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Synthesis of 1,3-Oxazepine Derivatives Derived from 2-(1H-Benzo[d][1,2,3]Triazol-1-yl) Acetohydrazide by Using Microwave Irradiation

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Abstract

A series of Schiff base and their derivative (oxazepine) have been synthesized. 8-hydrazone(schiff-bases) derivatives type (E)-2-(1H-benzo[d][1,2,3]triazole-1-yl)-N-(substituted benzylidene)acetohydrazide were prepared by condensation 1H-benzo[d][1,2,3]triazol-1-yl)hydrazine with various aromatic aldehyde in ethanol in the presence of dimethyl formamid or acetic acid as catalyst by using MWI to yield the Schiff bases. These Schiff's base on treatment with Maleic anhydride in dry conditions by using MWI to give 7-membered heterocyclic ring system (oxezapine) of (2-(1H)benzo[d][1,2,3]triazol-1-yl)-N-(2-(substituted penyl)-4, 7-dioxo-4,7-dihydro-1,3-oxazepin-3-(2H)-yl)acetamide. The purity of the compounds was confirmed by TLC. The final products were identified by their melting point, IR, 1HNMR, and UV-visible spectra.

Keywords

Hydrazone, Microwave, Oxezapine, Benzotriazole, Hydrazine

1. Introduction

The Schiff bases (or hydrazones) are considered to be precusor of (oxazepine) and other heterocyclic rings. Oxazepine, refers to any seven-membered ring containing oxygen in position one and nitrogen in position three in addition to the five carbon atoms. The 1,3-oxazepine is a branch of many types of heterocyclic oxazepine [1]-[6]. The core structure is 1,3-oxazepine-4,7-diones of seven-membered ring along with two carbonyl group. Over the years, the synthesis of oxazepine has been investigated and documented. It is prepared by the pericyc-

liccyclo addition of Schiff base or hydrazone with maleic, phthalic and succinic anhydrides [7] [8] [9] [10] [11] and also by green chemistry method [12] [13]. Oxazepine derivatives were found to exhibit a vast variety of biological activities like antibacterial [14], antifungal [15], hypnotic muscle relaxant [16], antagonistic [17], inflammatory [18] and antiepileptic [19]. Microwave-assisted reactions within shorter time are becoming popular for organic chemists [20] [21] [22] and had recently been reviewed [23] [24]. More interest has been focused on dry media synthesis under microwave irradiation and especially by carrying out the experiments with supported reagents on mineral oxide [25] [26]. This technology provides a promising alternative to environmentally unacceptable thermol procedures, which are usually time consuming, unsafe and cause solvent emission leading to pollution and wastedisposal problems. Under the framework of green chemistry an environmentally benign solvent-free approach has been developed for the synthesis of substituted hydrazide, substituted hydrazones and oxazepine by using microwave-assisted dry media reaction conditions.

2. Experimental

Melting pionts were determined in open capillary tubes and are uncorrected by using Stuart Melting Point Apparatus. The IR spectra (cm^{-1}) were recorded on Schimadzu FT-IR-8400S by using KBr disc.^{1}HMNR spectra (DMSO-d_6) were recorded on ultra shield 300 MHz Bruker (2003) NMR spectrometer using TMS as internal standard. Follow up of the reactions and the purity of the compounds by using TLC-techniqe on aluminium plates percoated with silica gel in various solvent system using iodine vapours as detecting agent. Reactions were carried out in domestic microwave oven (Bomann 02227 CB 700W). All the chemicals and solvents used were of laboratory grad.

