



## Research Article

## SYNTHESIS AND PHARMACOLOGICAL EVALUATION OF 6-SUBSTITUTED 2-AMINOBENZOTHAZOLES

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## ABSTRACT

Heterocyclic compounds and analogues have attracted strong interest due to their biological and pharmacological properties. Nitrogen containing heterocyclics, Benzothiazoles are an important class of heterocyclic compounds and the nucleus containing compounds involved in research aimed at evaluating new products that possess biological activities such as antibacterial, antifungal, anthelmintic, antidiabetic, and anticancer agents. In the present study various derivatives of 6-substituted 2-aminobenzothiazoles were prepared and evaluated for their antibacterial, antifungal, antioxidant and cytotoxic activity by various standard methods. Among the synthesized compounds, most of the compounds exhibited potent activity when compared with that of the standard drugs. Hence the versatile synthetic applicability and biological activity of these heterocyclic compounds will help the chemist to plan, organize and implement new approaches towards discovery of novel derivatives of Benzothiazoles.

**Keywords:** 2-aminobenzothiazoles, arylidines, anthelmintic, antidiabetic, cytotoxic activity.

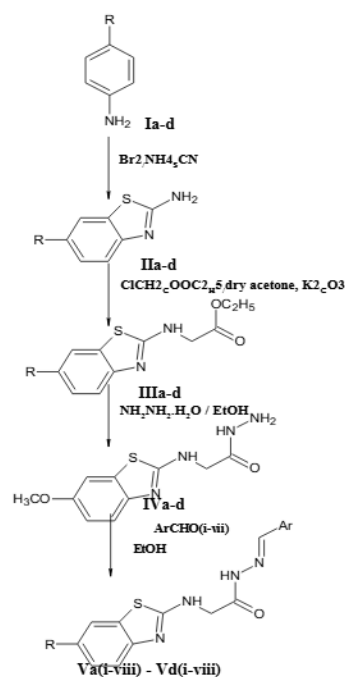
## INTRODUCTION

Benzothiazole is a privileged bicyclic ring system. Due to its potent and significant biological activities it has great pharmaceutical importance. Hence, synthesis of this compound is of considerable interest. The small and simple benzothiazole nucleus if present in compounds involved in research aimed at evaluating new products that possess interesting biological activities. 2-substituted benzothiazole has emerged in its usage as a core structure in the diversified therapeutically applications. The studies of structure–activity relationship interestingly reveal that change of the structure of substituent group at C-2 position commonly results the change of its biological activity. Among those 2-substituted benzothiazole derivatives with fluorine substituted molecules have already received considerable attention due to their potential bioactivities<sup>1-4</sup>.

2-substituted benzothiazoles are most commonly synthesized via one of two major routes. The most common direct method involves the condensation of an *o*-aminothiophenol with a substituted aromatic aldehyde, carboxylic acid, acyl chloride or nitrile. This method, however, is often not appropriate for many substituted 2-arylbenzothiazoles due to the difficulties encountered in the synthesis of the readily oxidisable 2-aminothiophenols bearing substituent groups. The other methods used extensively in the laboratories which are based on the potassium ferricyanide (Jacobsen cyclization) radical cyclization of thiobenzanilides which involve cyclization onto either carbon atom *ortho* to the anilido nitrogen produces only one product, hence, the Jacobsen cyclization is a highly effective strategy for the synthesis of benzothiazole<sup>5</sup>.

## MATERIALS AND METHODS

## SCHEME



Where R = - Cl, - F, - CH<sub>3</sub>, - OCH<sub>3</sub>  
 Ar = - C<sub>6</sub>H<sub>5</sub>, 4-OCH<sub>3</sub>C<sub>6</sub>H<sub>4</sub>, 4-N(CH<sub>3</sub>)<sub>2</sub>C<sub>6</sub>H<sub>4</sub>, 4-ClC<sub>6</sub>H<sub>4</sub>, 3,4-diOCH<sub>3</sub>C<sub>6</sub>H<sub>3</sub>, 2-OHC<sub>6</sub>H<sub>4</sub>, 3-NO<sub>2</sub>C<sub>6</sub>H<sub>4</sub>,  
 -CH=CH-C<sub>6</sub>H<sub>5</sub>

### Synthesis of 6-Substituted 2-Aminobenzothiazoles (II)

A mixture of *p*-substituted anilines (0.1mole) and ammonium thiocyanate (0.2mole) in 150 ml of glacial acetic acid were cooled in an ice bath and stirred mechanically. To the solution, bromine (0.2mole) in 25 ml of glacial acetic acid was added drop wise at such a rate to keep the temperature below 10° C throughout the addition. Stirring was continued for another thirty minutes after the bromine addition. The precipitate of the Benzothiazole hydrobromide was collected, dissolved in hot water and basified with a saturated sodium carbonate solution. The product was filtered under vacuum, washed with water, dried and recrystallized from the appropriate solvent<sup>6</sup>.

Four compounds (II<sub>a-d</sub>) were prepared by following above procedure. The physical data of all four compounds synthesized are given in Table 1.

### Synthesis of 6-Substituted Ethyl-2-(benzo[d]thiazol-2yl-amino)acetate (III)

0.01mole of (6-substituted 2-aminobenzothiazoles) was dissolved in 30ml of dry acetone. To this ethylchloroacetate (0.01mole) and freshly fused potassium carbonate (0.01mole) were added and refluxed on an oil bath at 120-140° C for 20-24 hrs and the progress of the reaction was monitored by TLC (Chloroform: n-hexane; 7:3 V/V). The reaction mixture was then poured into crushed ice; precipitate was filtered and washed with cold water. The product was dried and purified by crystallization from aqueous alcohol<sup>6</sup>.

Adopting the above procedure, four different 6-Substituted Ethyl-2-(benzo[d]thiazol-2ylamino)acetates were prepared, purified and characterized. The physical and analytical data are presented in Table 2.

