JETIR.ORG
ISSN: 2349-5162 | ESTD Year : 2014 | Monthly Issue
JOURNAL OF EMERGING TECHNOLOGIES AND INNOVATIVE RESEARCH (JETIR)
An International Scholarly Open Access, Peer-reviewed, Refereed Journal

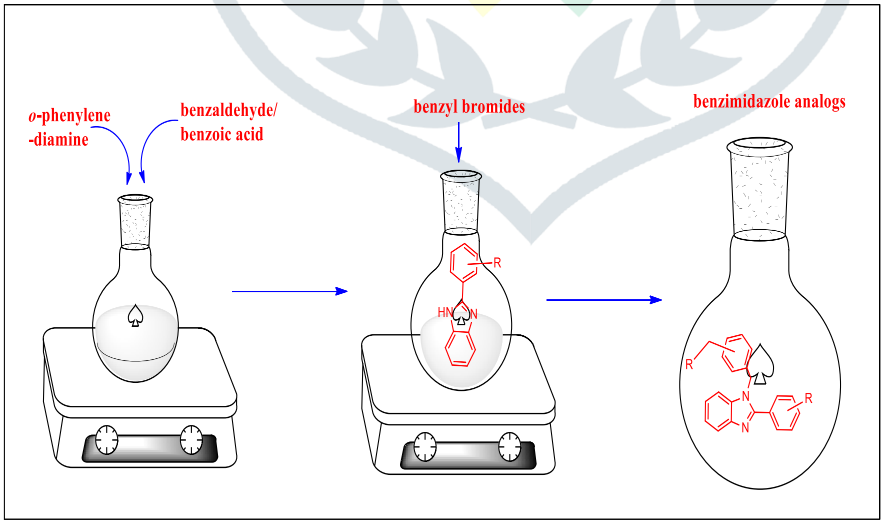
Synthesis, characterization and Biological Evaluation of Benzimidazole Analogs

Reetu Sangwan[1], Naz Perween Alam[2], Sudha Jain[3]
[1]Assistant Professor, [2]Assistant Professor & Head, [3]Former Professor and Head
[1]Department of Chemistry, R B College, Dalsing Sarai, Samastipur, L.N.M. University, Darbhanga, Bihar, India
[2] Department of Chemistry, M K College, Laheriasarai, L.N.M. University, Darbhanga, Bihar, India
[3] Department of Chemistry, University of Lucknow,Lucknow
Email: ritnikrana@gmail.com

Abstract: Benzimidazole and its analogs belong to important class of bioactive heterocycles in the field of medicinal chemistry. The 5,6-dimethyl-1-(a-D-ribofuranosyl)benzimidazole ring system was discovered in 1948 as an integral part of the structure of vitamin B12. By retrosynthetic analysis we synthesized total sixteen novel benzimidazole analogs along with the synthesis of two known benzimidazole analogs **4** and **5** precursors and two series (**7a-7h** and **8a-8h**) of eight analogs each and screened all the synthesized analogs for antioxidant (free radical scavenging activity) and antibacterial activity out of them only **8a** with phenyl substituent and **8g** with 3,5-dinitrophenyl substituent showed the highest free radical scavenging activity with % inhibition 98.73 % and 98.91 % as compared to standard BHT (99.24 %) at 100 µg/ml concentration. In the evaluation of antibacteraial activity it was found that all the novel analogs (**7a-7h** and **8a-8h**) were moderately active against all five bacterial strains *viz. Agrobacterium tumefaciens* (gram -ve), *Erwinia chrysanthemi* (gram -ve), *Xanthomonas phaseoli* (gram -ve), *Escherichia coli* (gram -ve) and *Bacillus subtilis* (gram +ve) with standard antimicrobial agent streptomycin.

IndexTerms: Antimicrobial, Antioxidant, Benzimidazole, DPPH.

Graphical Abstract



1. Introduction

Benzimidazole is one of the important nitrogen containing heterocyclic species because if its synthetic importance and various pharmacological, medicinal and agricultural application **[1]**. Benzimidazole drugs, due to their structural features and electron rich environment have ability to bind with different types of therapeutic targets. Benzimidazole is one of the oldest known nitrogen heterocycles, first synthesized by Hoebrecker and later by Ladenberg and Wundt during 1872–1878 **[2, 3]**. The benzimidazole derived alkaloids have limited occurrence in nature and only a few examples of natural products are known which have benzimidazole basic scaffolds **[4]**.

Benzimidazole and its analogs belong to important class of bioactive heterocycles in the field of medicinal chemistry[5]. They exhibit various biological activities including antimicrobial **[6]**, anti-inflammatory **[7]**, antitumour **[8]**, antiparasitic **[7]**, antiprotozoal **[9]**, anti-HIV **[10]**, anti-herpes (HSV-1) **[11]**, antimicobacterial [12], anti-influenza **[13]**, human cytomegalovirus (HCMV) **[14]**, anticonvulsant **[15]**, antineoplastic **[16]**, antiulcer **[17]**, anthelmintic **[18]**, antiproliferative **[19]**, analgesic **[20]**, cardiovascular applications **[21]**, antioxidant activity **[22]**, anti-ischemia–reperfusion injury **[23]**, anti-hypertension **[24]** and obesity **[25]**. They also act as ligands for modeling biological systems of transition metals **[26]**, asymmetric catalysis **[27]** and antimicrobial [**28-33]**. There is special scientific interest also to develop diverse types of antioxidants for medical treatment because antioxidants are very important for protecting living organisms against excessive reductive oxygen species (ROS) [**34-37].**

Literature survey revealed that none of the research group have attempted the substitution reaction at N-1 position of 4-(1H-benzodimidazol-2-yl)-2-methoxyphenol (4) and 2-(3,5-dinitrophenyl)-1H-benzodimidazole (5). It was therefore thought worthwhile to carry out the synthesis of C-2 substituted benzimidazoles (4) and (5). Further the substitution reactions will be done at N-1 position of these benzimidazole analogs (4 and 5) with different substituted benzyl bromides along with evaluation of *in vitro* antioxidant activity (free radical scavenging activity using DPPH) taking BHT as standard of all the synthesized compounds (7a-7h and 8a-8h). All the synthesized compounds (7a-7h and 8a-8h) will be also screened for antibacterial activity following cup-plate method using Muller-Hinton Agar for bacteria against standard drug streptomycin for bacteria.

2. Result and discussion

To the best of our knowledge, none of the presently synthesized benzimidazole analogs **(7a-7h** and **8a-8h)** have been reported in literature. We have synthesized eighteen **(7a-7h** and **8a-8h)** new analogs so as to evaluate their antioxidant and antimicrobial activity.

2.1. Synthesis of benzimidazole Analogs

The total syntheses of novel benzimidazole analogs **7a-7h** and **8a-8h** are shown in Scheme 1 and 2. The % yield, M.P. (^{0}C) and time required of various benzimidazole analogs are recorded in **Table 1.**

The structures of all benzimidazole analogs (**7a-7h** and **8a-8h)** and all the precursors **(4** and **5)** were established by their spectral data (IR, ^{1}H NMR, ^{13}C NMR and HRMS) except benzyl bromides, which were used in subsequent reactions. The detailed descriptions are as follows:

2.2. Synthesis of 4-(1H-benzo[d]midazol-2-yl)-2-methoxyphenol (4)

The *o*-phenylene diamine **(1)** was refluxed in water in the presence of catalytic amount of conc. HCl with vanillin (**2**). The reaction mixture was refluxed for 3 hrs. Usual workup afforded the respective substituted benzimidazole analogs **(4)** in excellent yield **(80%). (Scheme 1, Table 1)**

2.3. Synthesis of 2-(3, 5-dinitrophenyl)-1H-benzo[d]imidazole (5)

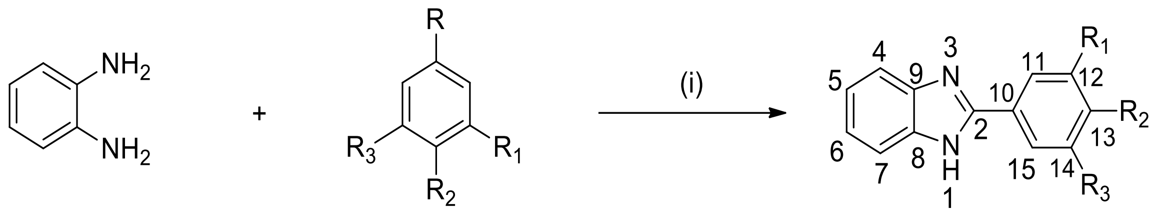
The *o*-phenylene diamine **(1)** was refluxed in water in the presence of catalytic amount of conc. HCl with 3,5dinitrobenzoic acid **(3).** The reaction mixture was refluxed for 4h. Usual workup afforded the respective substituted benzimidazole analogs **(5) (85%)**. **(Scheme 1, Table 1)**

2.4. Synthesis of substituted 1H-benzo[d]imidazol-2-yl (7a-7h)

The synthesized benzimidazole analog, **4** was stirred with substituted benzyl bromides **(6a-6h)** at room temperature in 10% ethanolic KOH solution. The reaction mixture was stirred for 3-5 hrs. Usual workup of the reaction mixture afforded the corresponding 1-N substituted benzimidazole analogs **(7a-7h)** in excellent yield (~85%). **(Scheme 2, Table 1)**

2.5. Synthesis of substituted 2-(3,5-dinitrophenyl)-1H-benzo[d]imidazole (8a-8h)

Similarly, another synthesized benzimidazole analog, **5** was stirred with benzyl bromide **(6a-6h)** at room temperature in 10% ethanolic KOH solution. The reaction mixture was stirred for 5-8 hrs. Usual workup afforded the corresponding 1-N substituted benzimidazole analogs **(8a-8h)** in excellent yield (~81%). **(Scheme 3, Table 1).**

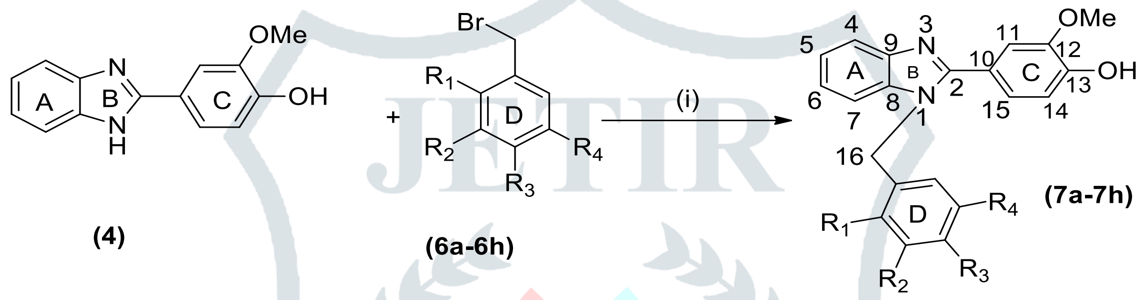


(1)

(2)
R=CHO, R_1= OMe, R_2=OH, R_3=H
(3)
R=COOH, R_1= NO_2, R_2=H, R_3=NO_2

(4)
R_1= OMe, R_2=OH, R_3=H
(5)
R_1= NO_2, R_2=H, R_3=NO_2

Scheme 1: (i) H_2O, Conc. HCl, reflux



(4)

(6a-6h)

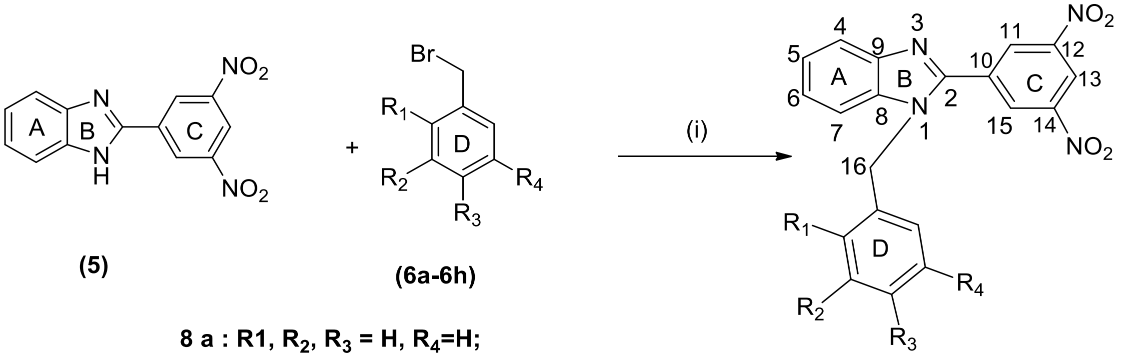
(7a-7h)

