



## Benzimidazolium picrate single crystal: Synthesis, growth, characterization and their biological activity

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Knowledge on charge-transfer complexes of drugs is important to understand the drug-receptor binding mechanism. Here, we have been synthesized the organic charge transfer complex of Benzimidazolium picrate (BP) and confirmed its molecular structure and carbon skeleton by <sup>1</sup>H and <sup>13</sup>C NMR spectral analysis. Asymmetric unit of BP consists of one benzimidazolium cation and a monovalent anion of picric acid. Decomposition pattern and thermal stability BP have been investigated using TG/DTA analysis. The interaction of the compound with calf thymus DNA is investigated by electronic absorption spectroscopy which indicates that the compound could interact with DNA through intercalation. The CT complex demonstrated significant free radical scavenging with 2,2-diphenyl-2-picryl-hydrazyl. The BP complex has been also screened for its antimicrobial activity.

**Keywords:** Benzimidazolium picrate, Charge-transfer complex, 2,2-Diphenyl-2-picryl-hydrazyl

All heterocyclic compounds have high significance in pharmaceutical chemistry. The benzimidazole moiety is bicyclic, consisting of fusion of benzene and imidazole, and is widely used in designing new drugs<sup>1</sup>. The benzimidazole derivatives represent one of the most biologically active class of compounds, possessing a wide spectrum of activities<sup>2,3</sup>. The benzimidazole nucleus has significance in medicinal chemistry, and many benzimidazole-containing compounds exhibit important biological activities viz. selective neuropeptide YY1 receptor antagonists, 5-lipoxygenase inhibitors for use as novel anti-allergic agents<sup>4</sup>, factor Xa (FXa) inhibitors<sup>5</sup>, poly (ADP-ribose) polymerase (PARP) inhibitors<sup>6</sup> and as human cytomegalovirus (HCMV) inhibitors<sup>7</sup>. The structural resemblance of benzimidazole nucleus to biological compounds, such as the purine base of the DNA and the occurrence of this nucleus in vitamin B12 are of interest in pharmaceutical industry<sup>8-12</sup>. Benzimidazole constitutes an important group of electron donor or proton acceptor and the study of their charge transfer complex can help elucidate many chemical phenomena<sup>13-17</sup>.

Proton or charge transfer complexes play an important role in the field of magnetic, electrical

conductivity and optical properties<sup>18-22</sup>. The crystalline charge transfer complexes (CTCs) are known to have a vital role in biological systems such as antimicrobial activity and DNA-binding as well as in laser technology, optoelectronics, optical communications, photocatalyst, optical signal processing and dendrimer fabrication for drug delivery systems<sup>23-32</sup>.

CT-interactions between aromatic electron acceptors and electron donors containing nitrogen, oxygen or sulphur atoms have been reported over the previous year<sup>33-36</sup>. Picric acid forms crystalline picrate salts with various organic molecules by virtue of its Lewis acid behaviour and serve as a better acidic ligand in the formation of salts through specific electrostatic or hydrogen bonding interactions<sup>37</sup>. Hence benzimidazole acts as the electron donor and picric acid acts as electron acceptor. Based on the above significance, sincere attempts were made for the successful synthesis of BP crystal. The current research work focuses on the UV-absorption and NMR characterisation, single crystal X-ray diffraction study of the BP crystal. In addition the DNA binding, antimicrobial activity and cleavage activities of the synthesised crystal were also investigated in detail.

## Materials and Methods

All the chemicals used were chemically pure and are AR grade. Solvents were purified and dried according to the standard procedures<sup>38</sup>. Calf-thymus (CT-DNA) was purchased from Genei, Bangalore, India. Tetracycline, Nystatin and Agar were purchased from Hi-Media, Mumbai.