### General procedure for the synthesis of 6-substituted 2-(benzo(d)thiazol-2yl-amino)acetohydrazides (IV)

A mixture of 6-Substituted Ethyl-2-(benzo[d]thiazol-2yl-amino)acetate (III) (0.01mole) and hydrazine hydrate (99%, 0.015mole) in absolute ethanol (25ml) was heated under reflux on a steam bath for 16-18 hrs. The solvent was removed from the reaction mixture to a possible extent and cooled. The compound was filtered, washed with cold water and dried. The product was purified by using alcohol<sup>6</sup>.

Adopting the above procedure four different 6-substituted 2-(benzo[d]thiazol-2ylamino)acetohydrazides were prepared and characterized. The physical and analytical data is presented in Table 3.

### General procedure for the synthesis of 6-substituted 2-(benzo(d)thiazol-2ylamino)-N-arylidene acetohydrazides (V)

An equimolar concentration of (0.01mole) each mixture of 6-substituted 2-(benzo[d]thiazol-2ylamino)acetohydrazides (IV) and differently substituted aromatic aldehydes (i-viii) in ethanol (25ml) and add 2-3 drops of acetic acid, the reaction mixture was refluxed on a water bath for 3-4 hrs. The solvent was distilled off under reduced pressure and the residue was poured into ice cold water to obtain the product. The compound was filtered off, washed with cold water and dried. The crude product was recrystallized from ethanol<sup>6</sup>.

Thirty two different benzylidene derivatives were synthesized and characterized by adopting the above procedure. The physical, analytical data are given in Table 4.

Four different known and unknown 2-amino-6-substituted-(1,3)benzothiazoles (II) have been prepared for the purpose from four different anilines (I), viz: 4-chloro aniline (I<sub>a</sub>, R=Cl); 4-fluoro aniline (I<sub>b</sub>, R=F); 4-methyl aniline (I<sub>c</sub>, R=CH<sub>3</sub>); and 4-methoxy aniline (I<sub>d</sub>, R=OCH<sub>3</sub>). Each of these anilines have been subjected to the reaction with ammonium thiocyanate in presence of bromine in acetic acid followed by the basification with ammonia. The product obtained from each of such reaction has been purified and identified in case of known compounds, based on their literature data and characterized the new compounds, on the basis of their spectral (IR, <sup>1</sup>H NMR & Mass) data.

**Infrared Spectrum** (KBr; cm<sup>-1</sup>) has been found to show the following absorptions: 3320 (NH, str), 3040 (C-H, str, aromatic), 2928 (C-H, str), 1695 (C=N, str), 1047 (C-F, str), 605 (C-S, str).

**<sup>1</sup>H NMR Spectrum** of the compound [in (CDCl<sub>3</sub>) 300 MHz] revealed the following characteristic proton signals (δ, ppm): 3.89 (s, 2H, NH<sub>2</sub>,br); 7.25-8.02 (m,3H, C<sub>4</sub>, C<sub>5</sub> & C<sub>7</sub> of benzthiozole).

**Mass Spectrum** (ESI, positive) of the compound has recorded its molecular ion: [M<sup>+</sup>] at m/z 168 equal to its mass (Mol.wt).

CHN analyses Calcd. for C<sub>7</sub>H<sub>5</sub>FN<sub>2</sub>S: C, 49.99, H, 3.00, N, 16.66. Found H, 49.82, H, 2.97, N, 16.59.

Based on the data recorded, the resultant compound has been characterized as **2-amino-6-fluoro-(1,3)benzothiazole (II<sub>b</sub>)**.

### Antibacterial Activity

The antibacterial activity of synthesized compounds will be conducted against two gram positive bacteria viz., *Bacillus subtilis* and *Staphylococcus aureus* and two gram negative bacteria viz., *Escherichia coli* and *Proteus vulgaris* by using cup-plate method<sup>7,8,9</sup>.

Culture Medium: Nutrient broth was used for the preparation of inoculum of the bacteria and nutrient agar for the screening method.

The test organisms will be subcultured using nutrient agar medium. The tubes containing sterilized medium will be inoculated with respective bacterial strain. After incubation at 37°C ± 1°C for 24 hours, store in refrigerator. The stock cultures will be maintained. Bacterial inoculum is prepared by transferring a loopful of stock culture to nutrient broth (100 ml) in conical flasks (250 ml). Then flasks have to be incubated at 37°C ± 1°C for 48 hours before the experimentation. (Table 5)

### Antifungal Activity

All those compounds screened for antibacterial activity were also tested by the same Cup-plate method for their antifungal activity. The fungi employed for screening were: *Candida albicans* and *Aspergillus niger*<sup>10,11</sup>.

The test organisms are sub-cultured using potato-dextrose-agar medium. The tubes containing sterilized medium are inoculated with test fungi and after incubation at 25°C for 48 hours, they are stored at 4°C in refrigerator.

The inoculum is prepared by taking a loopful of stock culture to about 100ml of nutrient broth, in 250 ml conical flasks. The flasks are incubated at 25°C for 24 hours before use.

The solutions of test compounds are prepared by a similar procedure described under antibacterial activity. A reference standard (1 mg/ml) was prepared by dissolving 10 mg of Cotrimoxazole in 10 ml of Dimethylformamide (AnalaR grade). Further, the dilution is made with dimethylformamide itself to obtain a solution of 100 µg/ml concentration. (Table 6)

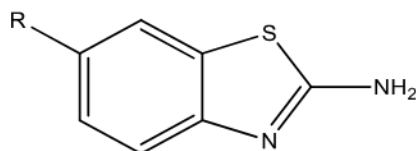
**Antioxidant Activity****DPPH method**

The method of Liyana-Pathiana and Shahidi (2005) was used for the determination of scavenging activity of DPPH free radical. To 1 ml of 0.135 Mm DPPH in methanol was mixed with 1 ml of test compounds ranging from 20-100 µg/ml. The reaction mixture was vortexed thoroughly and kept in dark at room temperature for 30 min. The absorbance was measured spectrophotometrically at 517 nm<sup>12,13</sup>. The scavenging ability of the test compounds was calculated using the standard equation. The % inhibition and IC<sub>50</sub> values were given in Tables 7.