6 a, 7 a : R1, R_2, R_3 = H, R_4=H;
6 b, 7 b : R1, R2 = H, R_3 = -OBn, R_4=H;
6 c, 7 c : R1, R_2 = H, R_3 = -OBz, R_4=H;
6 d, 7 d : R1, R_2, R_3 = -OMe, R_4=H;
6 e, 7 e : R1 = H, R2 = -OMe, R_3 = -OBz, R_4=H;
6 f, 7 f : R1, R_2 = H, R_3 = -OMe, R_4=H;
6 g, 7 g : R1 = H, R_2 = OMe, R_3= OH, R_4=Br, R_4=H;
6 h, 7 h : R1=-OMe, R_2, R_3 = H, R_4=H;

Scheme 2 : (i) C_2H_5OH, KOH, r.t., 3-6 hrs

Table 1: Benzimidazole analogs (4, 5, 7a-h, 8a-h)

S. No.	Compounds	M.P. (^{0}C)	Yield (%)	Time (h)
1.	4	160-162	80	3
2.	5	177–179[38]	85	4
3.	7a	183-184	85	4
4.	7b	196-198	82	3
5.	7c	230-212	80	5
6.	7d	240-241	89	3
7.	7e	256-157	87	4
8.	7f	212-214	85	4
9.	7g	264-267	84	6
10.	7h	220-223	87	4
11.	8a	203-204	81	5
12.	8b	236-138	88	6
13.	8c	242-244	83	5
14.	8d	255-258	82	7
15.	8e	270-271	87	5
16.	8f	232-234	86	4
17.	8g	285-286	83	8
18.	8h	251-255	89	4



(5) **(6a-6h)** **(8a-8h)**

8 a : R1, R_2, R_3 = H, R_4=H;
8 b : R1, R2 = H, R_3 = -OBn, R_4=H;
8 c : R1, R_2 = H, R_3 = -OBz, R_4=H;
8 d : R1, R_2, R_3 = -OMe, R_4=H;
8 e : R1 = H, R2 = -OMe, R_3 = -OBz, R_4=H;
8 f : R1, R_2 = H, R_3 = -OMe, R_4=H;
8 g : R1 = H, R_2 = OMe, R_3= OH, R_4=Br;
8 h : R1=-OMe, R_2, R_3 = H, R_4=H;

Scheme 3 : (i) C_2H_5OH, KOH, r.t., 3-7 hrs

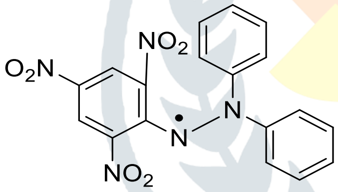
2.6. Biological Activity

All the synthesized analogs (**7a-7h** and **8a-8h)** were evaluated for *in vitro* free radical scavenging activity and anti microbial activity.

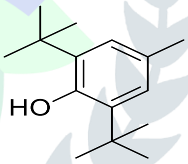
2.6.1. Experiment 1: Determination of free radical scavenging activity by 1, 1-diphenyl-2-picrylhydrazyl (DPPH) method (Table 2, Graph 1 and 2).

All the synthesized 4-(1H-benzo[d]midazol-2-yl)-2-methoxyphenol analogs **(7a-7h)** and 2-(3,5-dinitrophenyl)-1H-benzo[d]imidazole analogs **(8a-8h)** were separately dissolved in methanol to prepare samples of different concentrations (1000 µg/ml, 500 µg/ml, 250 µg/ml, 100 µg/ml) each as described in material and methods (**Experiment 1**).

The antioxidant activity in terms of percentage of DPPH scavenging activities were determined using BHT (butylated hydroxyl toluene) as standard, as described in materials and methods. The results are listed in Table 2 and Graph 1 and 2.



(DPPH)



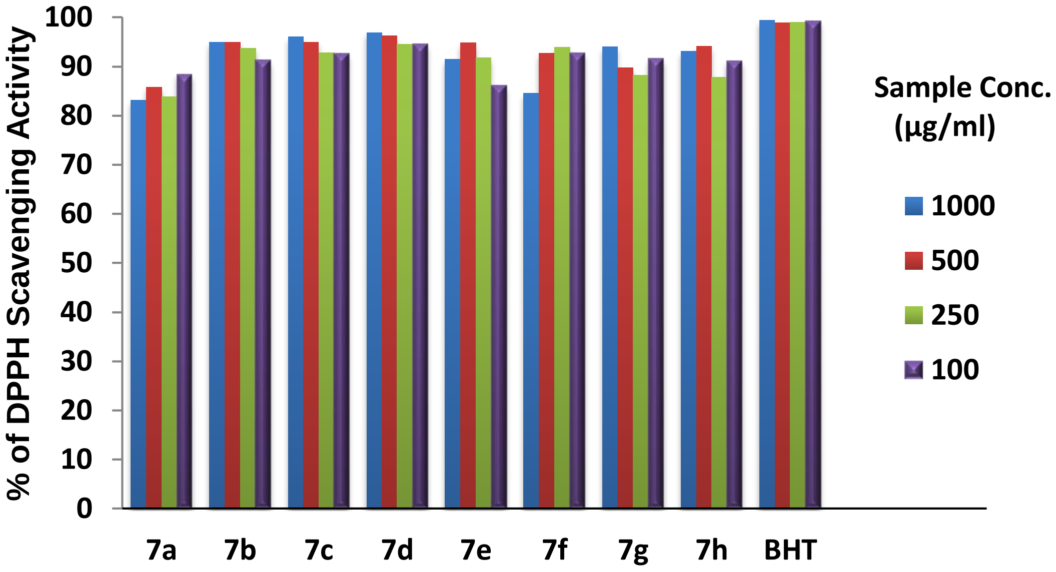
Butylated hydroxyl toluene (BHT)

The absorbance at 517nm for free radical scavenging activity by DPPH was compared with the standard BHT at 517 nm at four concentrations viz. 1000 µg/ml, 500 µg/ml, 250 µg/ml and 100 µg/ml. The standard BHT has shown an increase in absorbance as the concentration decreases. The absorbance obtained for BHT at the concentrations 1000 µg/ml, 500 µg/ml, 250 µg/ml and 100 µg/ml were 0.02, 0.0269, 0.026 and 0.0202 respectively (Table-2). Interestingly out of 16 samples (**7a-7h** and **8a-8h**) at concentration 1000 µg/ml only one compound (**7d**) has shown approximately comparable value of absorption (0.0844) with the standard BHT (0.02). At concentration 500 µg/ml, two samples (**8a** and **8g**) have shown absorption values 0.073 and 0.082 respectively comparable with the standard BHT (0.0269). Similarly at concentration 250 µg/ml, three samples **8a, 8c** and **8g** have shown absorption values 0.056, 0.042 and 0.057 respectively comparable with the standard BHT (0.026). Similarly at concentration 100 µg/ml, three samples **8d, 8f** and **8g** have shown absorption values 0.059, 0.061 and 0.033 respectively comparable with the standard BHT (0.020). All the results of absorption values at different concentrations are listed in Table 2.

The study of percentage of DPPH scavenging activity in 16 samples (**7a-h** and **8a-h**) has also been carried out and demonstrated that the three samples **8a, 8c** and **8g** have the highest value of % inhibition at concentrations 250 µg/ml (98.03, 98.56 and 98.01 respectively) and 100 µg/ml (98.73, 98.88 and 98.11 respectively) comparable with the standard BHT at concentrations 250 µg/ml (99.02) and 100 µg/ml (99.24) (**Table-2, Graph-2**).

Other analogs which have remarkable % of DPPH scavenging activity are **7b** with % inhibition 94.98 % and 94.99 % (1000 µg/ml and 500µg/ml respectively), **7b** with % inhibition 95.99 % (1000 µg/ml), **7d** with % inhibition 96.83% and 96.23% (1000 µg/ml and 500µg/ml respectively), **7g** with % inhibition 94.10% (1000 µg/ml), **7h** with % inhibition 94.17 % (500 µg/ml), **8d** with % inhibition 97.93% and 96.64% (250 µg/ml and 100µg/ml respectively) and **8f** with % inhibition 96.35% and 97.83% (250 µg/ml and 100µg/ml respectively) (**Table-2, Graph-1 and 2**).

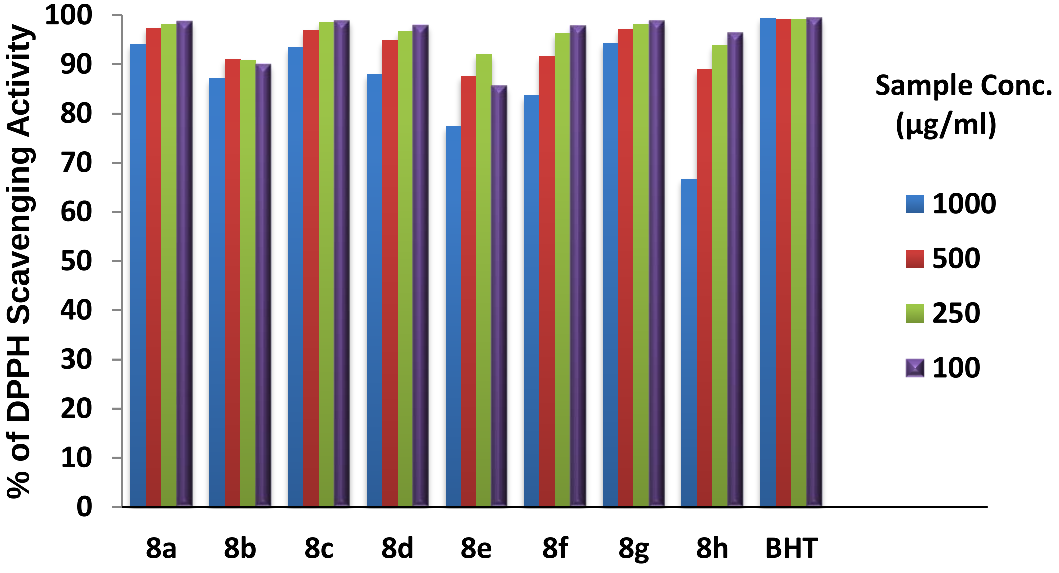
The results therefore demonstrated that the free radical scavenging activity and % inhibition are related to the concentration of the compound and the most suitable concentration of the synthesized compounds is 100µg/ml-250 µg/ml comparable to standard BHT.



Graph 1: % inhibition of DPPH scavenging activity of 7(a-h)

Table 2: Absorbance (517 nm) and % Inhibition at different concentrations for free radical scavenging activity by DPPH in compounds 7a-7h and 8a-8h and BHT

Sample	Concentration (µg/ml)							
	1000		500		250		100	
	Absorbance (517 nm)	% Inhibition	Absorbance (517 nm)	% Inhibition	Absorbance (517 nm)	% Inhibition	Absorbance (517 nm)	% Inhibition
7a	0.4509	83.1092	0.377	85.8775	0.4311	83.850	0.3102	88.37985
7b	0.134	**94.9803**	0.1337	**94.9915**	0.168	93.7066	0.2328	91.27927
7c	0.107	**95.9917**	0.1349	94.9466	0.1944	92.77	0.1966	92.63532
7d	**0.0844**	**96.8383**	0.1006	**96.231**	0.1453	94.5570	0.1466	94.50833
7e	0.2267	91.5077	0.1392	94.7855	0.2189	91.7999	0.3723	86.05357
7f	0.4121	84.5626	0.1967	92.6315	0.1612	93.9614	0.1945	92.71399
7g	0.1573	**94.1075**	0.2753	89.6872	0.3141	88.2337	0.2244	91.59393
7h	0.1831	93.1410	0.1554	**94.86**	0.3262	87.7804	0.2377	91.09571
8a	0.1631	94.0381	**0.073**	**97.4186**	**0.0567**	**98.0302**	0.0378	**98.73935**
8b	0.347	87.1384	0.2405	91.1342	0.2457	90.9391	0.27	90.02739
8c	0.84	93.4641	0.0859	96.9346	**0.0425**	**98.5630**	0.0338	**98.88943**
8d	0.3249	87.9675	0.143	94.7923	0.0935	**96.6495**	**0.0593**	**97.93269**
8e	0.6062	77.4134	0.3327	87.6749	0.2144	92.1134	0.3882	85.59262
8f	0.44	83.6491	0.2264	91.6632	0.1014	**96.3531**	**0.0618**	**97.83889**
8g	0.1539	94.3833	**0.082**	**97.081**	**0.057**	**98.0189**	**0.0331**	**98.91569**
8h	0.8922	66.6829	0.2982	88.9693	0.1692	93.8093	0.0995	96.42442
BHT	**0.02**	**99.355**	**0.0269**	**98.9923**	**0.026**	**99.0260**	**0.0202**	**99.2433**



Graph 2: % inhibition of DPPH scavenging activity of 8(a-h)

2.6.2. Experiment 2: Determination of antibacterial activity of benzimidazole analogs (7a-7h and 8a-8h) (Table 3, Graph 3 and 4)

All the benzimidazole analogues **(7a-7h)** and **(8a-8h)** were evaluated for their antibacterial activity and the results are summarized in **Table 3.**

Antibacterial Activity- The antibacterial activity was evaluated against five pathogenic bacterial strains *viz. Agrobacterium tumefaciens* (gram -ve), *Erwinia chrysanthemi* (gram -ve), *Xanthomonas phaseoli* (gram -ve), *Escherichia coli* (gram -ve) and *Bacillus subtilis* (gram +ve) with standard antimicrobial agent streptomycin.

