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Physicochemical properties and structural studies of a new hybrid compound potassium nitrate-ninhydrin: promising material for nonlinear applications.

Sirine Hawech^{2,3}, Isabelle Ledoux-Rak³, Amel Ben Slimane² and Nasreddine Ennaceur^{1,2,3*}

¹*University of Technology and Applied Sciences-ALRustaq, Sultanate of Oman.*

²*Lab AMEE, University of Gafsa, Faculty of Sciences of Gafsa, Tunisia.*

³*Lab LMI, Alembert Institute, Ecole Normale Supérieure of Paris-Saclay, France.*

*Corresponding Author: Pr. Nasreddine Ennaceur

* Email: nasreddin.alnaser.rus@cas.edu.om

Tel: (+216) 76 21 10 24

Abstract:

In this study, we have succeeded in synthesizing a new hybrid material $C_{18}H_{12}KNO_{11}.H_2O$, whose asymmetric unit is composed of a molecule of ninhydrin and potassium nitrate. This crystal was obtained using the slow evaporation method at room temperature. The grown crystal was subjected to different characterization techniques, whose lattice structure was determined by X-ray diffraction on single crystal. It showed that the compound is crystallized in a monoclinic system with a Cm space group. As for EDAX analysis, it confirmed the presence of the different elements present in the compound. Besides, the thermal study (TGA) proved the stability of the crystal up to 240°C. The transparency of the crystal has been studied by the UV spectrum. Finally, second-order non-linear optical measurements were carried out using the Kurtz-Perry technique. It proved that the second harmonic generation efficiency of the crystal was found to be 0.22 times more than that of KDP. This result seems to be advocate for its use as a doubling material of frequency.

Key words: crystal growth, hybrid material, x-ray diffraction, nonlinear optic, DFT

1. Introduction:

Nowadays, the search for second order nonlinear optical materials is attracting much attention thanks to their uses in the field of data storage, optical switching, image reconstruction, optical communication [1-3] and medicinal application. Indeed, using SHG technique in nonlinear optics with lasers, it is possible to extend the scope of photodynamic cancer therapy [4]. Moreover, many research works have been interested in the growth of hybrid materials due to their high nonlinear optical coefficients and their good stability compared to others existing in the past. The hybrid materials were obtained by the combination of two matrices, the organic and inorganic matrices. The ninhydrin which belongs to the family of gem-dihydroxy compounds is an organic matrix that can dissolve in many solvents, such as water, acetone and ethanol. It is known as a versatile reagent

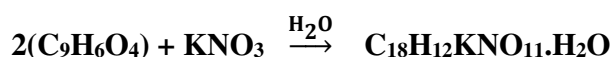
for the quantitative and qualitative determination of alpha-amino acids and proteins. The fields of using ninhydrin are various. An example of these fields is the chromatogram field in which ninhydrin reacts with acids and produces purple violet color. Besides, it can be helpful for food industry, microbiology, biochemistry, protein science and forensic science. Indeed, ninhydrin has aromatic rings with two hydroxyl groups attached to the same carbon. It allows many characteristics like the high nonlinear optical coefficient, high melting point and large band gap [5]. Ninhydrin crystallizes into a noncentrosymmetric crystal system $P2_1$ [6]. It also has a more important and efficient second harmonic generation than that of standard potassium dihydrogen phosphate (KDP) (2.5times higher than KDP) [7]. For this reason, it can be useful for making second harmonic generation devices out of the grown of other single crystals derived from this compound like the present work. In general, some organic crystals need to enhance their mechanical, thermal and nonlinear properties with other dopants because these properties are poor without combination with other metals [8, 9].

Therefore, this research work reports the bulk growth of a new hybrid material which is composed of the ninhydrin molecule and potassium nitrate. The inorganic matrix is a white crystalline salt that is soluble in ethanol and glycerol. Hence, the two mentioned matrices were mixed to obtain a crystal by slow evaporation method. This study will be supported by a quantitative and qualitative analysis of the presented elements. An X-ray diffraction study XRD is useful for a perfect description of the structure. The thermal stability is verified by a thermogravimetric analysis TGA. A high-added-value compound is revealed in its physical properties: nonlinear optics. To this end, we have carried out nonlinear optical measurements at 534 nm, which is reinforced by a theoretical calculation study of hyperpolarizability.