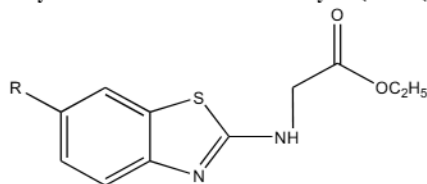
**Cytotoxic Activity****MTT Assay**

Stock solution of 10mg/ml stock solution in DMSO is prepared, from the above stock various dilutions were made with sterile water to get required concentration.

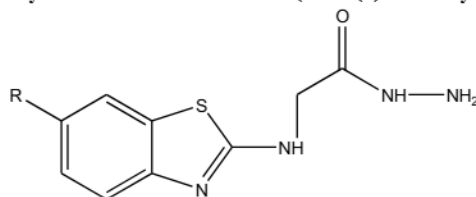
Toxicity of test compound in cells was determined by MTT assay based on mitochondrial reduction of yellow MTT tetrazolium dye to a highly colored blue formazan product. 1x10<sup>4</sup> Cells (counted by Trypan blue exclusion dye method) in 96- well plates were incubated with compounds with series of concentrations tested for 48 hrs at 37°C in DMEM/MEM with 10% FBS medium. Then the above media was replaced with 90µl of fresh serum free media and 10 µl of MTT reagent (5mg/ml) and plates were incubated at 37°C for 4h, there after the above media was replaced with 200µl of DMSO and incubated at 37°C for 10min. The absorbance at 570nm was measured on a spectrophotometer (spectra max, Molecular devices) IC<sub>50</sub> values were determined from plot: % inhibition (from control) versus concentration<sup>3,14</sup>. (Table 8)

**Table 1: Physical and analytical data of 6-substituted 2-aminobenzothiazoles**

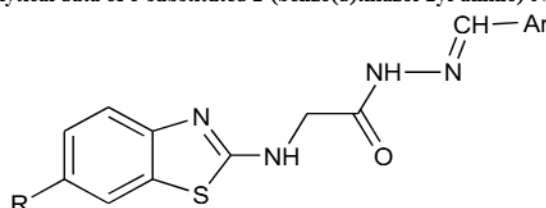
Compound	R	M.P. (°C)	Yield %	Molecular Formula	Mol. weight	Solvent for recrystallization	Elemental Analyses		
							Calculated (Found)		
							C	H	N
IIa	Cl	198-201	95	C <sub>7</sub> H <sub>5</sub> ClN <sub>2</sub> S	184	50% ethanol	45.53 (45.51)	2.73 (2.71)	15.17 (15.14)
IIb	F	184-187	90	C <sub>7</sub> H <sub>5</sub> FN <sub>2</sub> S	168	Ethanol	49.99 (49.92)	3.00 (2.97)	16.66 (16.62)
IIc	CH <sub>3</sub>	134-137	80	C <sub>8</sub> H <sub>5</sub> N <sub>2</sub> S	164	Benzene	58.51 (58.49)	4.91 (4.89)	17.06 (17.03)
IIId	OCH <sub>3</sub>	165-168	98	C <sub>8</sub> H <sub>8</sub> N <sub>2</sub> OS	180	Ethanol	53.31 (53.39)	4.47 (4.44)	15.54 (15.52)

**Table 2: Physical and analytical data of 6-Substituted ethyl-2-(benzo(d)thiazol-2-yl-amino)acetates**

Code	R	M.P. (°C)	Yield %	Molecular Formula	Molecular weight	Solvent for recrystallization	Elemental Analyses		
							Calculated (Found)		
							C	H	N
IIIa	Cl	162-165	82	C <sub>11</sub> H <sub>11</sub> ClN <sub>2</sub> O <sub>2</sub> S	270	Aq. Ethanol	48.80 (48.79)	4.10 (4.08)	10.35 (10.32)
IIIb	F	158-162	85	C <sub>11</sub> H <sub>11</sub> FN <sub>2</sub> O <sub>2</sub> S	254	Aq. Ethanol	51.96 (51.94)	4.36 (4.34)	11.02 (11.00)
IIIc	CH <sub>3</sub>	94-98	78	C <sub>12</sub> H <sub>14</sub> N <sub>2</sub> O <sub>2</sub> S	250	Aq. Ethanol	57.58 (57.55)	5.64 (5.62)	11.19 (11.17)
IIIId	OCH <sub>3</sub>	146-149	95	C <sub>12</sub> H <sub>14</sub> N <sub>2</sub> O <sub>3</sub> S	266	Aq. Ethanol	54.12 (54.10)	5.30 (5.28)	10.52 (10.49)

**Table 3: Physical and analytical data of 6-substituted 2-(benzo(d)thiazol-2-yl-amino)acetohydrazides**

Code	R	M.P. (°C)	Yield %	Molecular Formula	Molecular weight	Crystallization solvent	Elemental Analyses		
							Calculated (Found)		
							C	H	N
IVa	Cl	180-183	90	C <sub>9</sub> H <sub>9</sub> ClN <sub>4</sub> OS	256	Aq. Ethanol	42.11 (42.09)	3.53 (3.51)	21.82 (21.79)
IVb	F	172-175	92	C <sub>9</sub> H <sub>9</sub> FN <sub>4</sub> OS	240	Aq. Ethanol	44.99 (44.95)	3.78 (3.72)	23.32 (23.29)
IVc	CH <sub>3</sub>	178-182	85	C <sub>10</sub> H <sub>12</sub> N <sub>4</sub> OS	236	Aq. Ethanol	50.83 (50.80)	5.12 (5.09)	23.71 (23.69)
IVd	OCH <sub>3</sub>	182-184	90	C <sub>10</sub> H <sub>12</sub> N <sub>4</sub> O <sub>2</sub> S	252	Aq. Ethanol	47.61 (47.59)	4.79 (4.78)	22.21 (22.19)