The antibacterial activity was carried out against the above mentioned organisms by following the cup-plate method using Muller-Hinton Agar medium for bacteria. The compounds were screened for their antibacterial activity at 200 µg/ml concentration level using DMSO as solvent.

The zone of inhibitions of the synthesized compounds against five pathogenic (Gram positive and Gram negative) bacteria are presented in **Table 3**.

Among all the tested compounds **7c, 7e, 7g, 8c, 8e** and **8g** have shown mild antibacterial activity as compared to standard drug streptomycin (**Table 3**). After the evaluation of antibacterial activity the zone of inhibition for the most active compound **(7e)** amongst **7a-7h** was 20.67 for *A. tumefaciens*, 22.76 for *E. chrysanthemi,*15.54 for *X. phaseoli*, 18.35 for *E. coli and* 19.12 for *B. subtilis* strains. Other compounds showing mild activity were **7g** having zone of inhibition 19.66, 15.76, 14.55, 18.88 *and* 18.85 and **7c** having zone of inhibition 18.33, 14.56, 14.87, 17.54 and 18.07 respectively for *A. tumefaciens, E. chrysanthemi, X. phaseoli, E. coli* and *B. subtilis* strains.

Similarly amongst the series **8a-8h** compound **8e** was found to be most active antibacterial agent as compared to standard drug streptomycin. The zone of inhibition for the compound **8e** was **28.87** for *A. tumefaciens*, **27.86** for *E. chrysanthemi*, **19.96** for *X. phaseoli*, **22.87** for *E. coli and* **25.74** for *B. subtilis* strains. Other compounds showing mild activity were **8g** having zone of inhibition 27.66, 26.97, 19.88, 22.83 and 25.57 and **8c** having zone of inhibition 23.54, 25.87, 18.87, 21.05 and 24.08 for *A. tumefaciens, E. chrysanthemi, X. phaseoli, E. coli* and *B. subtilis* strains respectively.

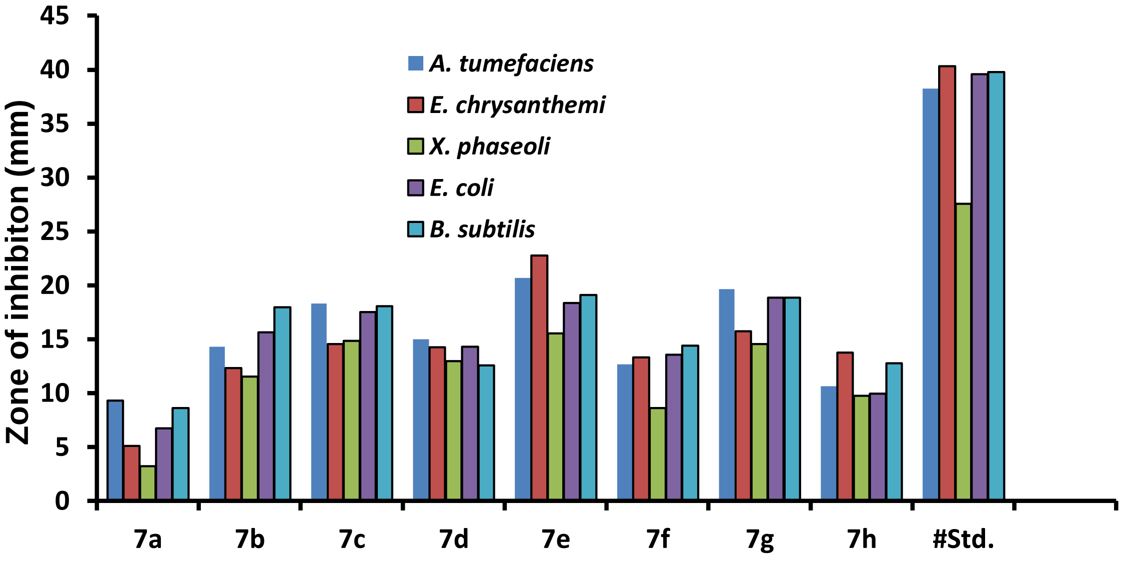
As predicted by the SAR in general it is clear that value of zone of inhibitions were greater for compounds **8a-8h** due to presence of nitro group (electron withdrawing group) than the compounds **7a-7h** bearing methoxy and phenolic group (electron donating group) in basic scaffold of substituted benzimidazoles. The compounds **7e** and **8e** both bearing -OBz group (electron withdrawing group) at 4th position and methoxy group at 3rd position of benzene ring showed higher antibacterial activity. The bromo group at 4th position of phenyl ring also enhances the activity in both compounds **7c** and **8c** thus supporting the SAR prediction that electron withdrawing groups enhance the activity.

The graphical representations of zone of inhibition of all the tested compounds are displayed in Graph 3 and 4.

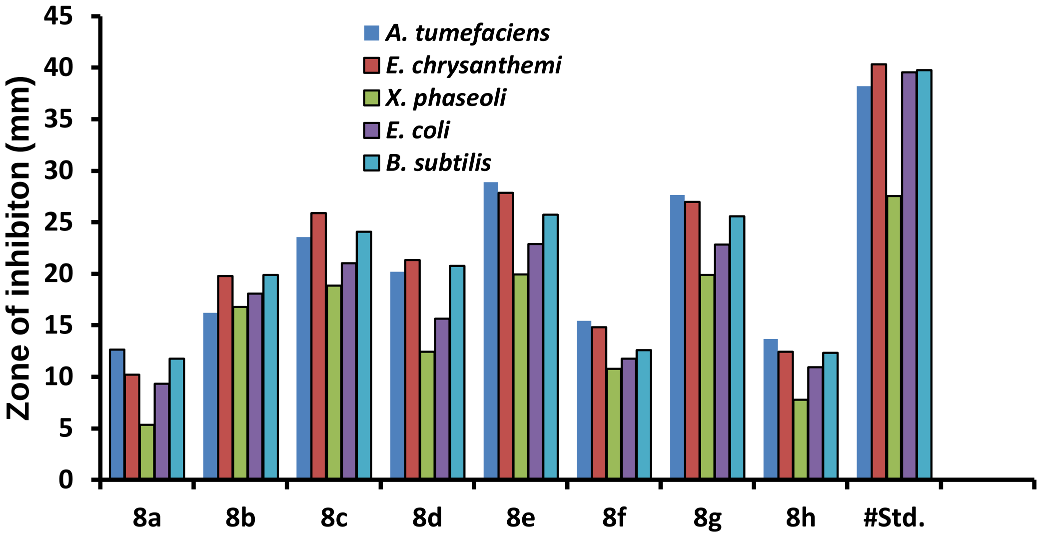
Table 3: Antibacterial activity (Zone of inhibition)* of benzimidazole derivatives (**7a-7h and 8a-8h**)

S.No.	**Samples**	**Gram negative bacteria**				**Gram Positive bacteria**
		A. tumefaciens	***E. chrysanthemi***	***X. phaseoli***	***E. coli***	***B. subtilis***
1.	**7a**	9.33	5.12	3.22	6.73	8.65
2.	**7b**	14.33	12.34	11.54	15.65	17.97
3.	**7c**	**18.33**	14.56	14.87	**17.54**	**18.07**
4.	**7d**	15.00	14.24	12.97	14.32	12.60
5.	**7e**	**20.67**	**22.76**	15.54	**18.35**	**19.12**
6.	**7f**	12.66	13.32	8.64	13.57	14.43
7.	**7g**	**19.66**	**15.76**	14.55	**18.88**	**18.85**
8.	**7h**	10.66	13.77	9.78	9.98	12.78
9.	**8a**	12.66	10.23	5.34	9.32	11.76
10.	**8b**	16.21	19.78	16.76	18.09	19.90
11.	**8c**	**23.54**	**25.87**	**18.87**	**21.05**	**24.08**
12.	**8d**	20.22	21.34	12.46	15.65	20.75
13.	**8e**	**28.87**	**27.86**	**19.96**	**22.87**	**25.74**
14.	**8f**	15.44	14.84	10.76	11.74	12.60
15.	**8g**	**27.66**	**26.97**	**19.88**	**22.83**	**25.57**
16.	**8h**	13.67	12.43	7.76	10.94	12.35
17.	**#Std.**	**38.23**	**40.33**	**27.57**	**39.57**	**39.76**

#Std. means Streptomycin (+control). All the compounds were screened at 200 µg/ml concentration



Graph 3: Zone of inhibition for compounds 7a-7h



Graph 4: Zone of inhibition for compounds 8a-8h

3. Experimental

Melting point was determined in a melting point apparatus. The IR spectra were recorded on a Perkin-Elmer RX-1 (4000-450 cm^{-1}) spectrophotometer using KBr pellets. ^1H NMR and ^{13}C NMR (Nuclear Magnetic Resonance) spectra were recorded on Bruker 300 MHz instrument using DMSO as a solvent. Chemical shifts are reported in parts per million (ppm) using Tetramethylsilane (TMS) as internal standard. Solvents and reagents were used without further purification, unless otherwise specified. The Positive Electron Spray Ionization (ESI) High resolution mass spectrometry of compound was recorded on Agilent 6520(Q-TOF) mass spectrometer and mass spectrum of the isolated compounds. Ultra violet (UV) spectrum was recorded on UV–visible Double-Beam Spectrophotometer (systronic-2203) instrument using chloroform as a solvent. The thin layer chromatography (TLCs) were visualized in an iodine chamber and carried out on precoated silica gel plates 60 F254 or RP-18 F254 plates (Merck). TLC was carried out on precoated silica gel plates 60 F254 (Merck) for monitoring of reactions. Spots were visualized by UV light or by iodine chamber. Column Chromatography was performed using silica gel (230-400 and 100-200 mesh size). 1, 1-Dipheny1-2-picryl-hydrazyl (DPPH) was purchased from Sigma-Aldrich Merck to determine the antioxidant activity of synthesized compounds in terms of free radical scavenging activities. *o*-Phenylenediamine (**157**), vanillin (**158**), 3, 5-dinitrobenzoic acid (**159**) and benzyl bromide (**162 a**) were purchased from Sigma Aldrich Chemical Co.

3.1. Synthesis of substituted benzimidazole 4 and 5

3.1.1. Synthesis of 4-(1H-benzo[d]imidazol-2-yl)-2-methoxyphenol (4)

A mixture of *o*-phenylenediamine (1.08 gm, 10 mmol) (**1**) and vanillin (1.52 gm, 10 mmol) (**2**) in water (15 ml) in presence of catalytic amount of conc. HCl was stirred for 3 hrs in an oil bath at 100^0C. The progress of reaction was monitored by TLC. After completion of reaction the reaction mixture was cooled to room temperature, solvent decanted and the residue extracted with $CHCl_3$ (3 x 20 ml). The organic layer was dried over anhydrous Na_2SO_4 and filtered. The residue was purified using silica gel column chromatography using $CHCl_3$: MeOH (98:2) as an eluent to obtain the corresponding benzimidazole analog **4** as yellowish brown solid.**Yield:** 85%; IR (KBr; υ_{max}, cm^{-1}) 3450.45 (-NH Stretching), 3196.05 (-CH Stretching), 1597.06 (C=N Stretching), 1508.33, 1436.97 (C=C Stretching), 1272.06 (C-N Stretching), 1222.42 (C-N Stretching), 1029.99 (C-O Stretching). ^1H (DMSO, 300 MHz; δ, ppm): 7.08-7.88 (4H, m, Ar-H, H-4, H-5, H-6, H-7), 7.55 (1H, dd, J = 8.8, Ar-H, H-14), 6.96 (1H, dd, J = 8.8, Ar-H, H-15), 7.46 (1H,s, Ar-H, H-11), 5.70 (1H, s, N$\underline{H}$), 5.35 (1H, s, O$\underline{H}$), 3.68 (3H, s, OC$\underline{H_3}$); ^{13}C (DMSO, 75 MHz; δ, ppm): 152.98 (C-2), 148.79 (C-13), 148.09 (C-12), 141.76 (C-8, C-9), 124.23 (C-10), 124.56 (C-15), 123.03 (C-5, C-6), 115.34 (C-14), 115.23 (C-4,C-7), 112.21 (C-11), 56.23 (O$\underline{C}H_3$). ESI-MS *m/z* 241 $M+H^+$; Mol. Formula: $C_{14}H_{12}N_2O_2$.