2. Experimental details:

2.1. Crystal growth:

An overview of the literature reveals that a great deal of research on the growth of aspects of ninhydrin has been conducted [10, 11] and the doped ninhydrin with many metals has been well developed. In this study we report the bulk growth of ninhydrin ($C_9H_6O_4$) and potassium nitrate (KNO_3) by slow evaporation solution method. The ninhydrin and potassium nitrate were mixed in a ratio of 2:1 using distilled water as a solvent. The prepared solution was stirred well using magnetic stirrer for 30 min. Then, it was filtered to remove the suspended impurities and allowed for slow evaporation. After a period of 30 days, we obtained homogeneous slightly whitish transparent plate crystals with good quality, according to the following reaction:



Which are used subsequently in different characterizations.

2.2. Characterization:

In order to see the suitability of the grown single crystal for its nonlinear optical properties, many analyses have been realized. To determine the basic composition of the crystal, we used an energy-dispersive X-ray (EDX) analysis using an analytical scanning electron microscopic (SEM) system (JOEL JSM -6380 LA). Furthermore, the grown crystal has been subjected to UV-visible spectrum using the Perkin Elmer Lambda 35 UV-vis spectrometer in the wavelength region 200-400 nm. Its structure was assessed by single crystal X-ray diffraction data using a Bruker Axs Kappa APEXII CCD area detector diffractometer. The thermal analysis (TGA) was determined using a Perkin Elmer DIAMOND with a heating rate of $10^\circ\text{C}/\text{min}$ in the temperature range from 30°C to 600°C in an air atmosphere. Finally, the second harmonic generation was performed by the Kurtz Perry technique with an Nd^{3+} : YAG nanosecond laser. All the results are presented and followed by detailed discussion in the sections that follow.

3. Results and discussion:

3.1. Energy-dispersive X ray analysis:

The energy-dispersive X-ray analysis is a very crucial technique to analyze the qualitative and the quantitative elemental percentages present in the single crystal. It is obvious that the typical peaks in the spectrum confirm the presence of various elements in the title compound which are carbon, oxygen, potassium and nitrogen (**Figure 1**).

The theoretical and experimental atom percentages are presented in the table below. A comparison of the results leads to the deduction that they are close except for the percentage of nitrogen, which confirms the validity of the proposed chemical formula (**Table 1**).

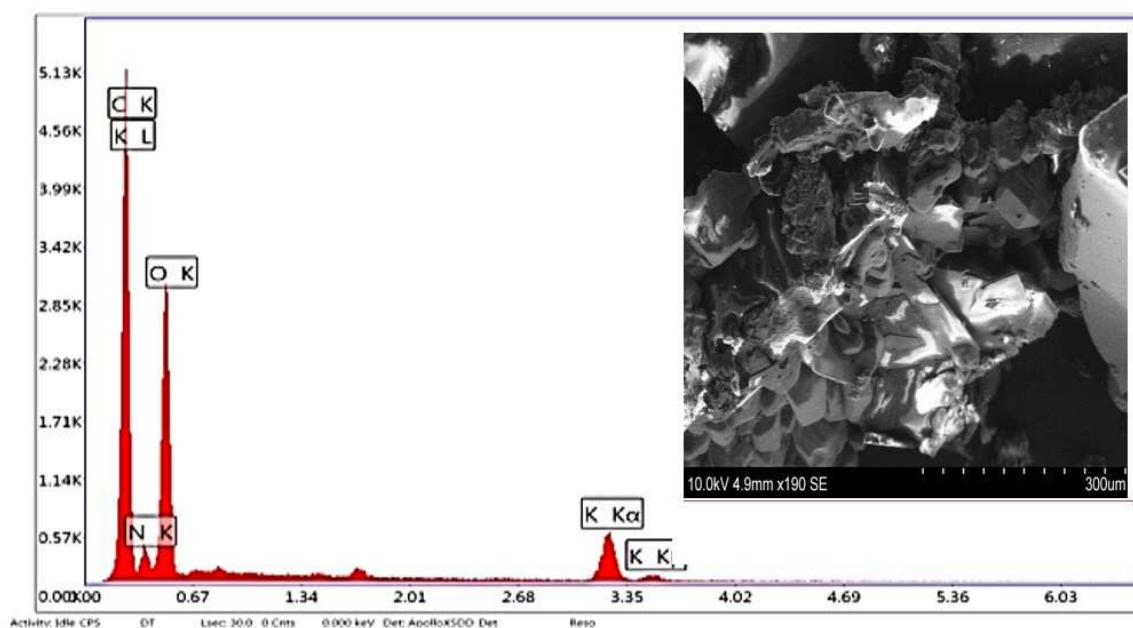


Figure. 1. Spectrum and scanning electron microscopy spots of potassium nitrate- ninhydrin single crystal.