### Physical characterisation techniques

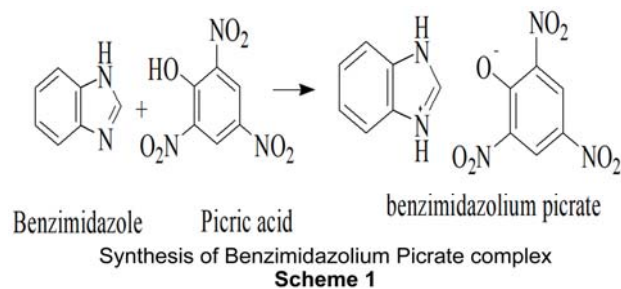
Micro analyses (C, H and N) were performed on a Vario EL III CHNS analyzer at STIC, Cochin University of Science and Technology, Kerala, India. The IR spectrum was recorded employing KBr pellet method in the region of 400–4000  $\text{cm}^{-1}$  using a PerkinElmer FTIR 8000 spectrophotometer. Electronic spectrum was recorded in methanol solution with a Systronics double beam UV–visible spectrophotometer 2202 in the range 200–800 nm.  $^1\text{H}$  and  $^{13}\text{C}$  NMR spectra were recorded on a Bruker AV III 500 MHz instrument using TMS as an internal reference at SAIF, Indian Institute of Technology, Madras. Melting points were recorded with Veego VMP-DS heating table and are uncorrected. The thermal analysis (TG and DTA) were carried out under nitrogen atmosphere with a heating rate of 20°C/min by NETZCHSTA 409C analyzers. DNA cleavage study was carried out using Gelstan - Gel documentation system. Antioxidant study was carried out at the Kovai Medical Centre and Hospital Pharmacy College, Coimbatore, Tamil Nadu, India.

### X-ray crystal structure determination

The crystallographic data of the complex has been measured at 298 K on a Bruker SMART APEX CCD, area detector system [ $\lambda$  (MoK $\alpha$ ) = 0.71073 Å]. A graphite monochromator, 2400 frames was recorded with an  $\omega$  scan width of 0.3°, each for 10 s, crystal-detector distance 60 mm, collimator 0.5 mm. Data reduction was carried out by SAINPTUS<sup>39</sup> and absorption correction was made using an empirical method SADBS<sup>40</sup>. The structure was studied and refined using SHELXL-97<sup>41</sup>. All non hydrogen is refined anisotropically. Cambridge Crystallographic Data Centre (CCDC No.1019637) contains the supplementary crystallo-graphic data for the complex.

### Synthesis of Benzimidazolium picrate (BP) complex

Equimolar ratio of benzimidazole in methanol and picric acid in water were dissolved and mixed together. The solution was stirred well for about 2 h at room temperature using magnetic stirrer to ensure homogeneous mixture, filtered using Whatmann 41



filter paper and kept aside unperturbed in a dust free room for the growth of single crystal. A good transparent needle shaped crystals was harvested in a growth period of 25-30 days. The purity of the synthesized crystal was further improved by repeated recrystallization process using methanol as solvent. (Scheme 1)

### Antioxidant behaviour

The 2,2-diphenyl-2-picryl-hydrazyl radical scavenging activity of the compound was measured according to the method of Elizabeth & Rao<sup>42</sup>. The DPPH is a free radical having a stable  $\lambda_{\text{max}}$  at 517 nm. A fixed concentration of the experimental compound (100  $\mu\text{L}$ ) was added to a solution of DPPH in methanol (0.3 mM, 1 mL) and the final volume were made up to 4 mL with double distilled water. DPPH solution with methanol was used as a positive control and methanol alone acts as blank. The solution was incubated at 37°C for 30 min in darkness. The decrease in absorbance of DPPH was measured at 517 nm. The tests were run in triplicate and various concentrations (20-100  $\mu\text{g}/\text{mL}$ ) of the compounds were used to fix a concentration at which the compounds showed 50% activity. In addition, the percentage of activity was calculated using the formula, % of suppression ratio =  $[(A_0 - A_c)/A_0] \times 100$ . Where,  $A_0$  and  $A_c$  are the absorbance in the absence and presence of the tested compounds, respectively. The 50% activity ( $\text{IC}_{50}$ ) can be calculated using the percentage of activity.

### DNA binding experiments

The binding affinity of the compound with CT-DNA was carried out in double distilled water with tris(hydroxymethyl)-aminomethane (Tris, 5 mM) and sodium chloride (50 mM). The pH was adjusted to 7.2 using hydrochloric acid. A result of CT-DNA in the buffer gave a ratio of UV absorbance with reference to 1.8-1.9 at 260 and 280 nm, indicating that the DNA was suitably free of protein. The DNA concentration per nucleotide was determined by absorption spectroscopy using the molar extinction coefficient

value of  $6600 \text{ dm}^3 \text{ mol}^{-1} \text{ cm}^{-1}$  at 260 nm. The compound was dissolved in a mixed solvent of 5% DMSO and 95 % tris HCl buffer for all the experiments. Stock solutions were stored at  $4^\circ\text{C}$  and used within 4 days. Absorption titration experiments were performed with a fixed concentration of the compound ( $25 \mu\text{M}$ ) with varying concentration of DNA (0-50  $\mu\text{M}$ ). While measuring the absorption spectra, an equal amount of DNA was added to all the test solutions and reference solution to eliminate the absorbance of DNA itself.