3.1.2. Synthesis of 2-(3,5-dinitrophenyl)-1H-benzo[d]imidazole (5)

The compound **5** was prepared similarly as described for the synthesis of **4** by the reaction of **1** with 3, 5-dinirobenzoic acid **(3)** as starting material for 4 hrs. The residue was purified using silica gel column chromatography, $CHCl_3$: MeOH (95:5) as an eluent to obtain the corresponding benzimidazole analog **5** as dark brown solid.**Yield:** 80%; IR (KBr; υ_{max}, cm^{-1}) 3400.23 (-NH Stretching), 3066.82 (-CH Stretching), 1620.34 (C=C Stretching), 1556.65 (C=N Stretching), 1544.23 (NO_2 sym Stretching), 1129.58 (C-O Stretching), 927 (N-O). ^1H (DMSO, 300 MHz; δ, ppm): 8.97 (2H, s, Ar-H, H-11, H-15), 8.36 (1H, s, Ar-H, H-13), 7.22-7.59 (4H, m, Ar-H, H-4, H-5, H-6, H-7), 5.05 (1H, s, NH); ^{13}C (DMSO, 75 MHz; δ, ppm): 150.27 (C-2), 147.37 (C-12, C-14), 140.47 (C-8, C-9), 132.47 (C-10), 126.87 (C-11, C-15), 122.56 (C-5, C-6), 122.34 (C-13), 121.09 (C-4, C-7). ESI-MS *m/z* 285 $M+H^+$; Mol. Formula: $C_{13}H_8N_4O_4$.

3.1.3. Synthesis of 4-(1H-benzo[d]imidazol-2-yl)-2-methoxyphenol (4) analogs (7a-7h)

3.1.3.1. 4-(1-benzyl-1H-benzo[d]imidazol-2-yl)-2-methoxyphenol (7a)

In a round bottom flask, 4-(1H-benzodimidazol-2-yl)-2-methoxyphenol **(4)** (0.240 mg, 1mmol) in ethanol (10 ml) was stirred with anhydrous KOH (500 mg) for 5 minutes at room temperature. Then benzyl bromide **(6a) (0.5 ml)** was added drop wise with the help of pressure equalizing dropping funnel and stirred for 4 to 6 hrss. The progress of the reaction was monitored with TLC. After completion of reaction the reaction mixture was poured in to ice cold water and then filtered. The pure compound **7a** was obtained as a yellow brown solid. **Yield:** 85%; IR (KBr; υ_{max}, cm^{-1}) 3146.06 (-CH Stretching), 1625.67 (C=C Stretching) 1610.50 (C=N Stretching), 1436.97 (C=C Stretching), 1382.53 (C-N Stretching), 1272.23 (C-N Stretching), 1167.59 (C-O Stretching). ^{1}H (DMSO, 300 MHz; δ, ppm): 6.90-7.59(12H, m, Ar-$\underline{H}$, H-5, H-6, H-7, H-4, H-11, H-14, H-15, H-17 to H-22), 5.79 (1H, s, NC$\underline{H_2}$, H-16), 5.45 (1H, s, O$\underline{H}$), 3.83 (3H, s, OC$\underline{H_3}$, H-23); ^{13}C (DMSO, 75 MHz; δ, ppm): 153.96 (C-2), 148.07 (C-13), 148.02 (C-12) , 142.62 (C-8, C-9), 137.30 (C-17), 128.64 (C-18, C-22), 127.89 (C-19, C-21), 125.70 (C-20), 124.23 (C-10), 123.09 (C-5, C-6, C-15), 119.05 (C-4, C-7), 115.84 (C-14), 112.71 (C-11), 56.13 (O$\underline{C}$H$_3$), 52.22 (C-16, N-$\underline{C}$H$_2$). ESI-MS *m/z* 331 M+H$^+$; Mol. Formula: $C_{21}H_{18}N_2O_2$. Anal. Calcd.: C 76.34, H 5.49, N 8.48, O 9.69.

3.1.3.2. 4-(1-(4-(benzyloxy)benzyl)-1H-benzo[d]imidazol-2-yl)-2-methoxyphenol (7b)

The compound **7b** was prepared in a similar manner as described earlier for the synthesis of **(7a)** by the reaction of **(4)** with **(6b)** (0.278 mg, 1 mmol). The pure compound **7b** was obtained as a brown solid. TLC Solvent System: 9:1 ($CHCl_3$: MeOH); **Yield:** 82%; IR (KBr; υ_{max}, cm^{-1}) 3156.56 (-CH Stretching), 1615.17 (C=C Stretching) 1560.12 (C=N Stretching), 1283.56 (C-N Stretching), 1262.27 (C-N Stretching), 1207.51 (C-O Stretching). ^{1}H (DMSO, 300 MHz; δ, ppm): 6.43-7.23 (16H, m, Ar-$\underline{H}$, H-5, H-6, H-7, H-4, H-11, H-14, H-15, H-25 to H-29, H-18, H-19, H-21, H-22), 5.69 (1H, s, NC$\underline{H_2}$, C-16), 5.35 (1H, s, O$\underline{H}$), 5.16(2H, s, OC$\underline{H_2}$Ph, H-23), 3.84 (3H, s, OC$\underline{H_3}$, H-30); ^{13}C (DMSO, 75 MHz; δ, ppm): 156.08 (C-20), 152.98 (C-2), 148.79(C-13), 148.09 (C-12), 141.76 (C-8, C-9), 136.56 (C-24), 130.5 (C-17), 129.6 (C-19, C-21), 128.50 (C-27), 127.34 (C-25, C-19), 127.10 (C-26, C-28), 124.23 (C-10), 124.56 (C-15), 115.34 (C-14), 114.2, 112.21 (C-11), 70.80 ($\underline{C}$H$_2$Ph, C-23), 56.12 (O$\underline{C}$H$_3$, C-30), 50.21(C-16, N-$\underline{C}$H$_2$).ESI-MS *m/z* 437 M+H$^+$; Mol. Formula: $C_{28}H_{24}N_2O_3$. Anal. Calcd.: C 77.04, H 5.54, N 6.42, O 11.0.

3.1.3.3. 4-((2-(4-hydroxy-3-methoxyphenyl)-1H-benzo[d]imidazol-1-yl)methyl) phenyl benzoate (7c)

The compound **7c** was prepared in a similar manner as described earlier for the synthesis of **(7a)** by the reaction of **(4)** with **(6 c)** (0.289 mg, 1 mmol). The pure compound **7c** was obtained as a brown solid. TLC Solvent System: 9:1 ($CHCl_3$: MeOH); **Yield:** 80%; IR (KBr; υ_{max}, cm^{-1}) 3309.89 (O-H Stretching), 3101.03 (-CH Stretching), 1756.34 (C=O Stretching), 1596.25 (C=C Stretching) 1515.23 (C=N Stretching), 1283.56 (C-N Stretching), 1267.45(C-N), 1262.27 (C-N Stretching), 1220.32 (C-N), 1120.11 (C-O Stretching); ^{1}H (DMSO, 300 MHz; δ, ppm): 8.22 (2H, dd, J=8.4, Ar-H, ring D), 7.60 (2H, dd, J=8.4, H-19, H-21) 7.25-7.50 (12H, m, Ar-H, H-5, H-6, H-7, H-4, H-11, H-14, H-15, H-25 to H-29), 5.79 (2H, s, NCH$_2$, H-16), 5.35 (1H, s, O$\underline{H}$), 3.84 (3H, s, OC$\underline{H_3}$, H-30); ^{13}C (DMSO, 75 MHz; δ, ppm): 165.23 (C=O), 153.30 (C-2), 146.40 (C-13), 141.55 (C-12), 146.45 (C-13), 146.40 (C-20), 142.40 (C-9), 138.95 (C-8), 137.28 (C-17), 133.99 (C-24), 131.42 (C-18, C-22), 130.34 (C-19, C-21), 129.40 (C-27), 127.50 (C-28, C-26), 128.34 (C-25, C-29), 123.67 (C-10), 123.05 (C-5, C-6), 121.32 (C-15), 116.76 (C-14), 119.56 (C-4, C-7), 110.20 (C-11), 56.23 (O$\underline{C}$H$_3$, C-30), 52.87(C-16, N-$\underline{C}$H$_2$); ESI-MS *m/z* 451 M+H$^+$; Mol. Formula: $C_{28}H_{22}N_2O_4$. Anal. Calcd.: C 74.65, H 4.92, N 6.22, O 14.21.

3.1.3.4. 2-methoxy-4-(1-(2,3,4-trimethoxybenzyl)-1H-benzo[d]imidazol-2-yl) phenol (7d)

The compound **7d** was prepared in a similar manner as described earlier for the synthesis of **(7a)** by the reaction of **(4)** with **(6 d)** (0.262 mg, 1 mmol). The pure compound **7d** was obtained as a brown solid. TLC Solvent System: 9:1 ($CHCl_3$: MeOH); **Yield:** 89%; IR (KBr; υ_{max}, cm^{-1}) 3359.67 (O-H Stretching), 3152.42 (-CH Stretching), 1550.27 (C=C Stretching), 1525.28 (C=N Stretching), 1213.34 (C-N Stretching), 1267.45(C-N), 1120.11 (C-O Stretching), 1105.80 (C-O Stretching); ^{1}H (DMSO, 300 MHz; δ, ppm): 6.90-7.59 (6H, m, Ar-H, H-5, H-6, H-7, H-4, H-14, H-15), 7.19 (1H, s, Ar-H, H-11), 6.57 (1H, d, J=7.5, Ar-H, H-21), 6.32 (1H, d, J=7.5, H-22), 5.46 (2H, s, NCH2, H-16), 5.35 (1H, s, O$\underline{H}$), 3.84 (12H, s, 4 x OC$\underline{H_3}$); ^{13}C (DMSO, 75 MHz; δ, ppm):153.3 (C-2), 151.4 ($\underline{C}$OCH$_3$, C-18), 150.98($\underline{C}$OCH$_3$, C-20), 148.76 (C-13), 148.56 (C-12), 142.43 ($\underline{C}$OCH$_3$, C-20), 124.42 (C-10), 143.42 (C-9), 138.40 (C-8), 148.09 (C-12), 123.06 (C-15), 119.07 (C-4, C-7), 115.67 (C-14), 112.43 (C-11), 104.63 (C-22), 123.67 (C-17), 119.60 (C-21), 60.80 (O$\underline{C}$H$_3$, C-23, C-24, C-25, C-26), 56.12 (C-16, N-$\underline{C}$H$_2$); ESI-MS *m/z* 421 M+H$^+$; Mol. Formula: $C_{24}H_{24}N_2O_5$. Anal. Calcd.: C 68.56, H 5.75, N 6.66, O 19.03.

3.1.3.5. 4-((2-(4-hydroxy-3-methoxyphenyl)-1H-benzo[d]imidazol-1-yl)methyl)-2-methoxyphenyl benzoate (7e)

The compound **7e** was prepared in a similar manner as described earlier for the synthesis of **(7a)** by the reaction of **(4)** with **6e** (0.321 mg, 1 mmol). The pure compound **7e** was obtained as a brown solid. TLC Solvent System: 9:1 ($CHCl_3$: MeOH); **Yield:** 87%; IR (KBr; υ_{max}, cm^{-1}) 3289.34 (O-H Stretching), 3092.78 (-CH Stretching), 1760.60 (C=O Stretching), 1576.85 (C=C Stretching), 1515.35 (C=N Stretching), 1223.90 (C-N Stretching), 1217.92(C-N Stretching), 1262.27 (C-O Stretching), 1220.32 (C-O Stretching), 1120.11 (C-O Stretching); ^{1}H (DMSO, 300 MHz; δ, ppm): 6.90-8.22 (15H, m, Ar-H, H-5, H-6, H-7, H-4, H-14, H-15, H-25 to H-19), 5.59 (2H, s, NCH$_2$, H-16), 5.38 (1H, s, O$\underline{H}$), 3.78 (6H, s, 2 x OC$\underline{H_3}$, H-30, H-31); ^{13}C (DMSO, 75 MHz; δ, ppm): 165.23 (C-23), 153.56 (C-2), 151.57 ($\underline{C}$-OCH$_3$, C-20), 148.79 (C-13), 148.07 (C-12), 142.43 (C-9), 142.45 (C-8), 137.81 (C-10), 135.40 (C-19), 134.90 (C-17), 130.30 (C-27), 130.09 (C-25, C-29), 133.30 (C-24), 128.66, 124.24 (C-18), 121.78 (C-21), 123.65 (C-5, C-6), 122.50 (C-15), 119.90 (C-14), 112.80 (C-11), 109.34 (C-22), 55.89(O$\underline{C}$H$_3$, C-30, C-31), 52.56(C-16, N-$\underline{C}$H$_2$); ESI-MS *m/z* 481 M+H$^+$; Mol. Formula: $C_{29}H_{24}N_2O_5$. Anal. Calcd.: C 72.49, H 5.03, N 5.03, O 16.65.