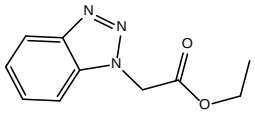
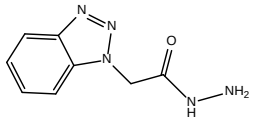
2.1. Preparation of Ethyl Benzotriazole Acetate(1a)

(0.03 mol) of benzotriazole was mixed with (0.03 mol) of ethyl –α-chloro acetate and (9.0 gm) of potassium carbonate dry in (70 mL) of acetone for 24 hr. After completation of the reaction the solvent was evaporated, the product was extracted by using diethyl ether. Evaporating the organic solvent (diethyl ether) gave soild needle crystals, its physical data illustrated in **Table 1**, yield (60-90%), M.p. (60˚C - 61˚C).

2.2. General Procedure for Microwave Assisted Preparation of: 2-(1H-Benzo[d][1,2,3]Triazol-1-yl)Acetohydrazide (2a)

(0.01 mol) of ester(1a)was mixed with (0.01 mol) hydrazine hydrate (80% conc.) in a 50 mL beaker, then was 2 mL of methanol added to the mixture. The mixture was exposed to microwave irradiation (80 W) for about 3 min. The progress of the reaction and the purity of the compounds were monitored with (TLC). The reaction mixture was cooled at 4˚C - 5˚C. The separated soild crystals were filtered and washed with cold ethanol. The crystals were dried and recrystallized from ethanol its physical data illustrated in **Table 1**.

Table 1. Physical data for starting compounds.

Comp. No.	Structure	Name	M.P. ˚C	Yield %	MWI	Reaction time	Colour
1a		ethyl 2-(1H-benzo[d][1,2,3]triazol-1-yl)acetate	60 - 61	73	-	24 hr	white
2a		2-(1H-benzo[d][1,2,3]triazol-1-yl)acetohydrazide	172 - 173	95	80 W	3 min	grayish

2.3. General Procedure for Microwave-Assisted Preparation of: (E)-2-(1H-Benzo[d][1,2,3]Triazol-1-yl)-N'-(Substituted Benzylidene)Acetohydrazide(3b,4b,5b,6b,8b,9b)

(0.01 mol) of hydrazide(2) mixed with (0.01 mol) substituted aromatic aldehyde (2-hydroxy,3-hydroxy, 3-chloro,4-dimethylamino, 2-bromo, 4-bromo)in beaker size 50 mL, 3 - 4 drops of dimethyl formaimde was added as catalyst. The mixture was exposed to microwave irradiation at different power and time interval (as showed in the Table 2). After completion of the reaction as indicated by TLC, the reaction mixture was cooled at room temperature and washed with mixed solvent (9:1) (ether: ethyl acetate). The products were recrystallized absolute ethanol to give yield (60% - 90%) pure crystal of substituted hydrazones (3, 4, 5, 6, 8, 9).

The same procedure to prepare **(7b,10b)** was followed using (0.01 mol) furfuraldehyde and (0.01 mol) cinnimaldehyde. its physical data illustrated in Table 2).

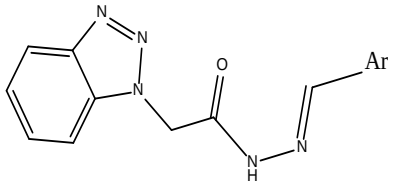
2.4. General Procedure for Microwave-Assisted Preparation of: 2-(1H-Benzo[d][1,2,3]Triazol-1-yl)-N-(2-(Substituted Phenyl)-4,7-Dioxo-4,7-Dihydro-1,3-Oxazepin-3(2H)-yl) Acetamide(11c,12c,13c,14c,15c,16c,17c,18c)

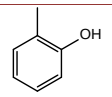
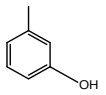
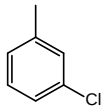
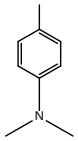
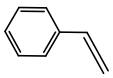
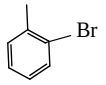
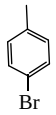
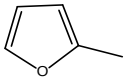
(0.001 mol) of substituted hyrazone mixed with (0.001 mol) of maleic anhydride in dry poceline mortar, to obaine fine mixed powder. The dry powder was irradiated in a microwave oven at different power and time of irradiation(showed in the Table 3) in 50 mL open small beaker. After completion of the reaction as indicated by TLC, the reaction mixture was cooled at room temperature. The product was washed with benzene and recrystallizated by dioxane to give good yield (60% - 90%) of pure crystal oxazepine derivatives (11 - 18). its physical data illustrated in Table 3.

