Synthesis and characterization of heterocyclic compound derivatives from 2-azo benzothiazole have possible biological activity

Entesar.O. AL-tememi*, Maha.K.Mahmmod**

* Department of chemistry, College of Science, Baghdad University, Baghdad, Iraq
** Department of chemistry, College of Science, Karbala University, Karbala, Iraq

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Abstract

Reaction of different substi tuents of 2-azobenzothiazolyl 4-phenols (I) with ethyl 2-bromo propionate to produce the derivatives (II) that react with thiosemicarbazide gave N-substituted thiosemicarbazide (III) also react with semicarbazide and which gave N-substituted semicarbazide (IV). The 1,2,4-triazole derivatives (V) and 1,3,4- thiadiazole derivatives (VI) were prepared by the cyclization of N-substituted thiosemicarbazide (III) with base and with concentrated sulphuric acid respectively . The 1,3,4-oxadiazole derivatives (VII) were prepared by the cyclization of N-substituted semicarbazide (IV) with acid. the new compounds were established on the basis of IR, 1H NMR and UV spectral data.

تحضير وتشخيص مركبات حلقية غير متجانسة للمركب 2-ازوبنزوثايازول ذات الفعالية البايلوجية المحتملة

ا.د. انتصار عبيد التميمي * م. م مها قاسم محمود **

* قسم الكيمياء / كلية العلوم / جامعة بغداد
** قسم الكيمياء / كلية التربية / جامعة كربلاء

الخلاصة

تفاعل مشتقات مختلفة للمركب 2-ازوبنزوثايازول 4- فينول (I) مع اثل 2-بروموبروبيونيت لتكوين مشتقات استرية مقابلة(II) التي بتفاعلها مرة مع الثايوسيميكاربازايد تعطي مشتقات N – ثايوسيميكاربازايد (III) ومع السيميكاربازايد لتحضير مشتقات N- مشتقات اليميكاربازايد (IV) . مشتقات 1,2,4– ترايازول (V) و مشتقات 1,3,4- ثاياديازول (VI) تم تحضيرها من عملية الغلق الحلقي لمشتقات الثايوسيميكاربازايد بوجود القاعدة والحامض بالتعاقب . مشتقات 1,3,4- اوكسودايازول تم تحضيرها عن طريق الغلق الحلقي لمشتقات السيميكاربازايد بوجود الحامض .

Introduction

Several 2-aminobenzothiazole derivatives have long been known for their diverse biological properties, mainly as antibacterial [1], antifungal[2] , antitumor[3] and anti-inflammatory[4] agents. In addition, heterocycles bearing 1,2,4 triazole or 1,3,4-thiadiazole or 1,3,4 oxadiazole moieties represent an interesting class of compounds possessing a wide spectrum of biological activities such as anti-inflammatory[5], antiviral[6], antimicrobial[7,8] , antiinflammatory[9], antitumor[10] , antifungal and antibacterial[11] properties. Similarly diazo compounds also shows a biological activity such as antibacterial[12], antiviral[13], antifungal[14] .

Experimental

Melting points were determined in open capillary tubes. IR spectra were recorded on a perkin-Elmer 157 spectrometer and ^{1}HNMR spectra on a Bucker WM-400 (400 MHZ FTNMR) spectrophotometer using TMS (Tetramethyl Silane) as internal reference (chemical shift in ppm). Purity of the prepared compounds was checked by TLC (Thin Layer Chromatography) on silica gel plates and spot were visualized by exposure to iodine vapours. The physical data of the prepared compounds are presented in Table 1, and the spectroscopic data are presented in Table 2 .

General procedure for the preparation of ethyl-{ρ-(2-diazo-substituted benzo-thiazolyl) substituted phenoxy }2-propanoate[15] (II)

To a solution of ρ-(2-diazo-substituted benzothiazolyl) subsituted phenols (0.5 mole) in dry acetone (150 mL), ethyl 2-bromopropanate (0.60 mole) were added followed by anhydrous K_2CO_3 (1.0 mole). After reflux for 20-24 hr. the reaction mixture was filtered and the solvent was removed under reduce pressure then poured into ice water, the mixture was standin for 3 days then the precipitate was filtered washed with water to give ester (II).