3.1.3.6. 2-methoxy-4-(1-(4-methoxybenzyl)-1H-benzo[d]imidazol-2-yl)phenol (7f)

The compound **7f** was prepared in a similar manner as described earlier for the synthesis of **(7a)** by the reaction of **(4)** with **6f** (0.201 mg, 1 mmol). The pure compound **7f** was obtained as a brown solid. TLC Solvent System: 9:1 ($CHCl_3$:MeOH); **Yield:** 85%; IR (KBr; υ_{max}, cm^{-1}) 3172.82 (-CH Stretching), 1566.86 (C=C Stretching), 1505.23 (C=N Stretching), 1203.93 (C-N Stretching), 1187.82(C-N Stretching), 1162.27 (C-O Stretching), 1120.32 (C-O Stretching), 1130.78 (C-O Stretching); ^1H (DMSO, 300 MHz; δ, ppm): 7.22-7.59 (4H, m, Ar-H, H-4, H-5, H-6, H-7), 7.19 (1H, s, Ar-H, H-11), 7.59 (2H, dd, J= 7.8, Ar-H, ring D), 7.02 (2H, dd, J= 8.6, Ar-H, ring D), 7.12 (2H, d, J= 7.8, Ar-H, H-15), 6.87 (2H, dd, J= 8.6, Ar-H, H-16), 5.79 (2H, s, NCH2, H-16), 5.39 (1H, s, $O\underline{H}$), 3.83 (6H, s, $OC\underline{H}_3$, H-23, H-24); ^{13}C (DMSO, 75 MHz; δ, ppm): 157.56 (C-20), 153.36 (C-2), 148.70 (C-13), 148.07 (C-12), 142.43 (C-9), 137.81 (C-8), 130.09 (C-17), 129.61 (C-18, C-22), 124.24 (C-10), 123.65 (C-15), 119.90 (C-4, (C-7), 115.80 (C-14), 114.26 (C-19, C-26), 112.70 (C-11), 58.89 ($O\underline{C}H_3$, C-23, C-24), 56.31(C-16, N-$\underline{C}H_2$); ESI-MS *m/z* 361 $M+H^+$; Mol. Formula: $C_{22}H_{20}N_2O_3$. Anal. Calcd.: C 73.79, H 5.59, N 7.77, O 13.32.

3.1.3.7. 2-bromo-4-((2-(4-hydroxy-3-methoxyphenyl)-1H-benzo[d]imidazol-1-yl)methyl)-6-methoxyphenol (7g)

The compound **7g** was prepared in a similar manner as described earlier for the synthesis of **(7a)** by the reaction of **(4)** with **6g** (0.295 mg, 1 mmol). The pure compound **7g** was obtained as a brown solid. TLC Solvent System: 9:1 ($CHCl_3$:MeOH); **Yield:** 84%; IR (KBr; υ_{max}, cm^{-1}) 3001.24(-CH Stretching), 1598.99 (C=C Stretching), 1550.77 (C=N Stretching), 1282.66 (C-N Stretching), 1263.37(C-N Stretching), 1141.86 (C-O Stretching), 1099.43 (C-O Stretching), 1043.49 (C-O Stretching); ^1H (DMSO, 300 MHz; δ, ppm): 6.75-7.45 (9H, m, Ar-H, H-5, H-6, H-7, H-4, H-14, H-15, ring D), 5.64 (2H, s, NCH2, H-16), 4.92 (1H, s, $O\underline{H}$), 3.81 (3H, s, $OC\underline{H}_3$, H-24), 3.79 (3H, s, $OC\underline{H}_3$, H-23); ^{13}C (DMSO, 75 MHz; δ, ppm): 157.56 ($\underline{C}$-OCH_3, C-21), 153.36 (C-2), 148.70 (C-13), 148.07 (C-12), 142.43 (C-9), 137.81 (C-8), 130.09 ($\underline{C}$-OH), 129.61 (C-17), 127.59 (C-18), 127.29 (C-22), 124.24 (C-10), 123.65 (C-5, C-6), 119.90 (C-4, C-7), 115.80 (C-14), 114.26 (C-11), 112.70 (C-Br), 58.89($O\underline{C}H_3$, C-23, C-24), 56.31(C-16, N-$\underline{C}H_2$); ESI-MS *m/z* 456 $M+H^+$; Mol. Formula: $C_{22}H_{19}BrN_2O_4$. Anal. Calcd.: C 58.04, H 4.21, Br 17.55, N 6.15, O 14.06.

3.1.3.8. 2-methoxy-4-(1-(2-methoxybenzyl)-1H-benzo[d]imidazol-2-yl)phenol (7h)

The compound **7h** was prepared in a similar manner as described earlier for the synthesis of **(7a)** by the reaction of **(4)** with **6h** (0.201 mg, 1 mmol). The pure compound 7 **h** was obtained as a brown solid. TLC Solvent System: 9:1 ($CHCl_3$:MeOH); **Yield:** 87%; IR (KBr; υ_{max}, cm^{-1}) 2999.31 (-CH Stretching), 1599.62 (C=C Stretching), 1500.62 (C=N Stretching), 1282.66 (C-N Stretching), 1274.95 (C-N Stretching), 1246.02 (C-O Stretching), 1122.57 (C-O Stretching), 1029.99 (C-O Stretching); ^1H (DMSO, 300 MHz; δ, ppm): 6.36-7.88 (11H, m, Ar-H, Ar-H, H-5, H-6, H-7, H-4, H-14, H-15, ring D), 5.44 (2H, s, NCH2, H-16), 5.32 (1H, s, $O\underline{H}$), 3.82 (6H, s, 2 x $OC\underline{H}_3$); ^{13}C (DMSO, 75 MHz; δ, ppm):155.36 ($\underline{C}$-OCH_3, C-18), 148.66 (C-2),146.30 (C-13), 132.75 (C-12), 130.78 (C-8), 130.16 (C-9), 128.33 (C-17), 127.65 (C-19, C-21), 122.49 (C-10), 119.31 (C-4, C-7), 118.97 (C-15, C-6, C-6), 116.01 (C-14), 111.35 (C-20), 111.13 (C-22), 110.93 (C-11), 55.91($O\underline{C}H_3$, C-23), 55.53 ($O\underline{C}H_3$, C-24), 47.79 (C-16, N-$\underline{C}H_2$); HRMS *m/z* 378 $M+NH_4^+$; Mol. Formula: $C_{22}H_{20}N_2O_3$. Anal. Calcd.: C 73.32, H 5.59, N 7.77, O 13.32.

3.1.4. Synthesis of 2-(3,5-dinitrophenyl)-1H-benzo[d]imidazole (5) analogs (8a-8h)

3.1.4.1. 1-benzyl-2-(3,5-dinitrophenyl)-1H-benzo[d]imidazole (8a)

The compound **8a** was prepared in a similar manner as described earlier for the synthesis of **(7a)** by the reaction of **(5)** with **(6 a)** (0.5 ml). The pure compound **8a** was obtained as a brown solid. TLC Solvent System: 8:2 ($CHCl_3$:MeOH); **Yield:** 81%; IR (KBr; υ_{max}, cm^{-1}) 3109.29 (C-H Stretching), 1610.04 (C=C Stretching), 1565.34 (C=N Stretching), 1534.53 (NO_2 sym Stretching), 1257.47 (C-N Stretching); ^1H (DMSO, 300 MHz; δ, ppm): 9.04 (2H, s, Ar-H, H-11, H-15), 8.72 (1H, s, Ar-H, H-13), 7.22-7.69 (9H, m, Ar-H, H-4, H-5, H-6, H-7, ring D), 5.49 (2H, s, NCH_2, H-16). ^{13}C (DMSO, 75 MHz; δ, ppm): 155.32 (C-2), 150.27 (C-12, C-14), 140.45 (C-9), 137.78 (C-8), 137.32, 132.40 (C-10), 128.66 (C-18, C-22), 127.87 (C-19, C-21), 125.79 (C-20), 123.04 (C-5, C-6), 119.56 (C-4, C-7), 129.08 (C-11, C-15), 118.07 (C-13), 52.23 (C-16). ESI-MS *m/z* 375 $M+H^+$; Mol. Formula: $C_{20}H_{14}N_4O_4$. Anal. Calcd.: C 64.17, H 3.77, N 14.97, O 17.10.

3.1.4.2. 1-(4-(benzyloxy)benzyl)-2-(3,5-dinitrophenyl)-1H-benzo[d]imidazole (8b)

The compound **8b** was prepared in a similar manner as described earlier for the synthesis of **(7a)** by the reaction of **(5)** with **(6b)** (0.278 mg, 1 mmol). The pure compound **8b** was obtained as a brown solid. TLC Solvent System: 8:2 ($CHCl_3$:MeOH); **Yield:** 88%; IR (KBr; υ_{max}, cm^{-1}) 3110.20 (C-H Stretching), 1612.23 (C=C Stretching), 1545.12 (C=N Stretching), 1530.03 (NO_2 sym Stretching), 1260.34 (C-N Stretching),1220.40(C-O Stretching); ^1H (DMSO, 300 MHz; δ, ppm): 9.04 (2H, s, Ar-H, H-11, H-15), 8.72 (1H, s, Ar-H, H-13), 6.87-7.59 (13H, m, Ar-H, H-4, H-5, H-6, H-7, ring D, OBn), 5.59 (2H, s, $NC\underline{H}_2$, H-16), 5.29 (2H, s, $C\underline{H}_2$, $OC\underline{H}_2Ph$); ^{13}C (DMSO, 75 MHz; δ, ppm):156.07 (C-20), 153.23 (C-2), 149.34 (C-12, C-14), 142.42 (C-9), 137.89 (C-8), 136.57 (C-24), 132.43 (C-10), 127.65 (C-27), 128.99 (C-25, C-29), 127.19 (C-26, C-28), 130.09 (C-17), 128.97 (C-18, C-22), 126.64 (C-15, C-11), 123.09 (C-5, C-6), 119.53 (C-4, C-7), 118.06 (C-13), 114.24 (C-19, C-21), 70.12 ($O\underline{C}H_2Ph$, C-23), 52.43 (C-16); ESI-MS *m/z* 481 $M+H^+$; Mol. Formula: $C_{27}H_{20}N_4O_5$. Anal. Calcd.: C 67.49, H 4.20, N 11.66, O 16.65.

3.1.4.3. 4-((2-(3,5-dinitrophenyl)-1H-benzo[d]imidazol-1-yl)methyl)phenyl benzoate (8c)

The compound **8c** was prepared in a similar manner as described earlier for the synthesis of **(7a)** by the reaction of **(5)** with **(6 c)** (0.278 mg, 1 mmol). The pure compound **8c** was obtained as a brown solid. TLC Solvent System: 8:2 ($CHCl_3$:MeOH); **Yield:** 83%; IR (KBr; υ_{max}, cm^{-1}) 3108.24 (C-H Stretching), 1735.76 (C=O), 1605.29 (C=C), 1592.23 (C=C Stretching), 1555.52 (C=N Stretching), 1532.83 (NO_2 sym Stretching), 1243.67 (C-N Stretching), 1210.40(C-O Stretching); ^1H (DMSO, 300 MHz; δ, ppm): 9.04 (2H, s, Ar-H, H-11, H-15), 8.72 (1H, s, Ar-H, H-13), 7.24-8.22 (9H, m, Ar-H, ring D, OBz), 7.25-7.30(6H, s, Ar-H, H-4, H-5, H-6, H-7), 5.59 (2H, s, $NC\underline{H}_2$, H-16); ^{13}C (DMSO, 75 MHz; δ, ppm):165.21(C-23), 156.07 (C-20), 153.23 (C-2), 149.34 (C-12, C-14), 142.42 (C-9), 137.89 (C-8), 136.57 (C-17), 133.95 (C-24), 132.43 (C-10), 131.57 (C-27), 121.57(C-19, C-21), 130.09 (C-18, C-22), 128.97 (C-25, C-29), 128.05 (C-26, C-28), 123.34 (C-5, C-6), 121.57 (C-19, C-21), 121.09 (C-15, C-11), 119.53 (C-4, C-7), 118.06 (C-13), 52.43 (C-16); ESI-MS *m/z* 495 $M+H^+$; Mol. Formula: $C_{27}H_{18}N_4O_6$. Anal. Calcd.: C 65.59, H 3.67, N 11.33, O 19.41.

