [18]) revealed only the existence of one hit [19] reported in 1986, two hits for its derivatives 4-chloro-2-nitroacetanilide (CSD refcode : ICUMAU [20]) and4′-Fluoro-2′-nitroacetanilide (CSD refcode: NELQIE [21]), in addition to six more hits for the three 4-methyl-2-nitroacetanilide polymorphs appearing twice in the literature [22–23]. Moreover,

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