## Results and Discussion

### Elemental analysis

The transparency and constituents of elements (CHN) of the synthesised complex were verified by the elemental analysis. The results shows that the compound BP contains C: 43.20 (44.97%), H: 2.12 (2.61%), N: 19.91 (20.17%), O 31.25 (32.25%). The data analysed show that the experimentally obtained values are in good agreement with theoretical values (within the bracket). The result confirms the formation of the compound in stoichiometric proportion and the compound is free of impurities.

### IR spectra

The characteristic vibrational frequencies of the functional group in the crystal lattice of BP are known through IR spectrum as shown in (Fig. 1) The formation of the charge transfer complex is strongly evidenced by the donor (picric acid) and acceptor (benzimidazole) molecules in the spectrum<sup>43,44</sup>. It is generally expected in acid – base interaction that there is a transfer of proton from the donor (acid) to acceptor (base). From the spectrum it is observed that the bands of the donor are slightly shifted to lower frequency and that of the acceptor are slightly shifted to higher frequency. This shift has been attributed to the charge transfer from the donor to acceptor upon the complexation. The band observed at  $3155 \text{ cm}^{-1}$  is due to the  $\text{N}^+\text{-H}$  stretching vibration.

The absorption peak at  $3663 \text{ cm}^{-1}$  is due to the N-H asymmetric stretching vibration and corresponding symmetric stretching vibration is observed at  $3093 \text{ cm}^{-1}$ . The aromatic C-H stretching vibration appears at  $3055 \text{ cm}^{-1}$ . The peak at  $933 \text{ cm}^{-1}$  is assigned to C- $\text{NO}_2$  stretching vibration. The frequencies at  $1080 \text{ cm}^{-1}$  is owed to C-H in plane bending vibration. The aromatic C=C stretching vibration is exhibited at  $1604 \text{ cm}^{-1}$ . The peak at  $1527 \text{ cm}^{-1}$  is due to  $\text{NO}_2$  asymmetric

stretching vibration and the corresponding symmetric stretching vibration band is observed at  $1327 \text{ cm}^{-1}$ . The band is at  $709 \text{ cm}^{-1}$  owes to the aromatic C-H out of plane bending vibration. The C-O stretching vibration appears at  $1256 \text{ cm}^{-1}$ . The  $\text{NO}_2$  scissoring and wagging vibrations are observed at  $840$  and  $748 \text{ cm}^{-1}$ .

### UV-visible absorption spectra

The optical absorption band is the most important parameter of the synthesised material for the implementation of various optical and biological applications. UV absorption spectra arise from the transition of electrons within a molecule from a lower electronic energy (LUMO) state to higher energy state (HOMO). When an atom or molecule absorbs the energy, electrons are promoted from their ground state to an excited state. The UV-visible absorption spectrum of the complex is shown in Fig. 2. The  $\pi\text{-}\pi^*$  and  $n\text{-}\pi^*$  transitions of BP appears 225 nm and 268 nm, respectively. The change in the band is due to the transfer of an electron from donor