Table1: EDAX analysis of the crystal

Element	Experimental	Theoretical
C	46.23	45.43
O	37.39	40.38
N	13.23	2.94
K	3.15	3.99
H	-	7.26
Total	100	100

3.2. Thermal analysis:

To determine the variation in the mass of a sample and its chemical properties such as decomposition, liberation of gas molecule, etc... thermogravimetric analysis (TGA) was performed. Furthermore, this technique is used to study the stability of the molecule in a precise temperature range controlled in an atmosphere of air at a heating rate of 10°C/min in the temperature range of 30-600°C. The initial mass which was 1.493 mg was put in a closed aluminum crucible with a drilled hole at the top. The TGA curve (**Figure 2**) reveals that there is a two-weight loss of about 7.8 and 11.3% in the temperature range of 100-188°C, respectively, which is due to the liberation of two and three water molecules. The next step is known by the beginning of decomposition of this crystal. In this process, the second weight loss of about 66% was observed at the temperature range of 188°C-238°C, which is due to the release of more CO₂. The decomposition point of the crystal was found at 238°C, which is different from that of pure ninhydrin found in the literature, namely 248.98°C [7]. Moreover, ninhydrin doped copper was found to be 249.81°C and that of ninhydrin doped cadmium was 254.31°C [12]. The values of

decomposition temperature for undoped and metal doped ninhydrin are in the same range. The difference is due to the occurrence of inorganic ions into the organic matrix.

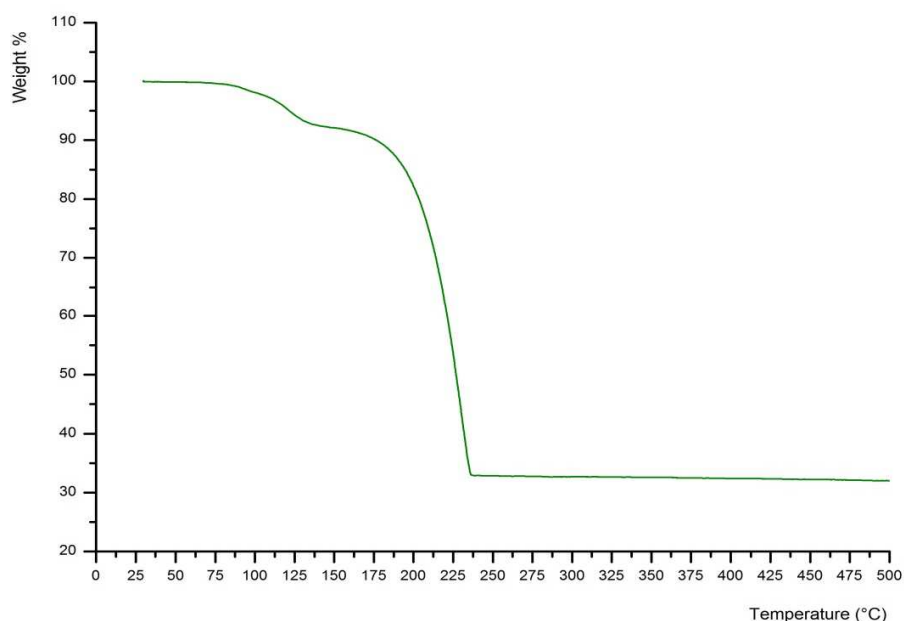


Figure.2: TGA thermogram of the $\text{C}_{18}\text{H}_{12}\text{KNO}_{11} \cdot \text{H}_2\text{O}$ crystal

3.3. UV –visible spectral analysis:

To identify the electronic transitions of the material, the UV-visible spectral analysis was performed. Indeed, we made use of the transmittance window or the absorbance to give information about these optoelectronic transitions. In the experimental absorption, the sample was put in liquid phase using ethanol as a solvent. Furthermore, the Perkin Elmer Lambda 35UV-visible spectrometer was used to study the UV-visible absorbance of the title compound. From the spectrum (**Figure 3**), it is obvious that there is no absorption in the visible region, which means that the crystal is transparent in that region and may be exploited for NLO applications at room temperature [13]. The two absorption bands centered at 230nm and 260nm are observed in the UV

region. In general, the organic materials have good nonlinear optical properties due to delocalized electrons $\pi-\pi^*$ and by doping these materials with the metal ions, the range of nonlinear optical applications will be large [14-18]. In fact, the organic material ninhydrin has important nonlinear optical properties due to delocalized electrons at $\pi-\pi^*$ orbitals. Furthermore, an intramolecular charge transfer emanates from the substitution of aromatic molecules with π - electron donors and acceptors [12], which enhances the response of our crystal to the optical field.