General procedure for the preparation of N- substituted thiosemicarbazide and N-subsituted semicarbazide[15](III),(IV).

To a round bottom flask, compound (I) (0.01 mol), thiosemicarbazide (0.01 mol) or (0.01 mol) semicarbazide and absolute alcohol (50 mL) were mixed. A condenser with calcium guard tube was attached to the flask and the mixture was refluxed for 4-5 hours on water bath. The reaction progression was monitored by thin layer chromatography (TLC),. The mixture was concentrated, cooled and poured in crushed ice. It was kept for 3-4 hours at room temperature and solid mass separated out was filtered and dried, recrystallized by methanol to give pure compounds (III,IV).

General procedure for the preparation of 5-{substituted } 2-amino (1,3,4) oxadiazole[16] (VII).

A mixture of N-substituted semicarbazide (IV) (0.01 M) and concentrated sulphuric acid (10 ml) was refluxed for half an hour and kept at room temperature for 24 h. The contents were poured into cold water and neutralized with diluted sodium carbonate solution. The product was isolated and crystallized from ethanol to give pure compounds (VII).

General procedure for the preparation of 5- {substituted} 3- mercapto- 1, 2, 4 - triazole[17] (V).

N-substituted thiosemicarbazide (III) (3 mmol) was added portion wise to 25 mL of 2N sodium hydroxide solution. The reaction mixture was refluxed, and the completion of the reaction was checked by TLC. The mixture was then allowed to cool to room temperature. It was filtered and then the filtrate was acidified with 2N hydrochloric acid. The precipitated solid

was filtered, washed thoroughly with water, dried and recrystallized from ethanol/water (80/20) to give pure compounds (V).

General procedures for the preparation of 2-amino-5(substituted)1-3-4 thiadiazole[18] (VI).

N-substituted thiosemicarbazid (III) (0.6 mmol) were added portion wise to 25mL of conc. sulfuric acid at ^{0} 0 C with continuous stirring. The reaction mixture was stirred further for 3 h at room temperature. Then it was poured into an ice-water mixture to precipitate a crude solid. The crude product was then recrystallized from a mixture of acetic acid and water (1:1 or 1:2) to furnish disubstituted 1,3,4-thiadiazole .

Results and Discussion

In view of the activities exhibit by benzothiazoles, diazo compounds and five member hetrocyclic compound, a new series of 2-azobenzothiazole derivatives, in which the adamantly moiety was attached to 1,2,4-triazole, 1,3,4- thiadiazole and 1,3,4-oxadiazole nucleus, have been synthesized as possible potential bioactive agents. The reaction sequence leading to the formation of desired heterocyclic compounds are outlined in Scheme 1. Reaction of compounds (I) with ethyl 2-bromo propionate in boiling absolute ethanol containing anhydrous potassium carbonate as catalyst, afforded the intermediate N-substituted thiosemicarbazi -de (III) and N-substituted semicarbazid (IV). The preparation of 5-subsituted-2-amino-1,3,4-thiadiazole (VI) and 5-subsituted-2-amino-1,3,4-oxadiazole (VII), respectively, was described in many reactions. As the initial substance, N-substituted thiosemicarbazide and N-substituted semicarbazide, respectively, is used.

Their cyclization happens in the presence of various substances, e.g. conc. sulphuric acid, phosphoric acid and acylchloride .
The mechanism of the reaction is shown in scheme (2) The preparation of 5-subsituted-1,2,4-triazol-3-thion was also described in various reactions. The cyclization of N-substituted semicarbazide happens best in the presence of hydroxides.
The mechanism of the reaction involves a nuclephilic addition as is shown in scheme(3).