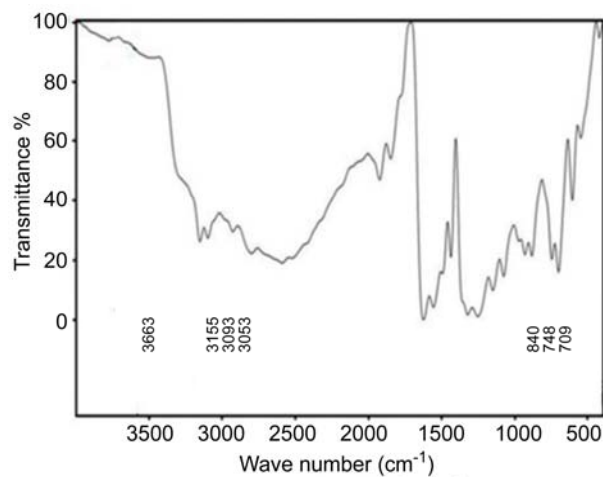


Fig. 1 — IR Spectrum of Benzimidazolium

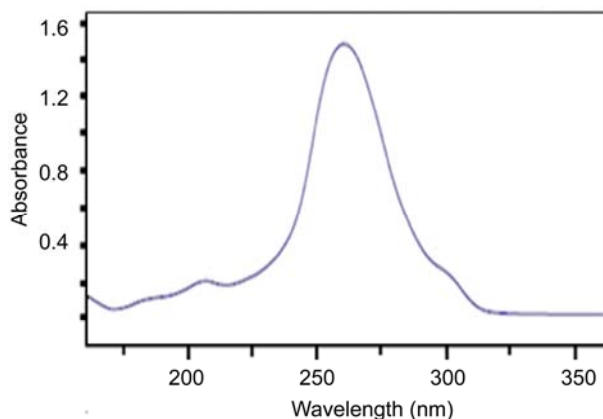


Fig. 2 — Electronic absorption spectrum of Benzimidazolium picrate

benzimidazole molecule to the acceptor picric acid molecule and appears on the longer wave length region. The charge transfer transition expected to occur in the longwave length region is gets submerged under the more intense  $n-\pi^*$  band.

#### NMR spectra

Molecular structure and carbon skeleton of BP crystals were confirmed by  $^1\text{H}$  and  $^{13}\text{C}$  NMR studies resulting from dissolution of crystalline BP in deuterated methanol are shown in Fig. 3 A and B, respectively. Six dissimilar proton signals appeared in the spectrum, confirming the presence of six different proton environments in the BP system. The downfield signal at 9.5 ppm corresponds to protons (H6 and H5A) of the picrate anion. The most intense singlet peak at  $\delta$  8.9 ppm is assigned to the ring proton (H11) of the benzimidazolium moiety. The absence of any signal above  $\delta$  11 ppm impute to the phenolic proton confirming the existence of the picrate anion in solution through proton transfer mechanism. In the spectrum, the signals at  $\delta$  8.1 and (two H atoms attached to  $\text{C}_{13}$  and  $\text{C}_8$ ) and  $\delta$  7.9 ppm (two H atoms attached to  $\text{C}_{12}$  and  $\text{C}_7$ ) authenticate the presence of two sets of chemically equivalent protons in the aromatic ring of benzimidazolium moiety.

A singlet signal at  $\delta$  5 ppm is attributed to the -NH protons of benzimidazolium moiety in the BP complex. The carbon signal appeared in the upfield at  $\delta$  139.5 is attributed to the  $\text{C}_2$  and  $\text{C}_{11}$  carbon atoms of BP. The weak carbon signal at  $\delta$  130 owes to the  $\text{C}_1$ ,  $\text{C}_3$  and  $\text{C}_4$  carbon atoms in which nitro group is present in the picrate moiety. The  $\text{C}_5$  and  $\text{C}_6$  carbon atoms of the same kind bring forth a signal at  $\delta$  126.4. The highly intense peak at  $\delta$  125.1 is due to  $\text{C}_7$ ,  $\text{C}_{12}/\text{C}_8$ ,  $\text{C}_{13}$  carbon atoms of the same environment in benzimidazolium moiety and the signal at  $\delta$  45 is assigned to solvent carbon atom of the compound.

#### Thermal analysis

The thermal stability and its decomposition pattern of BP has been established by consecutively employing TG/DTA analysis and the thermogram is shown in Fig. 4. From TG curve, it is contained that the title complex is stable up to  $146^\circ\text{C}$  and it decompose immediately after melting which is inferred that there is no inclusion of environmental moisture during crystallization. The decomposition occurs in a single stage. From the DTA curves, the sharp endothermic peak appearing at  $146^\circ\text{C}$