3.1.4.4. 2-(3,5-dinitrophenyl)-1-(2,3,4-trimethoxybenzyl)-1H-benzo[d]imidazole (8d)

The compound **8d** was prepared in a similar manner as described earlier for the synthesis of **(7a)** by the reaction of **(5)** with **(6 d)** (0.289 mg, 1 mmol). The pure compound **8d** was obtained as a brown solid. TLC Solvent System: 8:2 ($CHCl_3$:MeOH); **Yield:** 82%; IR (KBr; υ_{max}, cm^{-1}): 1600.26 (C=C), 1574.54 (C=C Stretching), 1530.50 (C=N Stretching), 1542.52 (NO_2 sym Stretching), 1212.90 (C-N Stretching), 1210.40(C-O Stretching), 1150.75 (C-O); ^1H (DMSO, 300 MHz; δ, ppm): 9.04 (2H, s, Ar-H, H-11, H-15), 8.72 (1H, s, Ar-H, H-13), 7.22-7.56(4H, m, Ar-H, H-4, H-5, H-6, H-7), 6.57(1H, d, J=7.6, Ar-H, ring D), 6.32(1H, d, J=7.6, Ar-H, ring D), 5.46 (2H, s, NCH_2, H-16), 3.83(9H, s, 3 x O-$C\underline{H_3}$,H-23, H-24, H-25); ^{13}C (DMSO, 75 MHz; δ, ppm):153.78 (C-2), 151.27 ($\underline{C}$-OCH_3, C-18), 150.09 ($\underline{C}$-OCH_3,C-20), 149.35 (C-12, C-14), 142.48 ($\underline{C}$-OCH_3), 142.32 (C-9), 137.86 (C-8), 132.45 (C-10), 128.98 (C-11, C-15), 123.34 (C-5, C-6), 123.03 (C-17), 119.58 (C-4, C-7), 119.05 (C-22), 118.03 (C-13), 104.67 (C-21), 60.80 (2 x $O\underline{C}H_3$, C-23, C-24), 56.30 ($O\underline{C}H_3$, C-25), 46.76 (C-16); ESI-MS *m/z* 465 $M+H^+$; Mol. Formula: $C_{23}H_{20}N_4O_7$. Anal. Calcd.: C 59.48, H 4.34, N 12.06, O 24.11.

3.1.4.5. 4-((2-(3,5-dinitrophenyl)-1H-benzo[d]imidazol-1-yl)methyl)-2-methoxyphenyl benzoate (8e)

The compound **8e** was prepared in a similar manner as described earlier for the synthesis of **(7a)** by the reaction of **(5)** with **6e** (0.321 mg, 1 mmol). The pure compound **8e** was obtained as a brown solid. TLC Solvent System: 8:2 ($CHCl_3$:MeOH); **Yield:** 87%; IR (KBr; υ_{max}, cm^{-1}): 3118.24 (C-H Stretching), 1745.56 (C=O), 1615.19 (C=C), 1592.23 (C=C Stretching), 1545.22 (C=N Stretching), 1542.83 (NO_2 sym Stretching), 1258.46 (C-N Stretching),1120. 04 (C-O Stretching); ^1H (DMSO, 300 MHz; δ, ppm): 9.04 (2H, s, Ar-H, H-11, H-15), 8.72 (1H, s, Ar-H, H-13), 7.22-8.23 (12H, m, Ar-H, ring D, OBz, H-4, H-5, H-6, H-7), 3.83 (3H, s, $OC\underline{H_3}$, H-30), 5.79 (2H, s, $NC\underline{H_2}$, H-16); ^{13}C (DMSO, 75 MHz; δ, ppm):165.21 (C-30), 156.07 (C-2), 153.23 ($\underline{C}$-OCH_3, C-20), 149.34 (C-12, C-14), 142.42 (C-9), 137.89 (C-8), 136.57 (C-19), 132.43 (C-17), 133.95 (C-24), 132.49 (C-10), 130.29 (C-27), 130.09 (C-25, C-29), 128.97 (C-26, C-28), 128.57 (C-11, C-15), 123.34, 123.05 (C-5, C-6), 121.09 (C-18, C-22), 119.53 (C-4, C-7), 118.06 (C-13), 109.35 (C-21), 55.88 (C-30), 52.43 (C-16) ;ESI-MS *m/z* 525 $M+H^+$; Mol. Formula: $C_{28}H_{20}N_4O_7$. Anal. Calcd.: C 64.12, H 3.84, N 10.68, O 21.35.

3.1.4.6. 2-(3,5-dinitrophenyl)-1-(4-methoxybenzyl)-1H-benzo[d]imidazole (8 f)

The compound **8f** was prepared in a similar manner as described earlier for the synthesis of **(7a)** by the reaction of **(5)** with **6f** (0.201 mg, 1 mmol). The pure compound **8f** was obtained as a brown solid. TLC Solvent System: 8:2 ($CHCl_3$:MeOH); **Yield:** 86%; IR (KBr; υ_{max}, cm^{-1}) 3115.25 (C-H Stretching), 1612.23 (C=C Stretching), 1545.12 (C=N Stretching), 1533.43 (NO_2 sym Stretching), 1238.32 (C-N Stretching), 1224.47(C-O Stretching); ^1H (DMSO, 300 MHz; δ, ppm): 9.04 (2H, s, Ar-H, H-11, H-15), 8.72 (1H, s, Ar-H, H-13), 7.22-7.33 (4H, m, Ar-H, H-4, H-5, H-6, H-7), 7.12 (2H, dd, J=8.6, Ar-H, ring D), 6.87 (2H, dd, J=8.6, Ar-H, ring D), 5.79 (2H, s, NCH_2, C-16), 3.83 (3H, s, $OC\underline{H_3}$, H-23); ^{13}C (DMSO, 75 MHz; δ, ppm): 157.89 ($\underline{C}$-OCH_3, C-20), 153.33 (C-2), 149.34 (C-12, C-14), 142.42 (C-9), 137.89 (C-8), 132.56 (C-17), 130.43 (C-18, C-22), 129.09 (C-19, C-21), 128.97 (C-11, C-15), 123.09 (C-5, C-6), 119.53 (C-4, C-7), 118.06 (C-13), 114.27, 58.34 ($O\underline{C}H_3$, C-23), 52.43 (C-16); ESI-MS *m/z* 405 $[M+H]^+$; Mol. Formula: $C_{21}H_{16}N_4O_5$. Anal. Calcd.: C 62.37, H 3.99, N 13.86, O 19.78.

3.1.4.7. 2-bromo-4-((2-(3,5-dinitrophenyl)-1H-benzo[d]imidazol-1-yl)methyl)-6-methoxyphenol (8g)

The compound **8g** was prepared in a similar manner as described earlier for the synthesis of **(7a)** by the reaction of **(5)** with **6g** (0.295 mg, 1 mmol). The pure compound **8g** was obtained as a brown solid. TLC Solvent System: 8:2 ($CHCl_3$:MeOH); **Yield:** 83%; IR (KBr; υ_{max}, cm^{-1}) 3303.57 (O-H), 3155.25 (C-H Stretching), 1614.27 (C=C Stretching), 1549.82 (C=N Stretching), 1538.23 (NO_2 sym Stretching), 1235.89 (C-N Stretching), 1224.47(C-O Stretching), 720.54 (C-Br); ^1H (DMSO, 300 MHz; δ, ppm): 9.04 (2H, s, Ar-H, H-11, H-15), 8.72 (1H, s, Ar-H, H-13), 7.22-7.33 (4H, m, Ar-H, H-4, H-5, H-6, H-7), 6.77 (2H, s, Ar-H, ring D), 5.79 (2H, s, NCH_2, H-16), 5.35 (1H, s, $O\underline{H}$), 3.82 (3H, s, $OC\underline{H_3}$, H-23); ^{13}C (DMSO, 75 MHz; δ, ppm): 152.33 ($\underline{C}$-OCH_3, C-21), 149.34 (C-2), 143.67 (C-12, C-14), 142.42 (C-OH), 142.03 (C-9), 138.89 (C-8), 132.43 (C-10), 132.05 (C-17), 138.87 (C-20), 128.09 (C-11, C-15), 127.97 (C-18), 123.09 (C-5, C-6), 119.53 (C-4, C-7), 118.06 (C-13), 111.78 (C-Br, C-19), 109.42 (C-22), 58.84 ($O\underline{C}H_3$, C-23), 51.13 (C-16); ESI-MS *m/z* 500 $M+H^+$; Mol. Formula: $C_{21}H_{15}BrN_4O_6$. Anal. Calcd.: C 50.52, H 3.03, N 11.22, O 19.23, Br 16.00.

3.1.4.8. 2-(3,5-dinitrophenyl)-1-(2-methoxybenzyl)-1H benzo[d]imidazole (8 h)

The compound **8h** was prepared in a similar manner as described earlier for the synthesis of **(7a)** by the reaction of **(5)** with **6h** (0.201 mg, 1 mmol). The pure compound **8h** was obtained as a brown solid. TLC Solvent System: 8:2 ($CHCl_3$:MeOH); **Yield:** 89%; IR (KBr; υ_{max}, cm^{-1}) 3115.25 (C-H Stretching), 1612.23 (C=C Stretching), 1545.12 (C=N Stretching), 1533.43 (NO_2 sym Stretching), 1260.62 (C-N Stretching), 1224.47(C-O Stretching); ^1H (DMSO, 300 MHz; δ, ppm): 9.04 (2H, s, Ar-H, H-11, H-15), 8.72 (1H, s, Ar-H, H-13), 6.48-7.19 (8H, m, Ar-H, H-4, H-5, H-6, H-7, Ring D), 5.49 (2H, s, NCH_2, H-16), 3.83 (3H, s, OCH_3, H-23); ^{13}C (DMSO, 75 MHz; δ, ppm): 158.89 ($\underline{C}$-OCH_3, C-18), 153.33 (C-2), 149.34 (C-12, C-14), 142.42 (C-9), 137.89 (C-8), 130.43 (C-10), 129.09 (C-11, C-15), 128.97 (C-17), 126.70 (C-19), 125.78 (C-21), 123.09 (C-5, C-6), 120.99 (C-20), 119.53 (C-4, C-7), 118.28 (C-13), 112.06 C-22), 56.34 (OCH_3, C-23), 46.43 (C-16); ESI-MS *m/z* 405 $M+H^+$; Mol. Formula: $C_{21}H_{16}N_4O_5$. Anal. Calcd.: C 62.37, H 3.99, N 13.86, O 19.78.

3.2. Biological activity

Experiment 1: Determination of free radical scavenging activity by 1, 1-diphenyl-2-picrylhydrazyl (DPPH) method

When a solution of DPPH is mixed with that of a substrate that can donate a hydrogen atom, then this in order to evaluate the antioxidant potential through free radical scavenging by the test samples, the change in optical density of DPPH radicals is monitored at 517 nm. A control was prepared by dissolving DPPH (7.89 mg, 0.2 mM) in 100 ml ethanol instead of samples. Then the solution was kept in dark for 2 hrs. Sample solutions were prepared by dissolving 3mg of each sample in 3 ml of methanol separately. 1ml of 0.2 mM DPPH methanolic solution was mixed with 3ml of each sample solutions of various concentrations (100µg/ml, 250µg/ml, 500µg/ml and 1000µg/ml) separately. The mixture was left in the dark for 30 min before its absorbance was rerecorded by at UV spectrophotometer 517 nm. Standard solution of BHT was prepared by dissolving 10 mg of BHT in 10 ml of methanol (1000 ppm) and then diluted it to prepare the different concentration (100µg/ml, 250µg/ml, 500µg/ml and 1000µg/ml) solutions. The DPPH radical scavenging activity (%) was calculated using the following formula.

$$\text{DPPH radical scavenging activity (\%)} = \frac{A_{control} - A_{sample}}{A_{control}} \times 100$$

Where, $A_{control}$ was the absorbance of the control and A_{sample} was the absorbance in the presence of the sample.

Experiment 2: Determination of antimicrobial activity

All glass apparatus were sterilized in hot air oven at 160°C for 1 hour 15 g of Muller-Hinton Agar and 32.5g of Sabouraud Dextrose Agar for bacteria were separately dissolved in 500 ml of distilled water by heating on water bath. The medium (100 ml) each was distributed, in 4 conical flasks and plugged with cotton, sterilized by autoc1aving at 15-lbs/sq inch. pressure at 121°C for a period of 15 minutes. After cooling the media to 45ºC, 1 ml of the 48 hours broth culture of stain was inoculated into each of the 4 different conical flasks.The medium was shaken thoroughly and distributed on petridishes (100 ml each). It was kept aside for some time for solidification and cups were made with sterile cork borer (internal diameter 6mm). Nine cups were made in each petridish and 0.2 ml of the test compounds solutions were added. To each plate 0.2 ml of the standard drug solution and control solution were also added. All the dilutions were made by using DMSO. Plates were kept at room temperature for 2 hours for diffusion and then plates were incubated for 16 hours at 37°C for bacteria. The zones of inhibition were measured at the end of stipulated time. The results obtained are recorded in **Table 3**.