**Table 4: Physical and analytical data of 6-substituted 2-(benzo(d)thiazol-2-yl-amino)-N-arylidene acetohydrazides**

Compound	R	Ar	Molecular Formula	Mol. Wt.	M.P. (°C)	Yield (%)	Elemental analyses		
							Calculated (Found)		
							C	H	N
Vai	-Cl	-C <sub>6</sub> H <sub>5</sub>	C <sub>16</sub> H <sub>13</sub> ClN <sub>4</sub> OS	344	172-175	70	55.73 (55.70)	3.80 (3.78)	16.25 (16.21)
Vaii	-Cl	-4-OCH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>	C <sub>17</sub> H <sub>15</sub> ClN <sub>4</sub> O <sub>2</sub> S	374	196-203	54	54.47 (54.42)	4.03 (4.01)	14.95 (14.92)
Vaiii	-Cl	-4-N(CH <sub>3</sub> ) <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	C <sub>18</sub> H <sub>18</sub> ClN <sub>5</sub> OS	387	162-165	60	55.74 (55.71)	4.68 (4.62)	18.06 (18.03)
Vaiv	-Cl	-4-ClC <sub>6</sub> H <sub>4</sub>	C <sub>16</sub> H <sub>12</sub> Cl <sub>2</sub> N <sub>4</sub> OS	379	164-169	68	50.67 (50.64)	3.19 (3.16)	14.77 (14.73)
Vav	-Cl	-3,4-diOCH <sub>3</sub> C <sub>6</sub> H <sub>3</sub>	C <sub>18</sub> H <sub>17</sub> ClN <sub>4</sub> O <sub>3</sub> S	404	158-163	45	53.40 (53.38)	4.23 (4.20)	13.84 (13.81)
Vavi	-Cl	-2-OH C <sub>6</sub> H <sub>4</sub>	C <sub>16</sub> H <sub>13</sub> ClN <sub>4</sub> O <sub>2</sub> S	360	172-175	78	53.26 (53.24)	3.63 (3.61)	15.53 (15.52)
Vavii	-Cl	-3-NO <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	C <sub>16</sub> H <sub>12</sub> ClN <sub>5</sub> O <sub>3</sub> S	389	150-155	84	49.30 (49.29)	3.10 (3.07)	17.97 (17.98)
Vaviii	-Cl	-CH=CH-C <sub>6</sub> H <sub>5</sub>	C <sub>18</sub> H <sub>15</sub> ClN <sub>4</sub> OS	370	174-178	90	58.30 (58.27)	4.08 (4.05)	15.11 (15.08)
Vbi	-F	-C <sub>6</sub> H <sub>5</sub>	C <sub>16</sub> H <sub>13</sub> FN <sub>4</sub> OS	328	148-153	75	58.52 (58.49)	3.99 (3.93)	17.06 (17.02)
Vbii	-F	-4-OCH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>	C <sub>17</sub> H <sub>15</sub> F N <sub>4</sub> O <sub>2</sub> S	358	182-185	56	56.97 (56.94)	4.22 (4.19)	15.63 (15.61)
Vbiii	-F	-4-N(CH <sub>3</sub> ) <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	C <sub>18</sub> H <sub>18</sub> FN <sub>5</sub> OS	371	169-172	68	58.21 (58.19)	4.88 (4.86)	18.86 (18.82)
Vbiv	-F	-4-ClC <sub>6</sub> H <sub>4</sub>	C <sub>16</sub> H <sub>12</sub> ClFN <sub>4</sub> OS	362	154-159	65	52.97 (52.91)	3.33 (3.31)	15.44 (15.41)
Vbv	-F	-3,4-diOCH <sub>3</sub> C <sub>6</sub> H <sub>3</sub>	C <sub>18</sub> H <sub>17</sub> FN <sub>4</sub> O <sub>3</sub> S	388	180-183	55	55.66 (55.62)	4.41 (4.39)	14.42 (14.40)
Vbvi	-F	-2-OH C <sub>6</sub> H <sub>4</sub>	C <sub>16</sub> H <sub>13</sub> FN <sub>4</sub> O <sub>2</sub> S	344	168-172	78	55.80 (55.79)	3.81 (3.78)	16.27 (16.25)
Vbvii	-F	-3-NO <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	C <sub>16</sub> H <sub>12</sub> FN <sub>5</sub> O <sub>3</sub> S	373	166-169	72	51.47 (51.42)	3.24 (3.21)	18.76 (18.71)
Vbviii	-F	-CH=CH-C <sub>6</sub> H <sub>5</sub>	C <sub>18</sub> H <sub>15</sub> FN <sub>4</sub> OS	354	182-185	60	61.00 (60.98)	4.27 (4.22)	15.81 (15.79)
Vci	-CH <sub>3</sub>	-C <sub>6</sub> H <sub>5</sub>	C <sub>17</sub> H <sub>16</sub> N <sub>4</sub> OS	324	296-302	54	62.94 (62.91)	4.97 (4.94)	17.27 (17.25)
Vcii	-CH <sub>3</sub>	-4-OCH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>	C <sub>18</sub> H <sub>18</sub> N <sub>4</sub> O <sub>2</sub> S	354	290-293	40	61.00 (60.59)	5.12 (5.10)	15.81 (15.78)
Vciiii	-CH <sub>3</sub>	-4-N(CH <sub>3</sub> ) <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	C <sub>19</sub> H <sub>21</sub> N <sub>5</sub> OS	367	288-292	44	62.10 (62.07)	5.76 (5.73)	19.06 (19.03)
Vciv	-CH <sub>3</sub>	-4-ClC <sub>6</sub> H <sub>4</sub>	C <sub>17</sub> H <sub>15</sub> ClN <sub>4</sub> OS	358	294-299	48	56.90 (56.87)	4.20 (4.17)	15.61 (15.57)
Vcv	-CH <sub>3</sub>	-3,4-diOCH <sub>3</sub> C <sub>6</sub> H <sub>3</sub>	C <sub>19</sub> H <sub>20</sub> N <sub>4</sub> O <sub>3</sub> S	384	268-270	30	59.36 (59.33)	5.24 (5.21)	14.57 (14.53)
Vcvi	-CH <sub>3</sub>	-2-OH C <sub>6</sub> H <sub>4</sub>	C <sub>17</sub> H <sub>16</sub> N <sub>4</sub> O <sub>2</sub> S	340	298-302	50	59.98 (59.94)	4.74 (4.71)	16.46 (16.42)