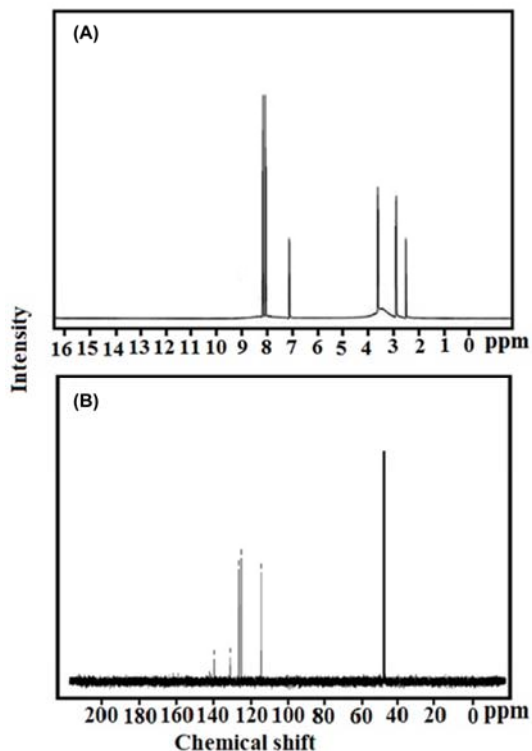


Fig. 3 — (A)  $^1\text{H}$  NMR spectrum and (B)  $^{13}\text{C}$  NMR of BP

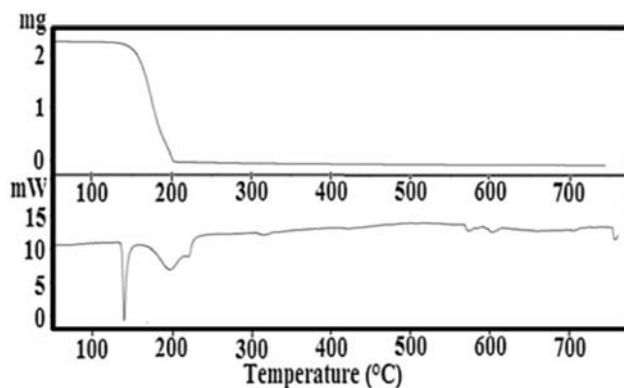


Fig. 4 — Thermal analysis graphs of BP

corresponds to the melting point of the material. This is followed by endothermic may be possible due to complex formation. No thermal anomaly was observed before the  $146^\circ\text{C}$ , this suggests that the BP was stable up to  $146^\circ\text{C}$ .

#### DNA binding studies

Electronic absorption spectroscopy is one of the most common and effective technique for investigation of the binding modes of organic compounds with CT-DNA. Compound binding through intercalation usually results in hypochromism with or without small red or blue shift, since the

intercalative mode involves a strong interaction between the aromatic chromophore and the base pairs of DNA<sup>45</sup>. Absorption spectra of the BP in the absence and presence of CT-DNA is given in (Fig. 5).

The binding mode of BP has been characterized through absorbance and shifts in the wavelength as a function of added concentration of DNA. Upon addition of increasing amounts of CT-DNA, a significant hypochromism is observed. This can be attributed to a strong interaction between DNA and compound and it is also likely that this compound binds to the DNA helix *via* intercalation. In order to illustrate quantitatively the consequence, the absorption data were analyzed to evaluate the intrinsic binding constant ( $K_b$ ), which can be determined from the following equation<sup>46-48</sup>,

$$[DNA]/(\varepsilon_a - \varepsilon_f) = [DNA]/(\varepsilon_b - \varepsilon_f) + 1/K_b(\varepsilon_b - \varepsilon_f)$$

where [DNA] is the concentration of DNA in base pairs, the apparent absorption coefficient  $\varepsilon_a$ ,  $\varepsilon_f$  and  $\varepsilon_b$  corresponds to  $A_{obs}/[\text{compound}]$ , the extinction coefficient of the free compound and the extinction coefficient of the compound when fully bound to DNA, respectively. From the plot of  $[DNA]/(\varepsilon_a - \varepsilon_f)$  versus [DNA],  $K_b$  is calculated by the ratio of slope to the intercept. The magnitude of intrinsic binding constant ( $K_b$ ) value for BP is  $4.9 \times 10^4 \text{ M}^{-1}$ . From the above DNA binding results, it is obvious that the title compound has planarity and extended  $\pi$  system which lead to the possibility of DNA intercalation.

#### Antioxidant activity

The DPPH radical has been used to test the ability of compound as free radical scavenger or hydrogen donor to evaluate the antioxidant activity. Hence experiments were carried out to explore the free radical scavenging ability of benzimidazolium picrate with DPPH radical<sup>49,50</sup> and compared with those of the positive control, ascorbic acid (Aca). The compound shows significant capacity for scavenging DPPH and the  $IC_{50}$  value (Fig. 6) indicated that it shows potent antioxidant activity. It also shows scavenging capacity beyond  $83 \mu\text{M/mL}$ . The activity of BP is also found to be good. (Table 1)

#### Single crystal X-ray diffraction method

A single crystal X-ray analysis is made at 296 K using suitable crystals for data collection. Accurate lattice parameters are resolved from least squares refinement of well centered reflection in the range  $2.58 \theta \text{ U } \theta 28.30$ . The complex, BP belongs to the monoclinic system with space group  $P2_1/c$ . The lattice