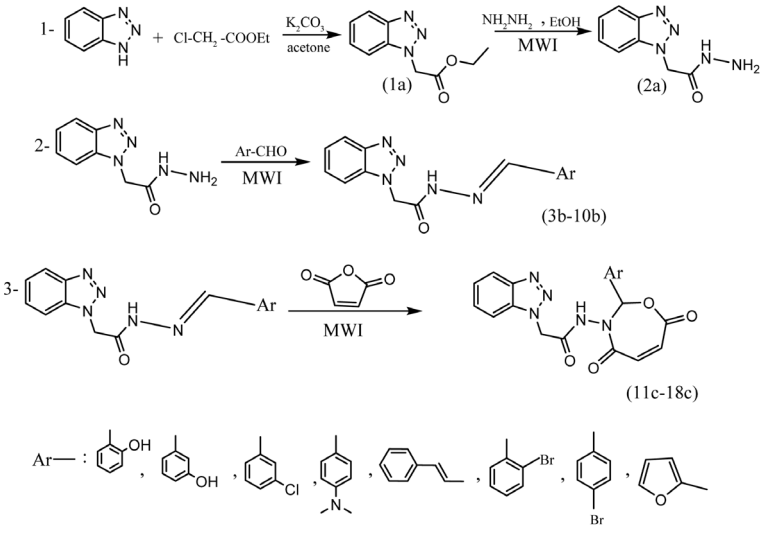
3. Result and Discussion

The Schiff base or (hydrazone) compounds [3b-10b] were synthesized from the reaction of benzotriazole acetohydrazide with different substituted aldehydes (Scheme 1). The synthesis of these compounds was carried out according to the steps outlined in scheme, by using microwave irradiation, and the physical properties are given in Table 2. Table 4 describe the important vibrational

Table 2. Physical data for hydrazone derivatives.

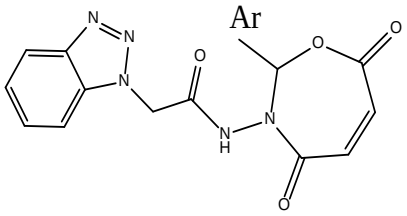


Comp. No.	Ar	Name	m.p. ˚C	Yield %	MWI Watt	Reaction Time/min	Colour
3b		(E)-2-(1H-benzo[d][1,2,3]triazol-1-yl)-N'-(2-hydroxybenzylidene) acetohydrazide	230	80	360	3	grayish
4b		(E)-2-(1H-benzo[d][1,2,3]triazol-1-yl)-N'-(3-hydroxybenzylidene) acetohydrazide	250 - 252	65	180	3	leady
5b		(E)-2-(1H-benzo[d][1,2,3]triazol-1-yl)-N'-(3-chlorobenzylidene) acetohydrazide	210	70	80	2	yellow
6b		(E)-2-(1H-benzo[d][1,2,3]triazol-1-yl)-N'-(4-dimethyl aminoben-zylidene) acetohydrazide	200	67	80	2	sepia
7b		2-(1H-benzo[d][1,2,3]triazol-1-yl)-N'-((1E,2E)-3-phenylallylidene) acetohydrazide	188 - 189	72	180	2	yellow
8b		(E)-2-(1H-benzo[d][1,2,3]triazol-1-yl)-N'-(2-bromobenzylidene) acetohydrazide	127 - 130	40	80	3	sepia
9b		E)-2-(1H-benzo[d][1,2,3]triazol-1-yl)-N'-(4-bromobenzyliden3e)acetohydrazide	245 - 247	90	80	3	white
10b		(E)-2-(1H-benzo[d][1,2,3]triazol-1-yl)-N'-(furan-2-ylmethylene) acetohydrazide	195 - 197	60	80	2	leady



Scheme 1. The steps of synthesis.