Vcvii	-CH <sub>3</sub>	-3-NO <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	C <sub>17</sub> H <sub>15</sub> N <sub>5</sub> O <sub>3</sub> S	369	270-274	55	55.27 (55.21)	4.09 (4.06)	18.96 (18.92)
Vcviii	-CH <sub>3</sub>	-CH=CH-C <sub>6</sub> H <sub>5</sub>	C <sub>19</sub> H <sub>18</sub> N <sub>4</sub> O <sub>5</sub> S	350	272-275	52	65.12 (65.08)	5.18 (5.14)	15.99 (15.94)
Vdi	-OCH <sub>3</sub>	-C <sub>6</sub> H <sub>5</sub>	C <sub>17</sub> H <sub>16</sub> N <sub>4</sub> O <sub>2</sub> S	340	150-155	70	59.98 (59.95)	4.74 (4.71)	16.46 (16.42)
Vdii	-OCH <sub>3</sub>	-4-OCH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>	C <sub>18</sub> H <sub>18</sub> N <sub>4</sub> O <sub>3</sub> S	370	194-197	57	58.36 (58.32)	4.90 (4.84)	15.12 (15.08)
Vdiii	-OCH <sub>3</sub>	-4-N(CH <sub>3</sub> ) <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	C <sub>19</sub> H <sub>21</sub> N <sub>5</sub> O <sub>2</sub> S	383	140-145	54	59.51 (59.49)	5.52 (5.49)	18.26 (18.21)
Vdiv	-OCH <sub>3</sub>	-4-ClC <sub>6</sub> H <sub>4</sub>	C <sub>17</sub> H <sub>15</sub> ClN <sub>4</sub> O <sub>2</sub> S	374	178-182	68	54.47 (54.42)	4.03 (3.99)	14.95 (14.91)
Vdv	-OCH <sub>3</sub>	-3,4-diOCH <sub>3</sub> C <sub>6</sub> H <sub>3</sub>	C <sub>19</sub> H <sub>20</sub> N <sub>4</sub> O <sub>4</sub> S	400	160-164	56	56.99 (56.95)	5.03 (5.00)	13.99 (13.94)
Vdvi	-OCH <sub>3</sub>	-2-OH C <sub>6</sub> H <sub>4</sub>	C <sub>17</sub> H <sub>16</sub> N <sub>4</sub> O <sub>3</sub> S	356	172-175	66	57.29 (57.25)	4.52 (4.49)	15.72 (15.69)
Vdvii	-OCH <sub>3</sub>	-3-NO <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	C <sub>17</sub> H <sub>15</sub> N <sub>5</sub> O <sub>4</sub> S	385	168-173	74	52.98 (52.94)	3.92 (3.87)	18.17 (18.12)
Vdviii	-OCH <sub>3</sub>	-CH=CH-C <sub>6</sub> H <sub>5</sub>	C <sub>19</sub> H <sub>18</sub> N <sub>4</sub> O <sub>2</sub> S	366	190-194	65	62.28 (62.22)	4.95 (4.91)	15.29 (15.23)

Table 5: Antibacterial activity of 6-Substituted -2-(benzo(d)thiazol-2-ylamino)-N-arylidene acetohydrazides

Code	R	Ar	Gram +ve		Gram -ve	
			<i>S. aureus</i>	<i>L. delbrueckii</i>	<i>P. vulgaris</i>	<i>E. coli</i>
Ciprofloxacin (10µg/cup)	-	-	20	15	16	18
Vai	-Cl	-C <sub>6</sub> H <sub>5</sub>	3	4	3	5
Vaii	-Cl	-4OCH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>	5	3	11	4
Vaiii	-Cl	-4-N(CH <sub>3</sub> ) <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	5	4	3	2
Vaiv	-Cl	-4-ClC <sub>6</sub> H <sub>4</sub>	4	3	2	2
Vav	-Cl	-3,4-diOCH <sub>3</sub> C <sub>6</sub> H <sub>3</sub>	10	8	8	8
Vavi	-Cl	-2-OH C <sub>6</sub> H <sub>4</sub>	12	6	5	4
Vavii	-Cl	-3-NO <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	5	4	6	3
Vaviii	-Cl	-CH=CH-C <sub>6</sub> H <sub>5</sub>	10	5	5	3
Vbi	-F	-C <sub>6</sub> H <sub>5</sub>	-	-	3	2
Vbii	-F	-4-OCH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>	7	6	5	4
Vbiii	-F	-4-N(CH <sub>3</sub> ) <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	6	5	10	3
Vbiv	-F	-4-ClC <sub>6</sub> H <sub>4</sub>	9	7	6	2
Vbv	-F	-3,4-diOCH <sub>3</sub> C <sub>6</sub> H <sub>3</sub>	-	-	2	4
Vbvi	-F	-2-OH C <sub>6</sub> H <sub>4</sub>	7	6	6	5
Vbvii	-F	-3-NO <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	12	9	9	10
Vbviii	-F	-CH=CH-C <sub>6</sub> H <sub>5</sub>	6	7	6	5
Vci	-CH <sub>3</sub>	-C <sub>6</sub> H <sub>5</sub>	5	7	3	4
Vcii	-CH <sub>3</sub>	-4-OCH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>	7	8	6	7
Vciii	-CH <sub>3</sub>	-4-N(CH <sub>3</sub> ) <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	5	3	6	5
Vciv	-CH <sub>3</sub>	-4-ClC <sub>6</sub> H <sub>4</sub>	8	7	7	9
Vcv	-CH <sub>3</sub>	-3,4-diOCH <sub>3</sub> C <sub>6</sub> H <sub>3</sub>	11	8	6	7
Vcvi	-CH <sub>3</sub>	-2-OH C <sub>6</sub> H <sub>4</sub>	9	8	7	7
Vcvii	-CH <sub>3</sub>	-3-NO <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	7	6	5	4
Vcviii	-CH <sub>3</sub>	-CH=CH-C <sub>6</sub> H <sub>5</sub>	-	-	4	3
Vdi	-OCH <sub>3</sub>	-C <sub>6</sub> H <sub>5</sub>	8	7	8	6
Vdii	-OCH <sub>3</sub>	-4-OCH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>	6	8	7	6
Vdiii	-OCH <sub>3</sub>	-4-N(CH <sub>3</sub> ) <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	9	8	9	7
Vdiv	-OCH <sub>3</sub>	-4-ClC <sub>6</sub> H <sub>4</sub>	9	5	6	4
Vdv	-OCH <sub>3</sub>	-3,4-diOCH <sub>3</sub> C <sub>6</sub> H <sub>3</sub>	3	4	5	4
Vdvi	-OCH <sub>3</sub>	-2-OH C <sub>6</sub> H <sub>4</sub>	12	10	10	11
Vdvii	-OCH <sub>3</sub>	-3-NO <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	4	3	2	4
Vdviii	-OCH <sub>3</sub>	-CH=CH-C <sub>6</sub> H <sub>5</sub>	7	9	9	2