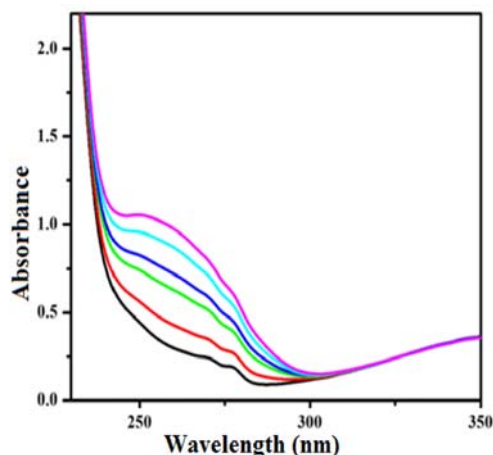


Fig. 5 — Electronic spectra of BP in Tris-HCl buffer upon addition of CT-DNA

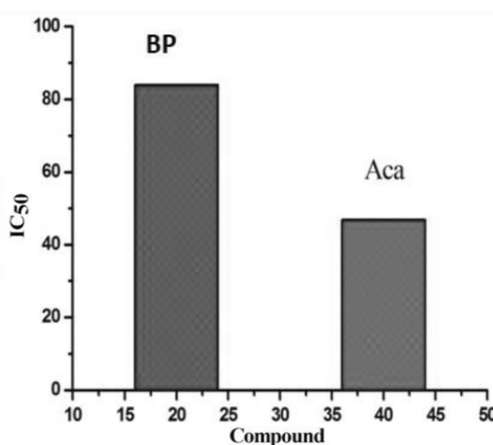


Fig. 6 — Antioxidant activity of BP

[Compound] =  $25 \mu\text{M}$ , [DNA] =  $0-50 \mu\text{M}$  DNA Solvent = (95% Water and 5%DMSO) (Inset: Plot between [DNA] and  $[DNA]/[\varepsilon_a - \varepsilon_f] \times 10^{-8}$ )

Table 1 — Comparison of antioxidant activity of Benzimidazolium picrate (BP) with other compounds

Compound	DPPH activity $IC_{50}\%$
2-Amino-4-methoxy-6-methylpyrimidine <sup>49</sup>	55
2,3-dimethylquinoxalinium-p-toluenesulfonate <sup>50</sup>	110
3-aminopyridium- p-toluenesulfonate <sup>51</sup>	125
Benzimidazolium picrate	83
Ascorbic acid	66.26

parameter obtained are  $a=9.2376(7) \text{ \AA}$ ,  $b=11.9355(7) \text{ \AA}$ ,  $c=12.8931(7) \text{ \AA}$ ,  $\alpha=90.00^\circ$ ,  $\beta=97.811(2)^\circ$ ,  $\gamma=90.00^\circ$  and the unit volume is  $1408.34(16) \text{ \AA}^3$ . The crystallographic data and structure refinements of BP complex are given in Table 2.

The benzimidazolium ion appears in crystalline lattice as single protonated cation and the picrate ion

Table 2 — Crystallographic data and structure refinement parameters of Benzimidazolium picrate (BP) complex

Empirical formula	C <sub>13</sub> H <sub>9</sub> N <sub>5</sub> O <sub>7</sub>
Formula weight	347.25
Temperature	296 (2) K
Wavelength	0.71073 Å
Crystal system	Monoclinic
Space group	P2 <sub>1</sub> /c
Cell dimensions	a = 9.2376(7) Å, α = 90.00° b = 11.9355(7) Å, β = 97.811(2)° c = 12.8931(7) Å, γ = 90.00°
Volume	1408.34(16) Å <sup>3</sup>
Z	4
Density (calculated)	1.638 mg/m <sup>-3</sup>
Absorption coefficient	0.137 mm <sup>-1</sup>
F <sub>(000)</sub>	712
Crystal size	0.40 × 0.35 × 0.30 mm <sup>-1</sup>
Reflections collected	11222
Range for data collection (deg)	2.58–28.30 °
Number parameters	227
Absorption correction	Semi-empirical from equivalents
Max and min. transmission	0.9602 and 0.9474
Limiting indices h, k, l	-12/8, -15/15, -17/15
Refinement method	Full-matrix least squares on F <sup>2</sup>
Goodness of F <sup>2</sup>	1.017

is present as deprotonated anion. X-ray single crystal structure of the charge-transfer complex of benzimidazole with picric acid shows the presence of mono protonated benzimidazolium cation and gives the intermolecular hydrogen bonding association. The ORTEP diagram of BP is depicted the Fig. 7.