Table 3. Physical data for oxazepine derivatives.



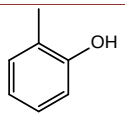
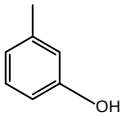
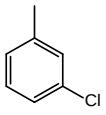
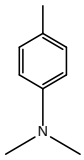
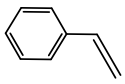
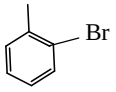
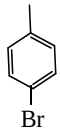
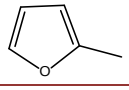
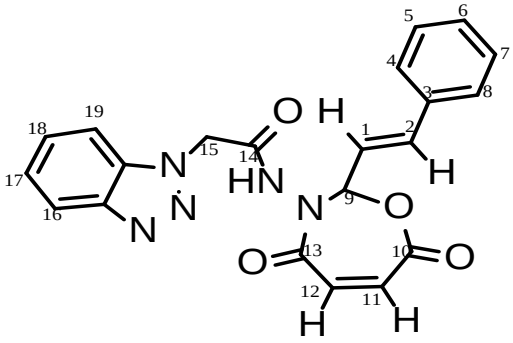
Comp. No.	Ar	Name	M.P. ˚C	Yield %	MWI Watt	Reaction Time/min	Colour
11c		2-(1H-benzo[d][1,2,3]triazol-1-yl)-N-(2-(2-hydroxyphenyl)-4,7-dioxo-4,7-dihydro-1,3-oxazepin-3(2H)-yl)acetamide	150 - 152	90	360	3	leady
12c		2-(1H-benzo[d][1,2,3]triazol-1-yl)-N-(2-(3-hydroxyphenyl)-4,7-dioxo-4,7-dihydro-1,3-oxazepin-3(2H)-yl)acetamide	87 - 90	85	180	3	black
13c		2-(1H-benzo[d][1,2,3]triazol-1-yl)-N-(2-(3-chlorophenyl)-4,7-dioxo-4,7-dihydro-1,3-oxazepin-3(2H)-yl)acetamide	140 - 142	76	180	3	gryisha
14c		2-(1H-benzo[d][1,2,3]triazol-1-yl)-N-(2-(4-dimethylaminophenyl)-4,7-dioxo-4,7-dihydro-1,3-oxazepin-3(2H)-yl)acetamide	120 - 123	78	180	2.5	red
15c		(E)-2-(1H-benzo[d][1,2,3]triazol-1-yl)-N-(4,7-dioxo-2-styryl-4,7-dihydro-1,3-oxazepin-3(2H)-yl)acetamide	112 - 115	85	180	3	yellow
16c		2-(1H-benzo[d][1,2,3]triazol-1-yl)-N-(2-(2-bromophenyl)-4,7-dioxo-4,7-dihydro-1,3-oxazepin-3(2H)-yl)acetamide	135 - 137	70	180	2	gryisha
17c		2-(1H-benzo[d][1,2,3]triazol-1-yl)-N-(2-(4-bromophenyl)-4,7-dioxo-4,7-dihydro-1,3-oxazepin-3(2H)-yl)acetamide	124 - 126	60	180	2.5	yellow
18c		2-(1H-benzo[d][1,2,3]triazol-1-yl)-N-(2-(furan-2-yl)-4,7-dioxo-4,7-dihydro-1,3-oxazepin-3(2H)-yl)acetamide	80 - 82	68	360	3.5	chestnut

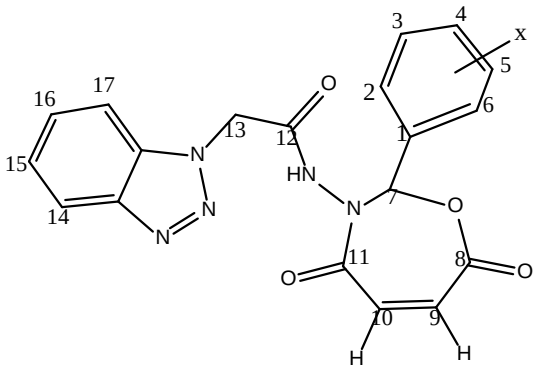
Table 4. IR and UV spectrum data for the synthesized compounds (3b-10b).