Table 6: Antifungal activity of Antibacterial activity of 6-Substituted -2-(benzo(d)thiazol-2-ylamino)-N-arylidene acetohydrazides

Compound	R	Ar	<i>Aspergillus niger</i>	<i>Candida albicans</i>
Clotrimazole (10µg/cup)	-	-	18	22
Vai	-Cl	-C <sub>6</sub> H <sub>5</sub>	3	2
Vaii	-Cl	-4OCH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>	6	2
Vaiii	-Cl	-4-N(CH <sub>3</sub> ) <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	3	4
Vaiv	-Cl	-4-ClC <sub>6</sub> H <sub>4</sub>	8	6
Vav	-Cl	-3,4-diOCH <sub>3</sub> C <sub>6</sub> H <sub>3</sub>	4	2
Vavi	-Cl	-2-OH C <sub>6</sub> H <sub>4</sub>	-	2
Vavii	-Cl	-3-NO <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	2	-
Vaviii	-Cl	-CH=CH-C <sub>6</sub> H <sub>5</sub>	5	3
Vbi	-F	-C <sub>6</sub> H <sub>5</sub>	4	1
Vbii	-F	-4-OCH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>	4	4
Vbiii	-F	-4-N(CH <sub>3</sub> ) <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	3	2
Vbiv	-F	-4-ClC <sub>6</sub> H <sub>4</sub>	2	-
Vbv	-F	-3,4-diOCH <sub>3</sub> C <sub>6</sub> H <sub>3</sub>	5	1

4bvi	-F	-2-OH C <sub>6</sub> H <sub>4</sub>	3	2
Vbvii	-F	-3-NO <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	7	4
Vbviii	-F	-CH=CH-C <sub>6</sub> H <sub>5</sub>	9	8
Vci	-CH <sub>3</sub>	-C <sub>6</sub> H <sub>5</sub>	3	3
Vcii	-CH <sub>3</sub>	-4-OCH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>	5	3
Vciii	-CH <sub>3</sub>	-4-N(CH <sub>3</sub> ) <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	3	-
Vciv	-CH <sub>3</sub>	-4-ClC <sub>6</sub> H <sub>4</sub>	4	6
Vcv	-CH <sub>3</sub>	-3,4-diOCH <sub>3</sub> C <sub>6</sub> H <sub>3</sub>	6	2
Vcvi	-CH <sub>3</sub>	-2-OH C <sub>6</sub> H <sub>4</sub>	7	5
Vcvii	-CH <sub>3</sub>	-3-NO <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	4	3
Vcviii	-CH <sub>3</sub>	-CH=CH-C <sub>6</sub> H <sub>5</sub>	3	2
Vdi	-OCH <sub>3</sub>	-C <sub>6</sub> H <sub>5</sub>	4	2
Vdii	-OCH <sub>3</sub>	-4-OCH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>	6	8
Vdiii	-OCH <sub>3</sub>	-4-N(CH <sub>3</sub> ) <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	4	2
Vdiv	-OCH <sub>3</sub>	-4-ClC <sub>6</sub> H <sub>4</sub>	8	5
Vdv	-OCH <sub>3</sub>	-3,4-diOCH <sub>3</sub> C <sub>6</sub> H <sub>3</sub>	3	2
Vdvi	-OCH <sub>3</sub>	-2-OH C <sub>6</sub> H <sub>4</sub>	7	6
Vdvii	-OCH <sub>3</sub>	-3-NO <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	1	3
Vdviii	-OCH <sub>3</sub>	-CH=CH-C <sub>6</sub> H <sub>5</sub>	4	2

Table 7: Antioxidant activity (% Inhibition, IC<sub>50</sub> Values) of 6-Substituted -2-(benzo(d)thiazol-2-ylamino)-N-arylidene acetohydrazides