The selected bond lengths and bond angle is given in Table 3, respectively. In the picrate anion, C-O bond distance of anion is evident for characteristic values with C2–O6 [1.2413(18) Å] which is intermediate between single and double bond characters. The loss of a proton from picric acid is confirmed by the lengthening of the C-C bonds. The C1–C2 and C1–C3 bond lengths are (1.447(2) Å) and (1.450(2) Å), respectively which deviate from the standard aromatic C-C bond length (1.40 Å). These differences are attributed to the loss of a hydroxyl proton at O6, leading to conversion from neutral to the anionic state where the negative charge is constrained to lie in the ring. The crystal packing diagram of BP is shown in Fig. 8.

The picrate anion plays a vital role in forming hydrogen bonds with cation and stabilizing the structure. The degree of twisting of the nitro group from the benzene plane does not depend upon the C-N bond distance. The C-O bond length of the phenolate [1.248(2)°] corresponds to a partial double bond

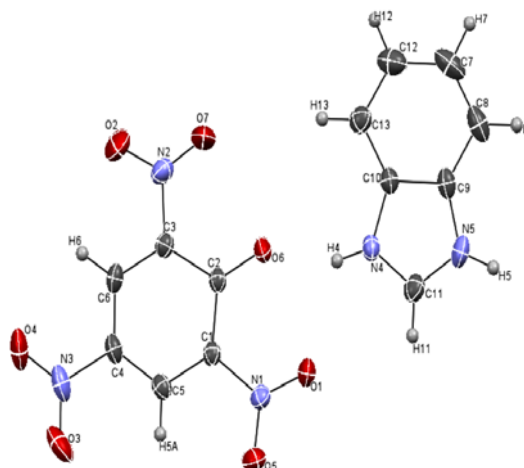


Fig. 7 — ORTEP diagram of BP

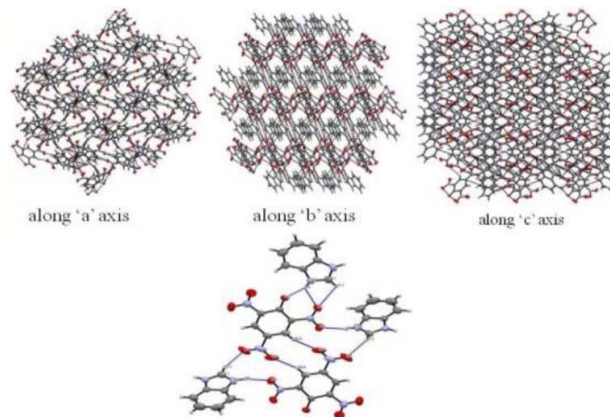


Fig. 8 — Crystal packing diagram of BP

character implying that the negative charge located on the phenolate oxygen atom is delocalized. The N-O bond distance of the nitro groups of the trinitrophenolate anion are typical double bonds.

The twist angles of the three nitro group from the benzene plane of the picrate ion are (O1–N1–O5, 121.10(14) Å), (O7–N2–O2, 121.68(17) Å) and (O3–N3–O4, 123.24(17) Å). Analysis of the picrate ion show that the ortho nitro groups in general, deviate away from the benzene plane due to steric interactions with the phenol group at C5 but para nitro group lies in the benzene plane. Deviation from the normal behaviour arises in many structures as a result of crystal packing criteria which involve N-H...O and C-H...O hydrogen bonds with the nitro group O atoms.

The formation of intermolecular hydrogen bond N-H...O and C-H...O in crystal structure lead to supramolecular networking. The hydrogen bonded networks of the BP indicates that the cation part