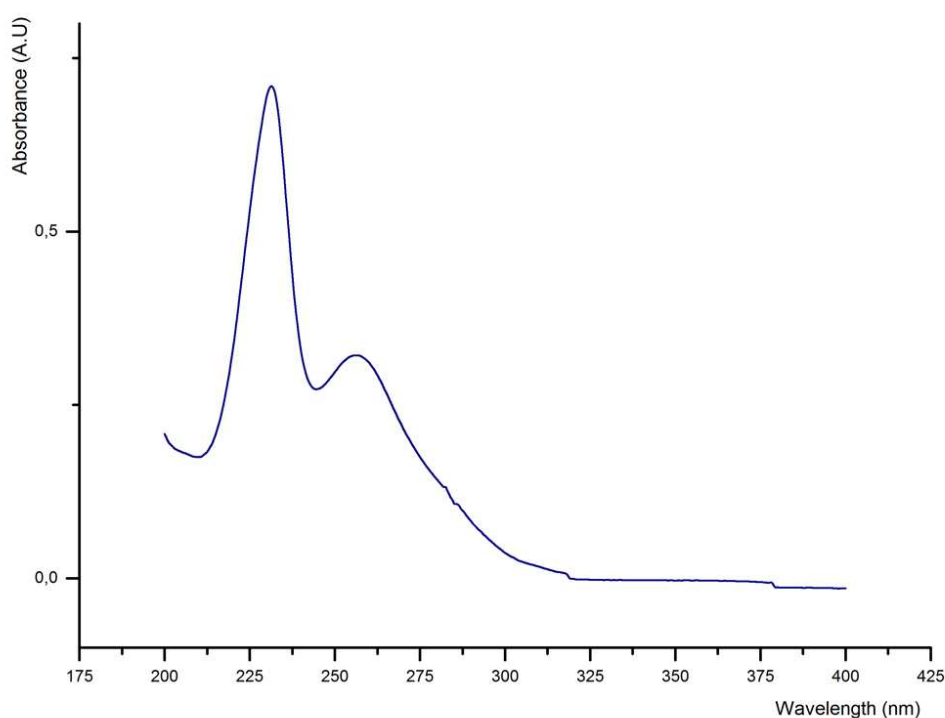


Figure.3: UV–visible spectrum of the $C_{18}H_{12}KNO_{11} \cdot H_2O$ crystal.

3.4. Single crystal X-ray diffraction:

We used the Burker Axs Kappa APEXII CCD to study the x-ray diffraction of the compound. The size of the crystal was $0.52 \times 0.3 \times 0.06 \text{ mm}^3$ and the temperature was 150.03 K. As for the resolution and the refinement of the structure, they were performed using the SHELXT 2014 [19] and SHELXL 2014 program [20], respectively. Moreover, the PLATON software was used to study

the structure solution and the refinement during the measurements. In order to determine the position of hydrogen, a Fourier difference was performed by using residual electronic densities. It is trusty to note that the crystal system was monoclinic with a C_m space group. Its lattice parameters were as follow: $a=7.9448\text{\AA}$; $b=20.8655\text{\AA}$; $c=5.7758\text{\AA}$ and $\alpha=\gamma=90^\circ$; $\beta=95.043^\circ$. The volume was found to be 953.76\AA^3 . These results are collected in **Table 2**.

Table 2: lattice parameters and structural refinement

Structure refinement	
Structure resolution	SHELXT and SHELXL
Refinement	based on F ²
Residual density (g.cm ⁻¹)	1.655
WR ₂	0.0763
R1	0.0310
Structural data	
Molecular formula	C₁₈H₁₂KNO₁₁.H₂O
Weight (g.mol ⁻¹)	475.40
Crystal system	monoclinic
Space group	Cm
a Å	7.9448
b Å	20.8655
c Å	5.7758
F (000)	488
V Å	953.76
Z	2
Conditions for collected intensities	
Diffractometer	Kappa Apex II
Radiation	Mo K α ($\lambda=0.71073$)
Absorption correction	multi -scan
Temperature (K)	150.03
hkl range	-10 \leq h \leq 10 ; -28 \leq k \leq 28 ; -7 \leq l \leq 7
Measured reflection	8575
Deposition Number- Cambridge Crystallographic Data Centre 2045358	

3.4.1. Description of the structure:

As shown in **Figure 4**, the asymmetric unit of the crystal is composed of two matrices, one of which is organic, revealed by the presence of one molecule of ninhydrin, and the other one is inorganic, revealed by the presence of potassium nitrate. The hydrogen and electrostatic bonds ensure the stability and the cohesion of the structure. Indeed, this structure can be described as being a three-dimensional network devised into two layers.