Comp. No.	*IR* v·cm^{-1} (KBr)							*UV*(EtOH) λ_{max}
	C=O *lactone*	C=O *amide*	C-N	C-O-C	C=C-C=O	Ar C=C	*Other*	
11c	1680	1610	1265	1160sy 1259as	1409	1453	3320(OH)	307
12c	1725	1632	1255	1080sy 1278as	1580	1450	3378(OH)	315
13c	1715	1612	1265	1165sy 1260as	1455	1433	746(C-Cl)	280
14c	1729	1660	1232	1065sy 1285as	1590	1446		296
15c	1720	1651	1229	1059sy 1266as	1507	1432	1622(C=C)	276
16c	1722	1646	1238	1049sy 1276as	1480	1447	624(C-Br)	284
17c	1756	1638	1242	1038sy 1266as	1496	1422	653(C-Br)	302
18c	1761	1621	1239	1122sy 1271as	1510	1445		298

modes of Schiff bases. The infrared spectra of Schiff bases exhibited the absence of absorption bands at (3345 - 3250 cm^{-1}) corresponding to stretching modes of NH_2 group of benzotriazole acetohydrazide and at (1700 cm^{-1}) C=O group of substituted benzaldehydes which refers to the formation of the Schiff bases as azomethine C=N linkage. This was confirmed by the appearance of new bands at (1580 - 1630) cm^{-1} assignable to C=N azomethine group. The table also describe the positions of the bands assigned to vibrational modes of amide NH groups at (3120 - 3230) cm^{-1} and C=O group of amide at (1660 - 1680) cm^{-1}. The reaction of Schiff bases [3b-10b] with maleic anhydride in dry condition by using microwave irradiation to give 1,3-oxazepine-4,7-dione derivatives. Cyclic addition reaction is achieved by ring formation, due to interaction between HOMO orbital of maleic anhydride with LUMO obital of (-C=N) group [27]. **Table 5** describes the important vibrational modes of oxazepine. The infrared spectra of oxazepine exhibited the absence of absorption bands at (1580 - 1630) cm^{-1} as azomethine C=N linkage and strong absorption for pure maleic anhydrid at (1800 - 1955) cm^{-1}. But the formation of oxazepine was confirmed by the presence of a new strong band at (1760 - 1680) cm^{-1} due to C=O group as lactone and C=O group as amide (lactam) at (1610 - 1660) cm^{-1}. The table also describe band assigned to vibrational modes of (C-O-C) group at (1260 - 1280) cm^{-1} as asymmetrical and the band assigned to C-N was observed at (1610 - 1660) cm^{-1} as in **Figure 1**. This confirmed the assigned seven-membered ring structure. UV spectrum of compounds in the **Table 4** and **Table 5** showed an absorption λ_{max} (270 - 320) nm which was attributed to different transitions of electrons.



The ^{1}HNMR spectrum of compund (c_{15} **Figure 2**) displayed a single peak appeared at 11.8 ppm which was assigned to chemical shift of NH and multiplet peak at 7.3 - 7.8 ppm which were assigned to chemical shifts of aromatic protons at carbons (4, 5, 6, 7, 8, 16, 17, 18, 19). The peak at 5.5 ppm was attributed to the chemical shift of carbon (15) proton its between carbonyl amide and benzotriazol moiety. The signal related to the protons at carbons (1, 2) appeared at 5.9 - 6 ppm due to conjegated with benzene ring. But protons at carbons (11, 12) in seven membered ring appeared at 6.1- 6.3 ppm, the peak at 8.1 ppm was attributed to the chemical shift of carbon (9) proton due to between two highly electronegativity atoms (oxygen and nitrogen).