Conclusion

Infrared spectra of all the prepared esters showed many absorption bands of stretching and bending vibration of different groups. In general, the FT-IR spectra showed a new bands appeared in compound (II) spectra, at (2723-2993) cm^{-1} due to the weaker C-H stretching for (CH_2, CH_3) groups, the characteristic band at (1250-1270) cm^{-1} for the C-O-C stretching indicate the presence of ether group also band at (1730-1750) cm^{-1} for (C=O) of ester stretching. The ^{1}H-NMR spectra showed a signal between δ7.421-823 ppm for five aromatic hydrogen and the signal at δ1.731 ppm (1H, multeplet) which was attributed to (-CH) proton, while the signal at δ2.512 ppm (2H, multeplet) was attributed to ($-CH_2$) proton, also signal at δ1.125 ppm (3H, triplet) was attributed to ($-CH_3$), and signal at δ2.889 ppm (3H, duplet) was attributed to ($-CH_3$). The ^{13}C-NMR of compound (II) showed the signals at 115-144 ppm for aromatic carbons, while the signal at 170 ppm for carbon of carbonyl ester, and signal at 65 ppm for carbon in thiazole ring, also signals at 16-25 ppm for aliphatic carbons . The FT-IR spectra of N-substituted thiosemicarbazide (III) and N-substituted semicarbazide (IV) showed.

characteristic bands for NH groups at 3210- 3240 cm^{-1}, and the absorption band for .
C=O amide group appeared at 1710-1730 cm^{-1} and absorption band for C=S group appeared at 1110-1120 cm^{-1} ,
the absorption band of C=O ester group was disappeared and this good indication for complete the reaction. The absorption bands of substituted thiosemicarbazides **(**III**)** was eliminated by the formation of 1,2,4-triazoles **(**IV**)** or 1,3,4-thiadiazoles **(**VI**)** rings, the substituted-s-triazole-3-thioles **(**V**)** showed a band in the region 1280-1290 cm^{-1} attributed to (C=S). None of that triazoles showed absorption in the region 2578-2650 cm^{-1} for SH.
The absence of S-H and presence of absorptions (C=S) and N-H established that all the isolated triazoles are in their thion rather than the thiole form. The absorption bands of substituted semicarbazides **(**IV**)** in the region between 1715 and 1733 cm^{-1},due to carbonyl, which was eliminated by the formation of 1,3,4-oxadiazoles (VII). In summary, we have described the synthesis of possible antimicrobial active 3-[(2-diazo substituted benzothiazole) 4-substituted phenoxy] -1,2,4-triazole-5-thiol, 2-[2-diaazo substituted benzothiazole [4-substituted phenoxy]-1,3,4-oxadiazole-5-amine and 2-[2-diazo substituted benzothiazole [4-substituted phenoxy]-1,3,4-thiadiazole-5-amine derivatives.

The absorption bands of N-substituted thiosemicarbazides **(III)** in the region between 1690 and 1722 cm^{-1}, due to carbonyl, which was eliminated by the formation of 1,2,4-triazoles **(V)** or 1,3,4-thiadiazoles **(VI)** rings, the substituted-s-triazole-3-thioles **(V)** showed a band in the region (1280-1290) cm^{-1} attributed to (C=S). None of that triazoles showed absorption in the region (2578-2650) cm^{-1} SH. The absence of S-H and presence of absorptions C=S and N-H established that all the isolated triazoles are in their thion rather than the thiole form.

The absorption bands of N-substituted semicarbazides **(IV)** in the region between 1715 and 1733 cm^{-1},due to carbonyl, which was eliminated by the formation of 1,3,4-oxadiazoles (**IV**).

The antimicrobial screening still to be done. However, the activities of these compounds are expected since it has three active groups thiazol ring, azo group and five member heterocyclic rings.