The inorganic part is composed of potassium nitrate. The KNO_3 is composed of the potassium, Nitrogen and 3 atoms of oxygen occupying an irregular tetrahedron (pyramidal form) outlined by angles and distances presented in Table 3. The values on the diagonal represent the N-O distances while those above the diagonal exhibit the symmetrical values of the angles O-N1-O. It is obvious from **Figure 5** that the anions are deployed in pairs and in zigzag along the b axis at 0 and $\frac{1}{2}$ to form anionic layers that are inserted between the cationic network.

Figure 6 shows the potassium cation forming an octahedral environment with the oxygen atoms, which is inserted between the ninhydrin and nitrate molecules in the b, c planes at $b = 0$ and $\frac{1}{2}$ and $c = \frac{1}{4}$. The organic matrix is composed of two molecules of protonated ninhydrin $(\text{C}_{18}\text{H}_{14}\text{O}_{11})^+$. Each molecule of ninhydrin is an aromatic molecule with two hydroxyl groups attached to the same carbon atom. An alternation of the long and short bonds is due to the electronic delocalization and a π -conjugated system. The distances separating them are O1-C1 (1.395 Å (2)), O2-C1 (1.395 Å (2)), O3-C2 (1.210 Å (3)) and O4-C5 (1.208 Å (3)). However, the C-C distances range from 1.385 Å (3) to 1.542 Å (3). Thus, the angles O-C-C, the C-C-C and the O-C-O angles are around 104.17 ° and 128.8 °. The organic cations are arranged in sandwich between the channels of anions developed in zigzag along a axis at $b=0$ and $\frac{1}{2}$.

