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Growth, physico-chemical properties, opto-electrical characteristics, thermo-mechanical and DFT studies of 4-aminoantipyrine single crystals

S. Dinagaran [a], J. Gajendiran [b], S. Gokul Raj [c], S. Gnanam [d]

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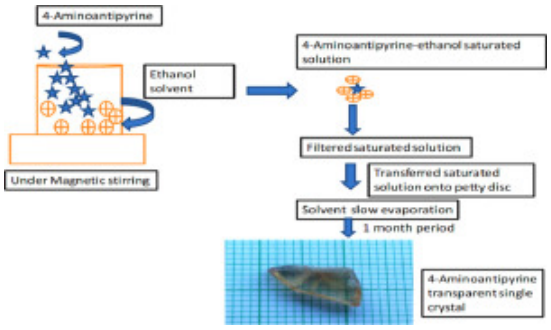
Abstract

The aim of this work was to grow organic single crystals of 4-Aminoantipyrine (4-AAP) under ambient conditions, using a solvent slow evaporation method, and then to investigate the crystal structure, optical band gap, optical transmittance behavior, second harmonic generation efficiency, thermal stability, material rigidity, and dielectric responses for opto-electrical based device applications. The powder X-ray diffraction (XRD) and single crystal XRD analyzes revealed the hexagonal crystal structure and lattice parameter values of the as-grown 4-AAP. The respective molecular vibration bands and functional groups of the as-grown 4-AAP were identified using the experimental and theoretical FT-IR analysis. The optical absorption and transmittance spectrum of the 4-AAP single crystals revealed good transparency throughout the visible region. The non-linear optical (NLO) characteristics of the 4-AAP crystals were tested using the Kurtz and Perry powder technique. Thermal gravimetric (TGA) and differential thermal analysis (DTA) were used on the as-grown crystals, to determine their weight loss and heat absorption/release process. The hardness vs load plot can be used to determine the hardness values and response of the hard as-grown 4AAP crystals under the influence of load. The electrical conductivity and dielectric behavior of the 4-AAP crystals changed as the applied frequency increased. Computational techniques were used to investigate the 4-AAP's optimized geometrical structure, theoretical optical band, molecular electrostatic potential, and Mulliken atomic charge population.

Graphical abstract



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Introduction

In recent years, some bioactive chemicals containing 4-aminoantipyrine, azithromycin, acriflavine keto-bromothymol blue, thiobarbituric acid, and carboxymethyl cellulose have been widely introduced in Schiff based derivatives, metal complexes, metal oxide and composite material compounds for potential applications such as biomedicine, non-linear optics- based devices, antibacterial activity, photocatalyst treatment, solar cells, optoelectronic devices, and removal of pollutants from the textile industry [1], [2], [3], [4], [5], [6], [7], [8], [9], [10], [11], [12], [13], [14], [15]. They have a good ability to react, which is followed by ease of attachment with the transition metal, Schiff based derivatives, and metal oxide complexes, resulting in tremendous unique bio-chemical properties, and optical and electrical characteristics that are non-toxic. There have been very few reports on the crystalline parameters, antibacterial, cytotoxic effects, and vibrational modes of the 4-AAP crystal [1], [2], [3], [4], [5], [6], [7]. More attention needs to be given to the NLO behavior and electrical properties of the 4-AAP crystal for testing its suitability to be applied to electro-optics based-devices.

There have recently been many web reports on single crystal's growth preparation using melt growth [16,17], Bridgman method [18], solid state reaction [19], Sankaranarayanan-Ramasamy method [20], solution growth and solvent slow evaporation at room temperature [21], [22], [23], [24], [25], [26], [27]. The optical absorption, transmittance behavior, and structural, vibrational, thermo-electrical, and mechanical properties of single crystals have been investigated in the above web reports. Among the methods, solution growth [21], [22], [23], [24], [25], [26], [27] has some merits, such as the lack of the need for special instruments, the elimination of time-consuming chemical synthesis steps, and the elimination of the need for high temperature reaction processes to grow single crystals with good optical transmission behavior.

In this current work, 4AAP single crystals are successfully grown using the slow solvent-evaporation method, without the aid of any special instruments or high temperature reaction processes, and their structural, optical, electrical, and thermo-mechanical properties are tested. The significance of this research work lies in the following characteristics: (i) From the UV-visible absorption spectrum, the

optical band gap value of the grown crystal is determined to be 4.42 eV, and the results of the broad band gap would imply that the grown crystal has good insulating properties. (ii) Based on the transmittance spectrum, the grown crystal revealed 92% transmittance. The detected transmittance values are appropriate for NLO based devices. (iii) Green colour emission radiation is observed after testing the laser SHG measurement analysis for the grown 4-AAP crystal, and the SHG efficiency is determined. Furthermore, the obtained SHG efficiency values are compared with those of the standard reference potassium dihydrogen phosphate (KDP) and urea crystal. (iv) The 4-AAP crystal is moderately soft material, according to Vicker's hardness test. (v) The grown crystal has been tested via dielectric studies, and the results show that it has a lower dielectric constant and dielectric loss, resulting in less imperfection and good optical responses.

Section snippets

Experimental

In a typical synthesis, 4-aminoantipyrine (4AAP) was grown under ambient conditions by the solvent slow evaporation method, using 4-aminoantipyrine salt ($C_{11}H_{13}N_3O$, Sigma-Aldrich (98%)) and ethanol (CH_3-CH_2-OH, Sigma-Aldrich (99.5%)) as the solvent medium. Fig. 1 depicts a schematic diagram of the steps for growing 4-aminoantipyrine single crystals.

Fig. 2a depicts the ORTEP diagram of the 4-aminoantipyrine molecule. It is a general view of the 4-aminoantipyrine molecule with an atomic numbering …

Single crystal X-ray diffraction analysis

The as-grown crystal was subjected to single crystal XRD analysis to determine the cell parameters given in Table 1. The derived grown crystal lattice parameters values from Table 1 were compared with values of the 4-AAP crystal's lattice parameters reported by Liu et al. [2], which confirmed that the grown crystal is hexagonal in structure with a space group $P6_1$ [28].

The calculated lattice parameters and cell volume of the 4-AAP crystals have a standard deviation error of ± 3%.

Using the single …

Conclusions

Using a solvent slow evaporation method, organic single crystals of 4-AAP were grown from a synthesized 4-AAP solution. The single crystal XRD and powder XRD analysis confirmed the hexagonal crystal structure of 4-AAP. Various molecular vibrational modes for the obtained 4-AAP were identified using the experimental and theoretical FT-IR spectra. The optical absorption plot revealed a larger energy gap (E_g=4.42 eV) value for the grown 4-AAP, indicating that the grown crystal has good…

CRediT authorship contribution statement

S. Dinagaran: Methodology, Writing – original draft. **J. Gajendiran:** Investigation, Writing – original draft, Writing – review & editing, Conceptualization. **S. Gokul Raj:** Methodology, Investigation, Writing – original draft, Writing – review & editing. **S. Gnanam:** Investigation, Writing – original draft, Writing – review & editing.…

Declaration of Competing Interest

The authors declare that they have no known competing financial interest or personel relationship that could have appeared to influence the work reported in this paper.…

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Recommended articles

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