CODE	R	Ar	% Inhibition					IC <sub>50</sub> (mg/ml)	IC <sub>50</sub> (Mm)	IC <sub>50</sub> (nm)
			20	40	60	80	100			
V ai	-Cl	-C <sub>6</sub> H <sub>5</sub>	15.87	17.46	20.63	22.22	25.40	167.79	0.67	670
V aii	-Cl	-4OCH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>	23.81	26.63	30.16	31.75	34.92	117.10	0.44	440
V aiii	-Cl	-4-N(CH <sub>3</sub> ) <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	47.62	49.21	52.38	55.56	57.14	67.84	0.23	226
V aiv	-Cl	-4-ClC <sub>6</sub> H <sub>4</sub>	28.57	31.75	34.92	38.10	39.68	100.60	0.35	353
V av	-Cl	-3,4-diOCH <sub>3</sub> C <sub>6</sub> H <sub>3</sub>	34.92	38.10	39.68	42.86	44.44	88.18	0.25	249
V avi	-Cl	-2-OH C <sub>6</sub> H <sub>4</sub>	57.14	60.32	63.49	66.67	69.84	55.93	0.15	151
V avii	-Cl	-3-NO <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	47.62	50.79	53.97	55.56	57.14	67.84	0.23	226
V aviii	-Cl	-CH=CH-C <sub>6</sub> H <sub>5</sub>	31.75	33.33	36.51	38.10	41.27	97.09	0.34	342
V bi	-F	-C <sub>6</sub> H <sub>5</sub>	50.79	53.97	55.56	58.73	60.32	63.86	0.21	211
V bii	-F	-4-OCH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>	55.56	58.73	60.32	63.49	66.67	58.48	0.18	184
V biii	-F	-4-N(CH <sub>3</sub> ) <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	44.44	47.62	50.79	53.97	55.56	70.03	0.24	246
V biv	-F	-4-ClC <sub>6</sub> H <sub>4</sub>	53.97	57.14	60.32	61.90	63.49	60.17	0.20	201
V bv	-F	-3,4-diOCH <sub>3</sub> C <sub>6</sub> H <sub>3</sub>	41.27	42.86	46.03	49.21	50.79	76.92	0.23	228
V bvi	-F	-2-OH C <sub>6</sub> H <sub>4</sub>	61.90	65.08	66.67	69.84	73.08	53.08	0.15	150
V bvii	-F	-3-NO <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	22.22	25.40	26.98	30.16	31.75	127.23	0.45	454
V bviii	-F	-CH=CH-C <sub>6</sub> H <sub>5</sub>	38.10	41.27	42.86	47.62	50.79	79.37	0.27	268
V ci	-CH <sub>3</sub>	-C <sub>6</sub> H <sub>5</sub>	49.21	50.79	53.97	57.14	58.73	65.88	0.21	210
V cii	-CH <sub>3</sub>	-4-OCH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>	53.97	55.56	58.73	60.32	61.90	61.65	0.19	187
V ciii	-CH <sub>3</sub>	-4-N(CH <sub>3</sub> ) <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	11.11	14.81	22.22	29.63	33.33	14.45	0.56	561
V civ	-CH <sub>3</sub>	-4-ClC <sub>6</sub> H <sub>4</sub>	29.63	37.04	40.74	44.44	55.85	83.06	0.27	270
V cv	-CH <sub>3</sub>	-3,4-diOCH <sub>3</sub> C <sub>6</sub> H <sub>3</sub>	22.22	25.93	33.33	37.04	40.74	104.60	0.39	390
V cvi	-CH <sub>3</sub>	-2-OH C <sub>6</sub> H <sub>4</sub>	18.52	22.22	29.63	37.01	39.07	109.41	0.38	380
V cvii	-CH <sub>3</sub>	-3-NO <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	37.04	40.74	48.15	55.56	59.26	70.42	0.19	192
V cviii	-CH <sub>3</sub>	-CH=CH-C <sub>6</sub> H <sub>5</sub>	44.44	51.85	55.56	59.26	66.67	62.19	0.16	161
V di	-OCH <sub>3</sub>	-C <sub>6</sub> H <sub>5</sub>	33.33	40.74	44.44	51.85	59.26	73.21	0.27	273
V dii	-OCH <sub>3</sub>	-4-OCH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>	51.85	59.26	66.67	70.37	77.78	52.85	0.18	184
V diii	-OCH <sub>3</sub>	-4-N(CH <sub>3</sub> ) <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	59.26	62.96	66.67	74.07	75.09	51.39	0.17	170
V div	-OCH <sub>3</sub>	-4-ClC <sub>6</sub> H <sub>4</sub>	62.96	66.67	77.78	81.48	85.19	46.55	0.14	145
V dv	-OCH <sub>3</sub>	-3,4-diOCH <sub>3</sub> C <sub>6</sub> H <sub>3</sub>	29.63	37.04	44.44	48.15	51.85	79.87	0.28	282
V dvi	-OCH <sub>3</sub>	-2-OH C <sub>6</sub> H <sub>4</sub>	40.74	48.15	55.26	66.67	62.97	59.26	0.20	202
V dvii	-OCH <sub>3</sub>	-3-NO <sub>2</sub> C <sub>6</sub> H <sub>4</sub>	59.26	70.34	77.78	81.48	85.19	46.43	0.13	135
V dviii	-OCH <sub>3</sub>	-CH=CH-C <sub>6</sub> H <sub>5</sub>	60.67	74.07	77.78	85.19	88.89	44.60	0.12	125

Table 8: Anticancer activity of Some Novel Synthesized compounds

Compound	R	Ar	A549	C-205	A431	PC-3
V <sub>ai</sub>	OCH <sub>3</sub>	-C <sub>6</sub> H <sub>5</sub>	39.8±2.3	24.7±2.8	33.3±2.5	22.3±2.2
V <sub>aiii</sub>	OCH <sub>3</sub>	-4OCH <sub>3</sub> C <sub>6</sub> H <sub>4</sub>	115.2±2.5	34.8±2.3	46.7±2.6	21.3±3.2
V <sub>avi</sub>	OCH <sub>3</sub>	-2-OHC <sub>6</sub> H <sub>4</sub>	172.28±3.2	127.9±3.3	100.3±3.5	100.0±2.4

RESULTS AND DISCUSSION

The antibacterial activity data of 6-Substituted -2-(benzo(d)thiazol-2-ylamino)-N-arylidene acetohydrazides given in Table 5 indicates that two of the present compounds are inactive against *S.aureus* & *L.delbrueckii* whereas compounds V<sub>dvi</sub> (R=OCH<sub>3</sub> & Ar= C<sub>6</sub>H<sub>4</sub>OH-2), V<sub>bvii</sub> (R=F & C<sub>6</sub>H<sub>4</sub>NO<sub>2</sub>-3), are found to be relatively more effective against both gram +ve and gram -ve organisms with the zones of inhibition (mm); 12,10,10,11 and 12,9,9,10 respectively. It is also noticed from the data that the compounds V<sub>av</sub> (R=C1 & Ar=C<sub>6</sub>H<sub>3</sub>(OCH<sub>3</sub>)<sub>2</sub>-3,4),