Table 3: the N-O distances and the O-N-O angles

N1	O5	O6	O7
O5	1.277	117.6	120.6
O6	117.6	1.245	121.8
O7	120.6	121.8	1.222

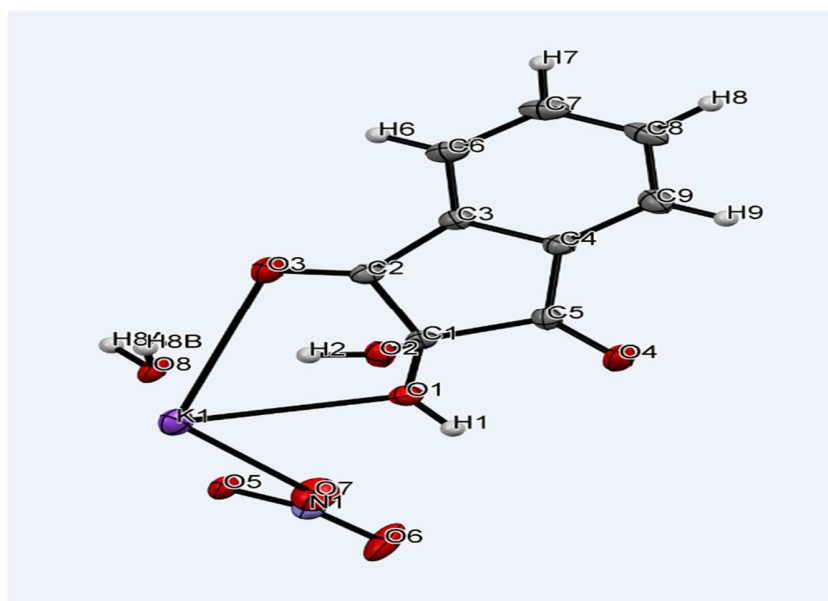


Figure.4: The asymmetric unit of the crystal

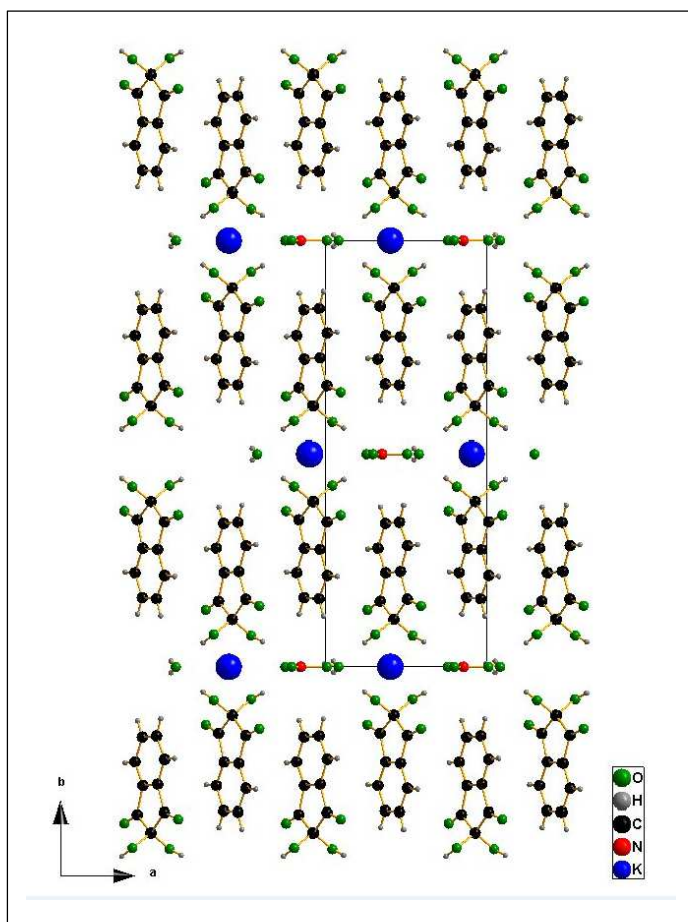


Figure.5: The projection of the structure along the (a, b) axis

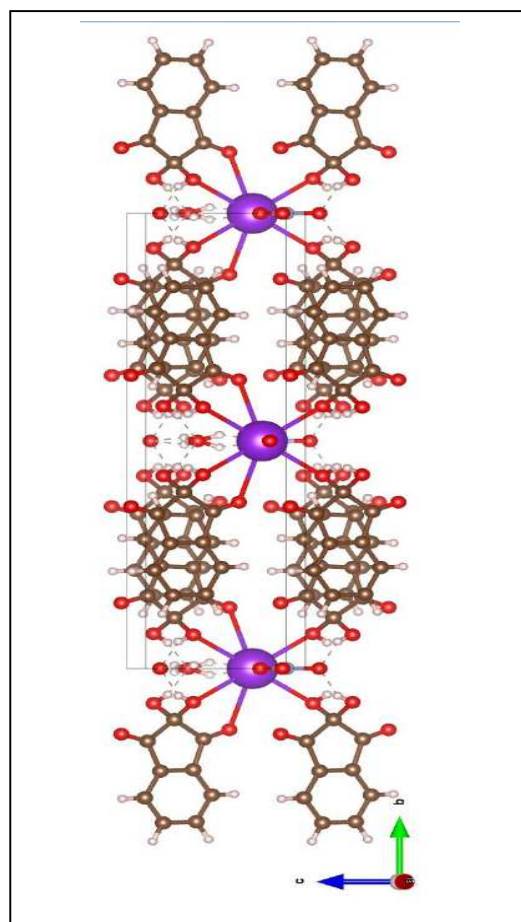


Figure.6: The projection of the structure along the (b, c) axis

3.5. Second harmonic generation

Kurtz and Perry powder technique is well known in the field of nonlinear optics [21]. The SHG measurements were performed by the Q-switched Nd:YAG laser operating at 1064 nm and pulse width 8ns with a repetition rate of 10 Hz [22]. The experimental method is based on the crush of the crystal and the reference (KDP), which are then put into sandwiches. After the alignment of the system optically, the samples were irradiated by a laser beam of 1064 nm wavelength. Then, an

oscilloscope revealed the electric signal indicating the SHG efficiency. The emission of a green light is an indicator for the presence of the second harmonic generation, as shown in **Figure 7**, in which the SHG response of the crystal and the reference KDP is obtained. The two intensities were measured and compared at 532nm. To calculate the susceptibility of the compound, we refer to the following equations:

$$\text{slope}_{\text{sample}} = \frac{I_{\text{sample}}^{2W}}{I_{\text{NPP}}^{2W}} \times \frac{(X_{\text{sample}}^2)^2}{(X_{\text{NPP}}^2)^2}$$

$$\text{Slope}_{\text{KDP}} = \frac{I_{\text{KDP}}^{2W}}{I_{\text{NPP}}^{2W}} \times \frac{(X_{\text{KDP}}^2)^2}{(X_{\text{NPP}}^2)^2}$$

$$X_{\text{sample}}^2 = \sqrt{\frac{\text{slope}_{\text{sample}}}{\text{slope}_{\text{KDP}}}} \times X_{\text{KDP}}^2$$

where χ^2 is the effective susceptibility; slope of the sample=4.75 and slope of KDP=5.71

$$\chi_{\text{sample}}^2 = \sqrt{\frac{4.75}{5.71}} \times 0.44 = 0.40 \text{ pm/v}$$