The 1HNMR spectrum of the compound (C11 Figure 3) displayed a single peak appeared at 11.8 ppm which was assigned to chemical shift of NH and multiplet peak at 7.2 – 7.9 ppm which were assigned to chemical shifts of aromatic protons at carbons (2, 3, 4, 5, 6, 14, 15, 16, 17). The peak at 5.5 ppm was attri-

Table 5. IR and VUspectrum data for the synthesized compounds (11c-18c).

Comp. No.	IR V·cm^{-1} (KBr)				UV (EtOH) λ_{max}
	N-H	C=O	C=N	other	
3b	3115	1660	1607	3200 (OH)	318
4b	3120	1671	1611	3191 (OH)	320
5b	3190	1665	1599	738(C-Cl)	305
6b	3211	1681	1619	1262(C-N)	317
7b	3230	1669	1600	1619(C=C)	302
8b	3220	1676	1580	624(C-Br)	309
9b	3235	1680	1616	1110(C-Br)	310
10b	3175	1662	1632	(C-OC)	303

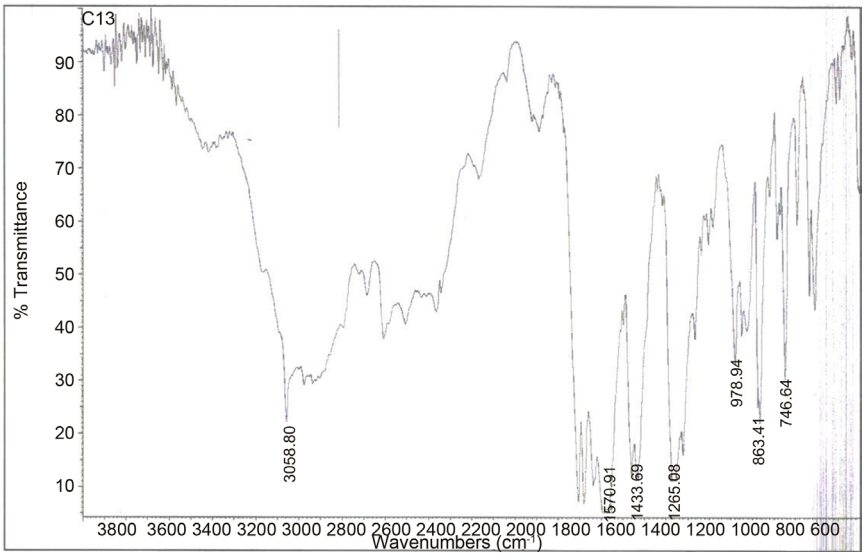


Figure 1. Infrared spectrum of 13C.