4. Conclusion

We have synthesized total sixteen novel benzimidazole analogs along with the synthesis of two known benzimidazole analogs **4** and **5** precursors. We have synthesized the two series (**7a-7h** and **8a-8h**) of eight analogs each and screened all the synthesized analogs for antioxidant (free radical scavenging activity) and antibacterial activity.The biological studies revealed that out of all synthesized analogs only **8a** with phenyl substituent and **8g** with 3,5-dinitrophenyl substituent showed the highest free radical scavenging activity with % inhibition 98.73 % and 98.91 % as compared to standard BHT (99.24 %) at 100 µg/ml concentration. In the evaluation of antibacteraial activity it was found that all the novel analogs (**7a-7h** and **8a-8h**) were moderately active against bacterial strains.

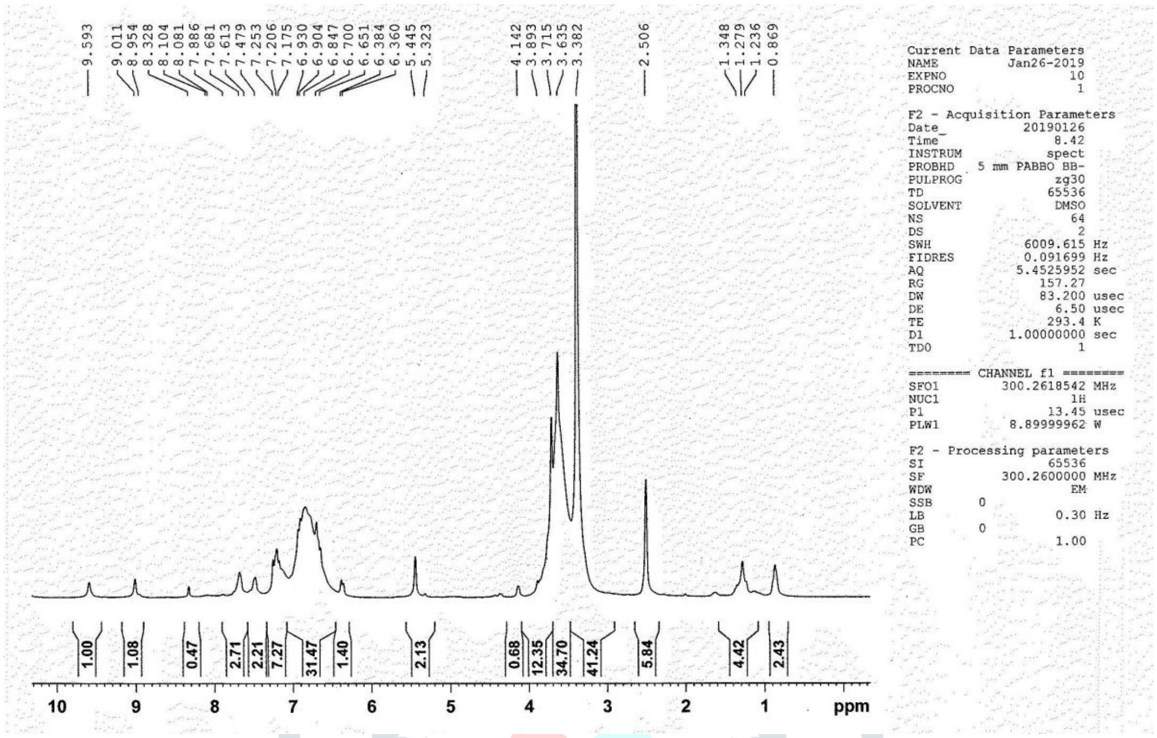
5. Acknowledgement

The authors acknowledge the Department of Chemistry, University of Lucknow, Lucknow, U.P. for supporting the NMR and FTIR spectroscopic facilities. The authors also acknowledge the SAIF division CDRI, Lucknow for HRMS analysis.

6. References

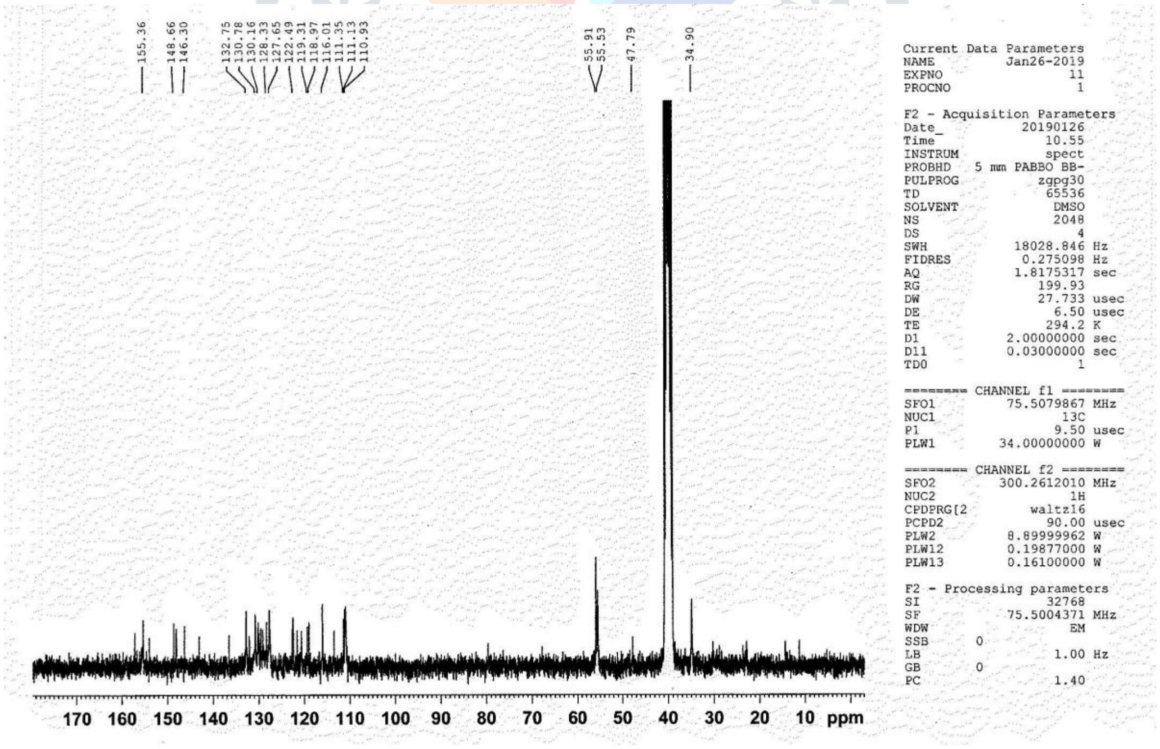
1. B. Yogita, O. Silakari, The therapeutic journey of benzimidazoles: A review. *Bioorganic & medicinal chemistry*, 20(21) (2012), pp. 6208-6236. https://doi.org/10.1016/j.bmc.2012.09.013
2. F. Hobrecker, Reduction-products of nitracetamide compounds. *Deut Chem Ges Ber*, 5 (1872), pp. 920-924.
3. S. Jyoti, P. K. Soni, R. Bansal, A. K. Halve, Synthetic approaches towards benzimidazoles by the reaction of o-phenylenediamine with aldehydes using a variety of catalysts: A review. *Current Organic Chemistry*, 22, (23) (2018), pp. 2280-2299. https://doi.org/10.2174/1385272822666181024120156
4. H.H. Yin, A new imidazole alkaloid from the marine sponge Leucetta microrhaphis. *The Journal of Organic Chemistry*, 57(7) (1992), pp. 2176-2178. https://doi.org/10.1021/jo00033a051
5. B. Ritchu, S. Arora, A Comprehensive Review on Therapeutic Potential of Benzimidazole: A Miracle Scaffold, *Journal of Pharmaceutical Technology, Research and Management*, 8(1) (2020), pp. 23-29. https://doi.org/10.15415/jptrm.2020.81004
6. G. Kumaraswamy, G. Brahmeshwari, M. Ravichander, U. Kusuma. Synthesis, antibacterial and molecular docking studies of new benzimidazole derivatives. *Egyptian journal of basic and applied sciences*, 4 (4) (2017), pp. 303-309. https://doi.org/10.1016/j.ejbas.2017.09.002
7. P. Chittari, H. Wang, S. R. Mulligan, Z. Chen, T. M. Stark, B. T. Gregg. Microwave-assisted synthesis of 1-aryl-1H-indazoles via one-pot two-step Cu-catalyzed intramolecular N-arylation of arylhydrazones, *Tetrahedron letters*, 46 (44) (2005), pp. 7553-7557. https://doi.org/10.1016/j.tetlet.2005.08.143
8. D. Nikunj, H. Bhatt, V. Chauhan, K. Joshi. Synthesis and Structural Elucidation of 1, 4 Dihydropyrimido [1, 2-a] Benzimidazole, *Asian Journal of Chemical Sciences*, (2022): pp. 10-16. https://doi.org/10.9734/ajocs%2F2022%2Fv11i119112
9. A. K. Dinesh, P. K. Goyal, Synthetic and Biological Aspects of Benzimidazole Derivatives, *Chemistry & Biology Interface*, 11(1) (2021).
10. S. Nomi, J. Banerjee, S. Srivastava, A review on chemistry and biological significance of benzimidazole nucleus, *IOSR J Pharm*, 4(12) (2014), pp. 28-41.
11. M. T. Michael, J. Girardet, J. A. Walker, G. W. Koszalka, S. D. Chamberlain, J. C. Drach, L. B. Townsend. Design, synthesis, and antiviral activity of α-nucleosides: D-and L-isomers of lyxofuranosyl-and (5-deoxylyxofuranosyl) benzimidazoles, *Journal of medicinal chemistry*, 41(8) (1998), pp. 1242-1251. https://doi.org/10.1021/jm970545c
12. K. S. Rangappa, C. K. Rajappa, S. A. Patil, B. M. Nagaraja, Benzimidazole-core as an antimycobacterial agent, *Pharmacological Reports*, 68(6) (2016), pp. 1254-1265. https://doi.org/10.1016/j.pharep.2016.08.002
13. W. Miao, X. Han, Z. Zhou, New substituted benzimidazole derivatives: A patent review , *Expert opinion on therapeutic patents*, 25(5) (2015), pp. 595-612. https://doi.org/10.1517/13543776.2015.1015987