The SHG efficiency of the crystal is 0.20 times that of KDP. Indeed, some aromatic molecules are characterized by a large non-linearity because of the nature and the length of π -conjugated chain. Besides, the presence of asymmetrical charge transfer process influences the efficiency of the NLO response [23]. Furthermore, due to the difference of electronegativity between the end groups, an area of charge transfer from donor to acceptor is created [24]. The values of hyperpolarisability β , which are interesting, especially in organic materials, represent one of the important reasons for the high magnitude of the SHG [25-27]. In the present case, the high response of the title compound is probably due to the incorporation of the metal ions as the dopants in the lattice of the crystal. The electron-phonon interaction between the nitrate of potassium and the phonon subsystems are responsible for the enhancement of the SHG efficiency [28]. Moreover, the hydrogen bonds, which play an important role in the crystal packing, ensure the stability of crystal. They also increase the

charge distribution between the two rings with the potassium nitrate anions [29] without neglecting the role of parallel stacking and π - π interaction to enhance the second harmonic generation efficiency of the nonlinear materials [30].

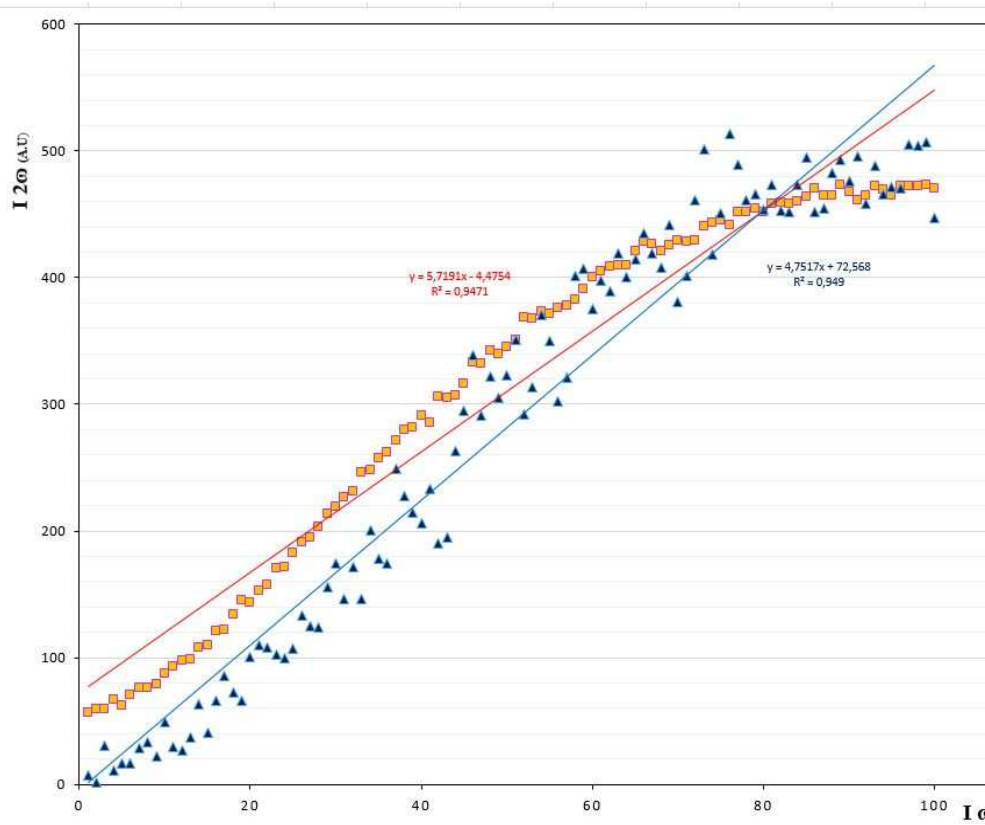


Figure.7: Power dependence of $I_{2\omega}$ vs I_{ω}^2 of the title compound crystal and KDP

3.6. Hyperpolarisability calculations:

The hyperpolarisability calculation gives an indication about the nonlinear optical response of the title compound. It is also used to inform about the links between the structure of the crystal and their nonlinear properties. Indeed, the nonlinear optical response can be presented as a Taylor series expansion of the total dipole moment μ in an electric field.

$$\mu = \mu_0 + \mu_i E_i + \beta_{jk} E_j E_k + \dots$$

where α is the linear polarisability, μ_0 is the permanent dipole moment and β_{ijk} are the first hyperpolarisability tensors components [31,32].