Table 3 — Selected bond length (Å) and bond angle (°) of BP complex

Atom	observed	Atom	Observed	Atom	observed
O(1)-N(1)	1.2188(18)	C(10)-C(13)	1.381(2)	C(5)-C(4)-N(3)	119.36(16)
O(2)-N(2)	1.214(2)	C(11)-H(11)	0.93	C(6)-C(4)-N(3)	119.01(16)
O(3)-N(3)	1.217(2)	C(12)-C(13)	1.363(3)	C(4)-C(5)-C(1)	118.94(15)
O(4)-N(3)	1.229(2)	C(12)-H(12)	0.93	C(4)-C(5)-H(5A)	120.5
O(5)-N(1)	1.2345(18)	C(13)-H(13)	0.93	C(1)-C(5)-H(5A)	120.5
O(6)-C(2)	1.2413(18)	O(1)-N(1)-O(5)	121.10(14)	C(3)-C(6)-C(4)	119.52(15)
O(7)-N(2)	1.187(2)	O(1)-N(1)-C(1)	120.80(14)	C(3)-C(6)-H(6)	120.2
N(1)-C(1)	1.444(2)	O(5)-N(1)-C(1)	118.09(14)	C(4)-C(6)-H(6)	120.2
N(2)-C(3)	1.456(2)	O(7)-N(2)-O(2)	121.68(17)	C(8)-C(7)-C(12)	122.5(2)
N(3)-C(4)	1.441(2)	O(7)-N(2)-C(3)	120.28(16)	C(8)-C(7)-H(7)	118.7
N(4)-C(11)	1.312(2)	O(2)-N(2)-C(3)	118.03(16)	C(12)-C(7)-H(7)	118.7
N(4)-C(10)	1.381(2)	O(3)-N(3)-O(4)	123.24(17)	C(7)-C(8)-C(9)	116.29(19)
N(4)-H(4)	0.86	O(3)-N(3)-C(4)	118.99(18)	C(7)-C(8)-H(8)	121.9
N(5)-C(11)	1.320(2)	O(4)-N(3)-C(4)	117.76(19)	C(9)-C(8)-H(8)	121.9
N(5)-C(9)	1.381(2)	C(11)-N(4)-C(10)	108.82(14)	N(5)-C(9)-C(8)	132.91(17)
N(5)-H(5)	0.86	C(11)-N(4)-H(4)	125.6	N(5)-C(9)-C(10)	105.98(15)
C(1)-C(5)	1.373(2)	C(10)-N(4)-H(4)	125.6	C(8)-C(9)-C(10)	121.11(19)
C(1)-C(2)	1.447(2)	C(11)-N(5)-C(9)	108.82(14)	N(4)-C(10)-C(13)	131.23(15)
C(2)-C(3)	1.450(2)	C(11)-N(5)-H(5)	125.6	N(4)-C(10)-C(9)	106.38(15)
C(3)-C(6)	1.356(2)	C(9)-N(5)-H(5)	125.6	C(13)-C(10)-C(9)	122.39(17)
C(4)-C(5)	1.369(2)	C(5)-C(1)-N(1)	115.95(14)	N(4)-C(11)-N(5)	110.01(16)
C(4)-C(6)	1.381(2)	C(5)-C(1)-C(2)	124.01(14)	N(4)-C(11)-H(11)	125
C(5)-H(5A)	0.93	N(1)-C(1)-C(2)	120.04(13)	N(5)-C(11)-H(11)	125
C(6)-H(6)	0.93	O(6)-C(2)-C(1)	124.88(14)	C(13)-C(12)-C(7)	121.7(2)
C(7)-C(8)	1.358(3)	O(6)-C(2)-C(3)	123.06(14)	C(13)-C(12)-H(12)	119.1
C(7)-C(12)	1.393(4)	C(1)-C(2)-C(3)	112.02(13)	C(7)-C(12)-H(12)	119.1
C(7)-H(7)	0.93	C(6)-C(3)-C(2)	123.90(15)	C(12)-C(13)-C(10)	115.93(18)
C(8)-C(9)	1.383(3)	C(6)-C(3)-N(2)	116.19(14)	C(12)-C(13)-H(13)	122
C(8)-H(8)	0.93	C(2)-C(3)-N(2)	119.89(14)	C(10)-C(13)-H(13)	122
C(9)-C(10)	1.386(2)	C(5)-C(4)-C(6)	121.60(14)		

joined to the anion part via extended hydrogen bonding. In crystal structure the cation and anion linking by strong N-H...O and C-H...O hydrogen bonding. The combination of all type of intermolecular hydrogen forms three dimensional networks. This hydrogen bonding link layer of cation with the layer anion form straight chain along in the b axis.

### Conclusions

A novel organic charge transfer crystal of Benzimidazolium picrate was synthesized and successfully grown by the slow evaporation solution growth technique at ambient temperature. The single crystal analysis indicated that the crystal belongs to monoclinic with P21/c space group. The CT complex is found to be biologically significant with regards to its DNA binding. The radical scavenging activity showed that the CT complex possesses the greater activity against DPPH radical. FTIR was substantial the presence of functional group, especially  $\text{NH}^+$  stretching vibration at  $3155\text{ cm}^{-1}$ . The UV spectrum confirmed the formation of CT complex. TG and

DTA analysis inferred the thermal stability of the synthesized complex. The proton and carbon positions were analyzed by NMR spectra which establish molecular structure of the crystal.

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### Conflict of interest

The authors declare no conflict of interests in this study.

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