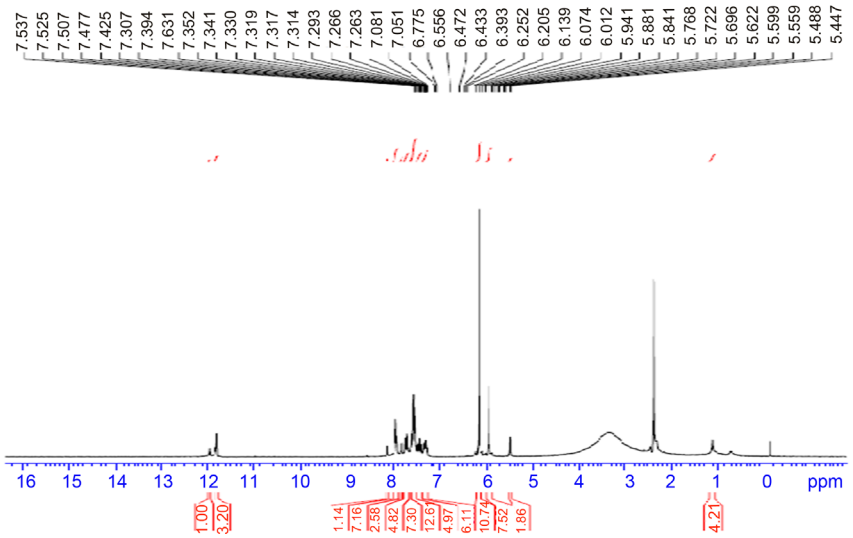


Figure 2. [1]HNMR spectrum of compound 15c.

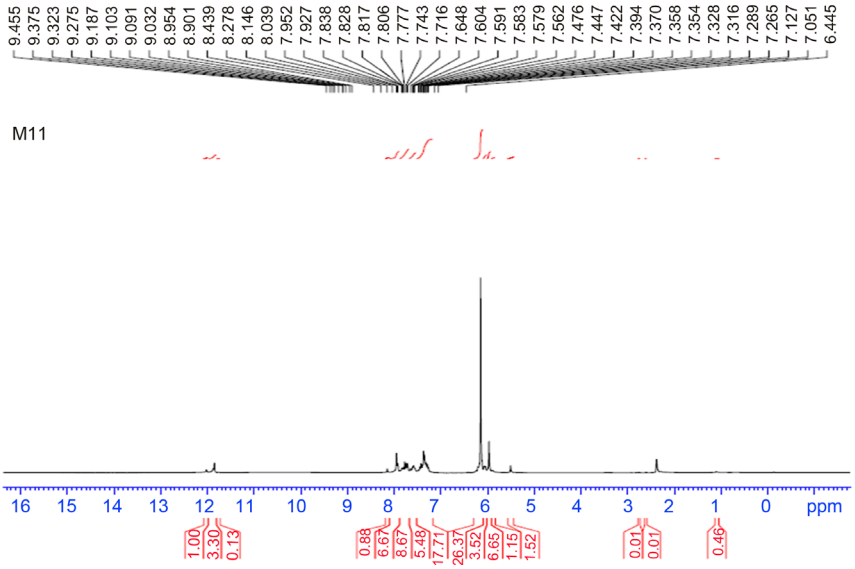


Figure 3. [1]HNMR spectrum of compound 11c.

Table 6. Proton NMR spectra data for the synthesis compounds (11c-18c).

Comp. No.	*Proton values* (δ, ppm)					
	C-H aromatic	CH=CH	N-H	-CH2-aliphatic	C-H (ring)	Others
11c	7.2 - 7.9	6.3 - 6.9	11.8	5.3	8.01	(OH)12
12c	7.15 - 8	6.3 - 6.91	10.9	5.4	8	(OH) 9.01
13c	7.3 - 7.8	6 - 6.92	11.1	5.3	8.01	
14c	7.1 - 8	6.4 - 6.9	11.3	5.3	7.9	(-$N(CH_3)_2$)2.6
15c	7.2 - 7.8	6.1 - 6.3	11.9	5.4	8.1	(-CH=CH-)=5.9-6
16c	6.9 - 7.7	6.3 - 6.8	11.1	5.6	7.9	
17c	7 - 7.9	6.2 - 6.9	10.9	5.2	8	
18c	7.1 - 7.9	6.3 - 6.9	11.2	5.4	8.02	

buted to the chemical shift of carbon (13) proton its between carbonyl amide and benzotriazol moiety. The signal related to the protons at carbons (9, 10) appeared at 6.1 - 6.3 ppm in seven membered ring, the peak at 8.1 ppm was attributed to the chemical shift of carbon (7) proton due to between two highly electronegativity atoms (oxygen and nitrogen). The values of NMR are illustered in Table 6.

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