The calculations of polarizability α_0 and first-order hyperpolarizability were realized through the system B3LYP/6-31(d) level basis set [33]. Furthermore, the two equations mentioned below are used to calculate the isotropic linear polarizability and the first hyperpolarizability α_{tot} and β_{tot} , respectively [34, 35].

$$\alpha_{tot} = 1/3 (\alpha_{xx} + \alpha_{yy} + \alpha_{zz})$$

$$\beta_{tot} = \sqrt{(\beta_{xxx} + \beta_{xyy} + \beta_{xzz})^2 (\beta_{yyx} + \beta_{yxx} + \beta_{yzz})^2 (\beta_{zzx} + \beta_{zxx} + \beta_{zyy})^2}$$

After calculations, the values are presented in atomic unit (a.u), which are converted into electrostatic unit (e.s.u) by this transformation $\alpha.1a.u=0.1482.10^{-24}$ esu and $\beta.1a.u = 8.6393.10^{-33}$ esu.

As seen in **Table 4**, the polarizability α_{tot} of the compound is $3.34321.10^{-23}$ esu and the largest value of α_{zz} ($5.33605.10^{-23}$ esu) shows that the molecule is strongly polarized along the Z direction. Besides, the calculated value β_{tot} is evaluated as $1.74231.10^{-28}$ esu, which gives information about the direction of charge delocalization or polarization. It is also obvious that this value is higher than that of the reference crystal KDP and urea 254 times and 467 times, respectively ($6.85.10^{-31}$ for KDP and $3.728.10^{-31}$ for urea). This result means that the title compound is a potential candidate for nonlinear optic. In the same context, the highest value of the first hyperpolarisability is in the order of $1.75655.10^{-28}$ esu and it is dominated by the diagonal component (YYY). Therefore, the domination of this component shows that while there is a substantial delocalization of charges in this direction, the other directions show close values. It can be deduced that there are two types of charge transfer, namely intramolecular and intermolecular, caused by the electron cloud movement.

Furthermore, it is possible now to refer to the value of β_{tot} to determine the source of nonlinear optical response, which is the electronic delocalization in the different directions between the two rings of the molecule and the role of hydrogen bonds as being kind of links with the potassium nitrate anions. Hence, the title compound will be a good material for NLO applications.

Table 4: Calculated polarisability (α) and hypperpolarisability (β) of the crystal

components	Values (esu)	Components	Values (esu)
α_{xx}	$4.92114.10^{-23}$	β_{xxx}	$2.62109.10^{-29}$
α_{xy}	$2.1896.10^{-24}$	β_{xxy}	$-1.05172.10^{-29}$
α_{yy}	$-2.2755.10^{-24}$	β_{xyy}	$1.62327.10^{-29}$
α_{xz}	$8.04554.10^{-24}$	β_{yyy}	$1.75655.10^{-28}$
α_{yz}	$9.19832.10^{-25}$	β_{xxz}	$-5.36273.10^{-30}$
α_{zz}	$5.33605.10^{-23}$	β_{xyz}	$5.26592.10^{-30}$
α_{tot}	$3.34321.10^{-23}$	β_{yyz}	$-1.46944.10^{-29}$
		β_{xzz}	$1.85277.10^{-30}$
		β_{yzz}	$1.32881.10^{-30}$
		β_{zzz}	$-6.07908.10^{-30}$
		β_{tot}	$1.74231.10^{-28}$

Conclusion:

The title compound was grown using slow evaporation solution technique at room temperature. It was found to crystallize in the monoclinic system with a Cm space group. Its structure was determined using X-ray analysis. This technique showed that the asymmetric unit contains two parts, the organic part and the inorganic one which are linked by hydrogen bonds and π - π interactions. Besides, an alternation in sandwich between the anionic and cationic layers was observed. The presence of the different chemical elements was assessed by EDX analysis. Moreover, the title compound's stability up to its melting point was identified using the TG analysis. This study proved the stability of the crystal up to 240°C. The UV-visible confirmed that the crystal is transparent in the entire visible region. As for the SHG measurements, they have been carried out and the SHG efficiency was found to be about 0.22 times that of the KDP. This is due to the different kinds of electronic delocalization along the semi organic crystal. Thus, the quantum chemical calculations provided information about the direction of charge delocalization or polarization. Furthermore, the value of the first-order polarizability (β) was found to be $1.74231 \cdot 10^{-28}$ esu, which is approximately 254 times and 467 times that of KDP and urea, respectively. This represents an encouraging result to classify the crystal of ninhydrin and nitrate of potassium as a good material for nonlinear applications.

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