V<sub>diii</sub> (R=OCH<sub>3</sub>, Ar=C<sub>6</sub>H<sub>4</sub>N(CH<sub>3</sub>)<sub>2</sub>-4) and V<sub>cvi</sub> (R=CH<sub>3</sub>, Ar(C<sub>6</sub>H<sub>4</sub>OH-2)) are next in the order of antibacterial activity. The test compounds V<sub>biii</sub> (R=F, Ar=C<sub>6</sub>H<sub>4</sub>N(CH<sub>3</sub>)<sub>2</sub>-4), V<sub>dviii</sub> (R=OCH<sub>3</sub>, Ar=C<sub>6</sub>H<sub>5</sub>-CH=CH) are selectively more active against *P.vulgaris*, with zones of inhibition: 11, 10 & 9mm, whereas compound V<sub>avi</sub> (R=C1, Ar=C<sub>6</sub>H<sub>4</sub>OH-2), V<sub>bvii</sub> (R=F, Ar=C<sub>6</sub>H<sub>4</sub>NO<sub>2</sub>-3), V<sub>dvi</sub> (R=OCH<sub>3</sub>, Ar=C<sub>6</sub>H<sub>4</sub>OH-2), V<sub>cvi</sub> (R=CH<sub>3</sub>, Ar=C<sub>6</sub>H<sub>3</sub> dioCH<sub>3</sub>-3,4), V<sub>av</sub> (R=C1, Ar=C<sub>6</sub>H<sub>3</sub>.dioCH<sub>3</sub>-3,4), V<sub>avii</sub> (R=C1, Ar=C<sub>6</sub>H<sub>5</sub>.CH=CH), are found to possess higher inhibitory activity against *S.aureus*, with zones of inhibition : 12, 12, 12, 11, 10 and 10 mm,

respectively. The rest of the compounds showed mild to moderate antibacterial activity.

From the above antibacterial evaluation for the Series-I compounds,  $V_{bvii}$  and  $V_{dvii}$  were found to be more active in the Series and as well as less active compared to the standard Ciprofloxacin.

The antifungal data of new 6-Substituted -2-(benzo(d)thiazol-2ylamino)-N-arylidene acetohydrazides given in Table 6 indicates that some of the compounds of this series could show the antifungal activity against both the strains of the fungi employed with a degree of variation. Among all the test compounds, compound  $V_{bviii}$  (R=F, Ar=C<sub>6</sub>H<sub>5</sub>, CH=CH) is found to be relatively more effective against *A.niger*. Compounds  $V_{aiv}$  (R=C1, Ar=C<sub>6</sub>H<sub>4</sub>.C1-4),  $V_{div}$  (R=OCH<sub>3</sub>, Ar=C<sub>6</sub>H<sub>4</sub>.C1-4) are found to be next in the order of inhibitory activity. Compound  $V_{avi}$  (R=C1, Ar=C<sub>6</sub>H<sub>4</sub>.OH-2) is found to be in active against *A.niger*. Compounds  $V_{bviii}$  (R=F, Ar=C<sub>6</sub>H<sub>5</sub>, CH=CH) and  $V_{dii}$  (R=OCH<sub>3</sub>, Ar=C<sub>6</sub>H<sub>4</sub>.OCH<sub>3</sub>-4) are equi & more potent against *C.albicans* with zone of inhibition 8 mm. Only three compounds  $V_{avii}$  (R=C1, Ar=C<sub>6</sub>H<sub>4</sub>.NO<sub>2</sub>-3),  $V_{biv}$  (R=F, Ar=C<sub>6</sub>H<sub>4</sub>.C1-4) and  $V_{ciii}$  (R=CH<sub>3</sub>, Ar=C<sub>6</sub>H<sub>4</sub>.N(CH<sub>3</sub>)<sub>2</sub>-4) are inactive against *C.albicans*. Remaining all the compounds shown mild to moderate antifungal activity.

The antioxidant data of compounds given in Table 7 reveals that the six compounds such as  $V_{dviii}$  (R=OCH<sub>3</sub>, Ar=C<sub>6</sub>H<sub>5</sub>-CH=CH-),  $V_{div}$  (R=OCH<sub>3</sub>, Ar=C<sub>6</sub>H<sub>4</sub>.NO<sub>2</sub>-3),  $V_{dii}$  (R=OCH<sub>3</sub>, Ar=C<sub>6</sub>H<sub>4</sub>.C1-4),  $V_{bvi}$  (R=F, Ar=C<sub>6</sub>H<sub>4</sub>.OH-2),  $V_{avi}$  (R=C1, Ar=C<sub>6</sub>H<sub>4</sub>.OH-2), and  $V_{cviii}$  (R=C1, Ar=C<sub>6</sub>H<sub>5</sub>.CH=CH) showed more potent antioxidant activity with IC<sub>50</sub> values: 125, 135, 145, 150, 151 and 161 nm respectively, When compared with standard (IC<sub>50</sub> value 270 nm). Compounds  $V_{ai}$  (R=C1, Ar=C<sub>6</sub>H<sub>5</sub>),  $V_{aii}$  (R=C1, Ar=C<sub>6</sub>H<sub>4</sub>.OCH<sub>3</sub>-4),  $V_{aiv}$  (R=C1, Ar=C<sub>6</sub>H<sub>4</sub>.C1-4),  $V_{aviii}$  (R=C1, Ar=C<sub>6</sub>H<sub>5</sub>.CH=CH),  $V_{bvii}$  (R=F, Ar=C<sub>6</sub>H<sub>4</sub>.NO<sub>2</sub>-3),  $V_{ciii}$  (R=CH<sub>3</sub>, Ar=C<sub>6</sub>H<sub>4</sub>.N(CH<sub>3</sub>)<sub>2</sub>-4),  $V_{cv}$  (R=CH<sub>3</sub>, Ar=C<sub>6</sub>H<sub>3</sub>.diOCH<sub>3</sub>-3,4),  $V_{cvi}$  (R=CH<sub>3</sub>, Ar=C<sub>6</sub>H<sub>4</sub>.OH-2),  $V_{dv}$  (R=OCH<sub>3</sub>, Ar=C<sub>6</sub>H<sub>3</sub>.diOCH<sub>3</sub>-3,4), are less active than standard (BHT). The remaining compounds showed potent to moderate potent antioxidant activity with IC<sub>50</sub> values ranging from 170-670 nm.

The anticancer activity of some novel benzthiazoles as presented in Table 8 reveals that only compound  $V_{di}$  is shown to have mild anticancer activity compared with that of standard and rest of the compounds have not shown any significant anticancer activity.

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