14. K. S. Rangappa, A. Hiremathad, S. Budagumpi, B. M. Nagaraja, Comprehensive review in current developments of benzimidazole-based medicinal chemistry, *Chemical biology & drug design*, 86(1) (2015), pp.19-65. **https://doi.org/10.1111/cbdd.12462**
15. Y. Geeta, S. Ganguly, Structure activity relationship (SAR) study of benzimidazole scaffold for different biological activities: A mini-review, *European Journal of Medicinal Chemistry*, 97 (2015), pp. 419-443. doi, 10.1016/j.ejmech.2014.11.053
16. A. S. RAHMAN, B. K. Das, M. S. Hossain, U. T. T. O. M. Kumar, A. S. S. Rouf., Synthesis and antioxidant activity of 2-substituted-5-nitro benzimidazole derivatives, *Int J Pharm Pharm Sci*, 9(1) (2017), pp. 308-310. https://doi.org/10.22159/ijpps.2017v9i1.14972
17. S. Y. Cho, S. K. Kang, S. S. Kim, C. HG, J. K. Choi, E. K. Yum. Synthesis and SAR of benzimidazole derivatives containing oxycyclic pyridine as a gastric H+/K+-ATPase inhibitors, Bull Korean Chem Soc, 22(2001), pp. 1217-23
18. K.bSreena, R. Ratheesh, M. Rachana, M. Poornima, C. Shyni, Synthesis and anthelmintic activity of benzimidazole derivatives. (2009).
19. R. Yassine, M. H. Geesi, O. Ouerghi, O. Dehbi, A. Elsanousi, R. Azzallou. Synergistic Catalytic Effect of the Combination of Deep Eutectic Solvents and Hierarchical H-TiO2 Nanoparticles toward the Synthesis of Benzimidazole-Linked Pyrrolidin-2-One Heterocycles: Boosting Reaction Yield, *Polycyclic Aromatic Compounds*, (2021), pp. 1-15. https://doi.org/10.1080/10406638.2021.1991397
20. D., Seref, A. C. K., I. Kayagil, U. Uçucu, R. Beis. Synthesis and analgesic activities of some 2-(benzazolylacetyl) amino-3-ethoxycarbonylthiophene derivatives, *Phosphorus, Sulfur, and Silicon and the Related Elements*, 180(8) (2005), pp. 1841-1848. https://doi.org/10.1080/104265090889503
21. A. S. Ibrahim. Synthetic approaches to benzimidazoles from o-phenylenediamine: A literature review, *Journal of Saudi Chemical Society*, 21(2) (2017), pp. 229-237. https://doi.org/10.1016/j.jscs.2016.08.001
22. G. Singh, K. Maninderjit, C. Mohan, Benzimidazoles: The latest information on biological activities, *Int. Res. J. Pharm*, 4(1) (2013), pp. 82-87.
23. S. Hemant, N. Kaur, N. Singh, D. O. Jang, Synergetic catalytic effect of ionic liquids and ZnO nanoparticles on the selective synthesis of 1, 2-disubstituted benzimidazoles using a ball-milling technique, *Green chemistry*, 17(8) (2015), pp. 4263-4270. **https://doi.org/10.1039/C5GC00536A**
24. S. Rohit, F. A. Sofi, P. Rana, P. V. Bharatam, Bimetallic Cu–Mn B spinel oxide catalyzed oxidative synthesis of 1, 2-disubstituted benzimidazoles from benzyl bromides, *New Journal of Chemistry*, 43(10)(2019), pp. 4013-4016. **https://doi.org/10.1039/C8NJ05504A**
25. E. Tsolaki, P. Nobelos, A. Geronikaki, E. A. Rekka, Selected heterocyclic compounds as antioxidants, Synthesis and biological evaluation, *Current Topics in Medicinal Chemistry*, 14(22) (2014), pp. 2462-2477.
26. W. B. Michael., J. Feng, Z. Zhang, R. J. Skene, L. Shi, C. L. Caster, D. B. Kassel, R. Xu, S. L. Gwaltney II, Structure-based design and synthesis of benzimidazole derivatives as dipeptidyl peptidase IV inhibitors, *Bioorganic & medicinal chemistry letters*, 18(7) (2008), pp. 2362-2367. https://doi.org/10.1016/j.bmcl.2008.02.071
27. S. Ramineni, K. R. Kumar, P.V. V. Satyanarayana, Facile and efficient method for synthesis of benzimidazole derivatives catalyzed by zinc triflate, *Green and Sustainable Chemistry*, (2014). DOI:10.4236/gsc.2014.41006
28. J. Deepkumar, K. Parikh, Synthesis and evaluation of novel benzimidazole derivatives as antimicrobial agents, *Medicinal Chemistry Research*, 23(3) (2014), pp. 1290-1299. https://doi.org/10.1007/s00044-013-0732-z
29. K. Gopakumar, A. Sivan, Exploring the Versatility of Benzimidazole Scaffolds as Medicinal Agents: A Brief Update.
30. R. Mudumala, V.Gangireddy, C. S. Reddy, Y. T. Jeong, Molybdate sulfuric acid (MSA): an efficient reusable catalyst for the synthesis of tetrahydrobenzo [4, 5] imidazo [2, 1-b] quinazolin-1 (2 H)-ones under solvent-free conditions and evaluation of their in vitro bioassay, *RSC advances*, 5(15) (2015), pp. 11423-11432. https://doi.org/10.1039/C4RA13440K
31. C. Chao, H. Wang, Y. Hu, C. Liu, S. Ma, X. Li, J. Cao, S. Ma, Synthesis and antibacterial activity of novel 4 ″-O-benzimidazolyl clarithromycin derivatives, *European journal of medicinal chemistry*, 46(7) (2011), pp. 3105-3111. https://doi.org/10.1016/j.ejmech.2011.04.004
32. S. Padalkar, V. B. N. Borse, V. D. Gupta, K. R. Phatangare, V. S. Patil, P. G. Umape, N. Sekar, Synthesis and antimicrobial activity of novel 2-substituted benzimidazole, benzoxazole and benzothiazole derivatives, *Arabian Journal of Chemistry*, 9, (2016), pp. S1125-S1130. https://doi.org/10.1016/j.arabjc.2011.12.006
33. S. Weijie, T. Zhang, Y. Li, D. She, W. Pan, Z. Gao, J. Ning, X. Mei, Synthesis and biological activity of novel benzimidazole derivatives as potential antifungal agents, *Journal of Pesticide Science*, 41(1) (2015), pp. 15-19. https://doi.org/10.1584/jpestics.D15-037
34. Y. Fatih, N. Karaali, S. Şaşmaz, Microwave-assisted synthesis of some nitro-benzimidazoles and their salicyl and isatin Schiff bases, *Bulletin of the Chemical Society of Ethiopia*, 31(2) (2017), pp. 351-359. **DOI:** 10.4314/bcse.v31i2.17
35. A. A. SELEN, G. KILCIGİL, E. D. ÖZDAMAR, T. ÇOBAN, B. Eke, Synthesis and evaluation of antioxidant activities of novel 1, 3, 4-oxadiazole and imine containing 1H-benzimidazoles, *Turkish Journal of Chemistry*, 39(1), (2015), pp. 42-53. https://doi.org/10.3906/kim-1403-44
36. K. Canan, G. Ayhan-Kılcıgil, S. Özbey, F. B. Kaynak, M. Kaya, T. Çoban, B. Can-Eke, Synthesis and antioxidant properties of novel N-methyl-1, 3, 4-thiadiazol-2-amine and 4-methyl-2H-1, 2, 4-triazole-3 (4H)-thione derivatives of benzimidazole class, *Bioorganic & medicinal chemistry*, 16, (8) (2008): pp. 4294-4303. https://doi.org/10.1016/j.bmc.2008.02.077
37. Z. A. Alagoz, C. Kus, T. Çoban, Synthesis and antioxidant properties of novel tetrahydro-naphthalene fragments, *J Enzym Inhib Med Chem*, 20 (2004), pp. 325.
38. K. Kantharaju, Synthesis and pharmacological profile of benzimidazoles, *Chemistry and applications of benzimidazole and its derivatives. London, UK: IntechOpen*, (2019), pp. 51-69.



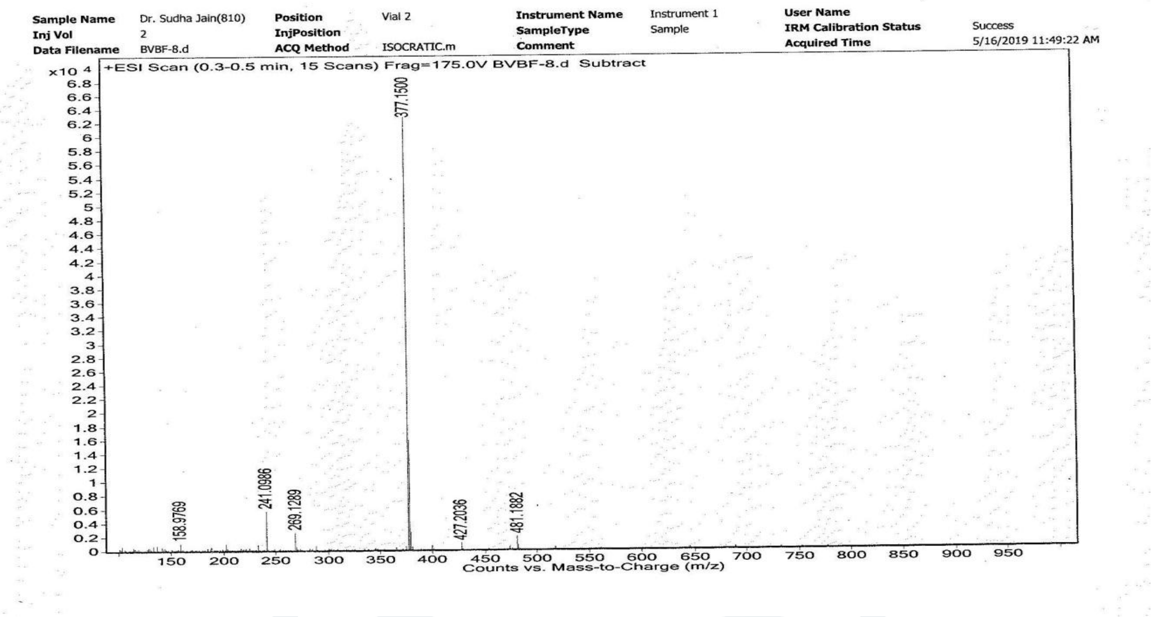
```
Current Data Parameters
NAME             Jan26-2019
EXPNO                    10
PROCNO                    1

F2 - Acquisition Parameters
Date_              20190126
Time                   8.42
INSTRUM               spect
PROBHD   5 mm PABBO BB-
PULPROG                zg30
TD                    65536
SOLVENT                DMSO
NS                       64
DS                        2
SWH                6009.615 Hz
FIDRES             0.091699 Hz
AQ                5.4525952 sec
RG                   157.27
DW                   83.200 usec
DE                     6.50 usec
TE                    293.4 K
D1               1.00000000 sec
TD0                       1

======== CHANNEL f1 ========
SFO1            300.2618542 MHz
NUC1                     1H
P1                    13.45 usec
PLW1             8.89999962 W

F2 - Processing parameters
SI                    65536
SF              300.2600000 MHz
WDW                      EM
SSB      0
LB                     0.30 Hz
GB       0
PC                     1.00
```

^{1}H NMR Spectra of 7h

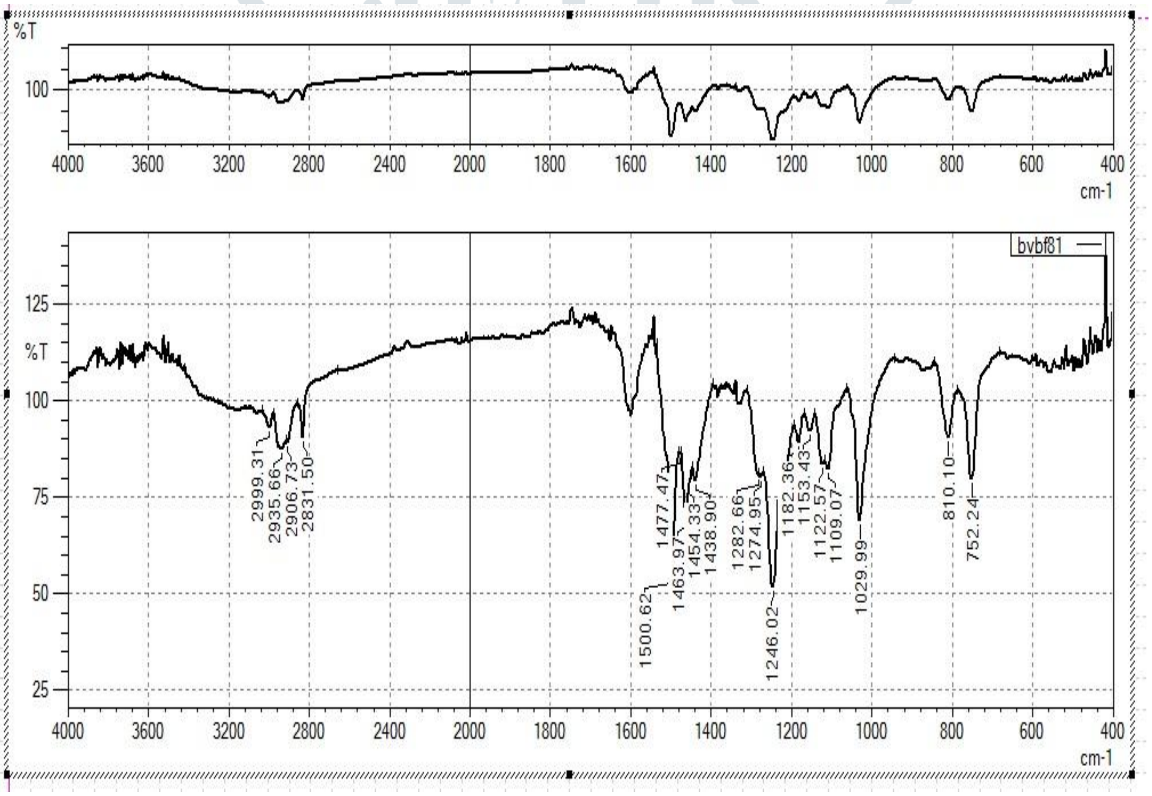


```
Current Data Parameters
NAME             Jan26-2019
EXPNO                    11
PROCNO                    1

F2 - Acquisition Parameters
Date_              20190126
Time                  10.55
INSTRUM               spect
PROBHD   5 mm PABBO BB-
PULPROG              zgpg30
TD                    65536
SOLVENT                DMSO
NS                     2048
DS                        4
SWH               18028.846 Hz
FIDRES             0.275098 Hz
AQ                1.8175317 sec
RG                   199.93
DW                   27.733 usec
DE                     6.50 usec
TE                    294.2 K
D1               2.00000000 sec
D11              0.03000000 sec
TD0                       1

======== CHANNEL f1 ========
SFO1             75.5079867 MHz
NUC1                    13C
P1                     9.50 usec
PLW1            34.00000000 W

======== CHANNEL f2 ========
SFO2            300.2612010 MHz
NUC2                     1H
CPDPRG[2            waltz16
PCPD2                 90.00 usec
PLW2             8.89999962 W
PLW12            0.19877000 W
PLW13            0.16100000 W

F2 - Processing parameters
SI                    32768
SF               75.5004371 MHz
WDW                      EM
SSB      0
LB                     1.00 Hz
GB       0
PC                     1.40
```

^{13}C NMR Spectra of 7h



Mass Spectra of 7h



IR spectra of 7h