Acknowledgement

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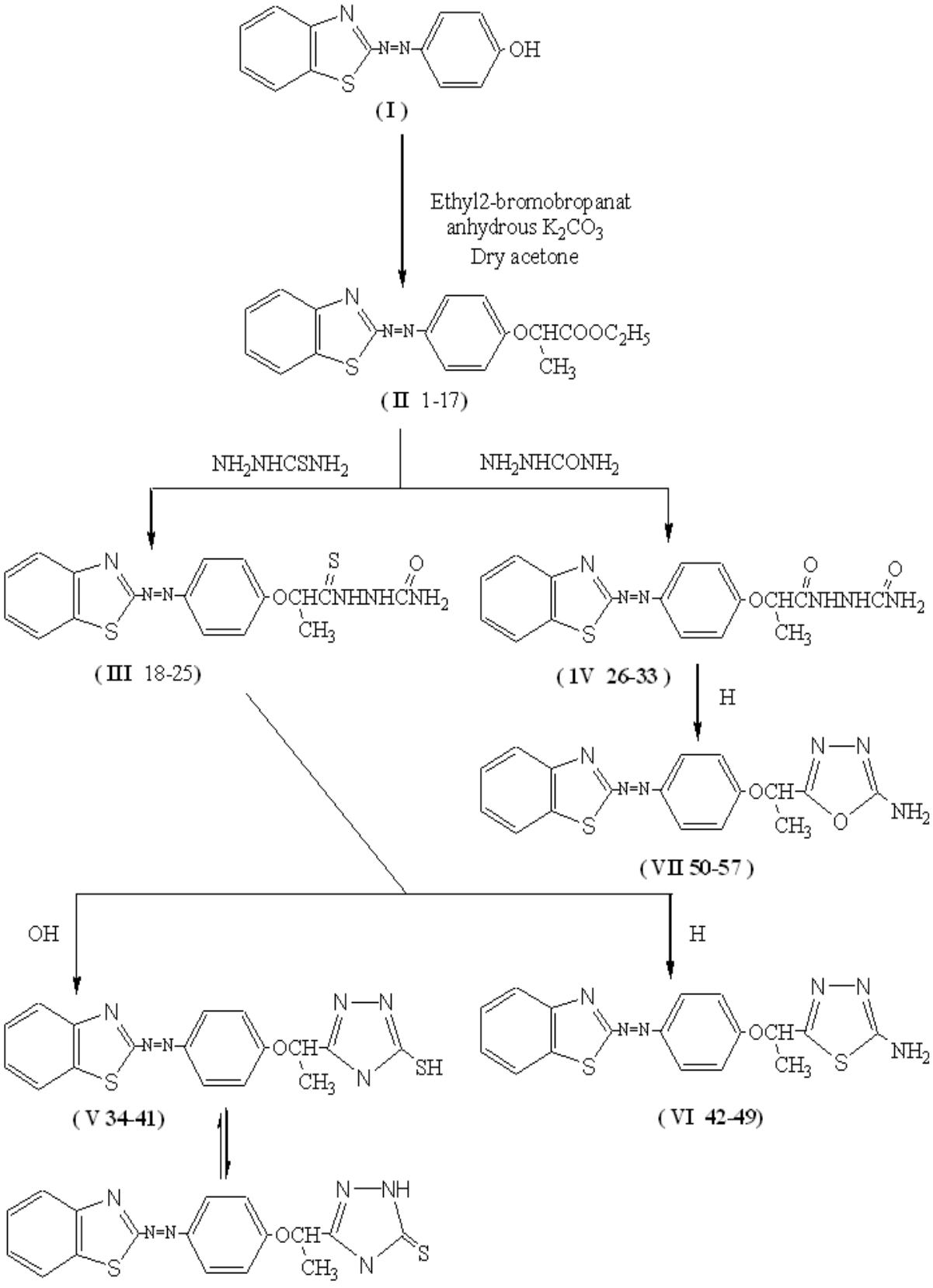
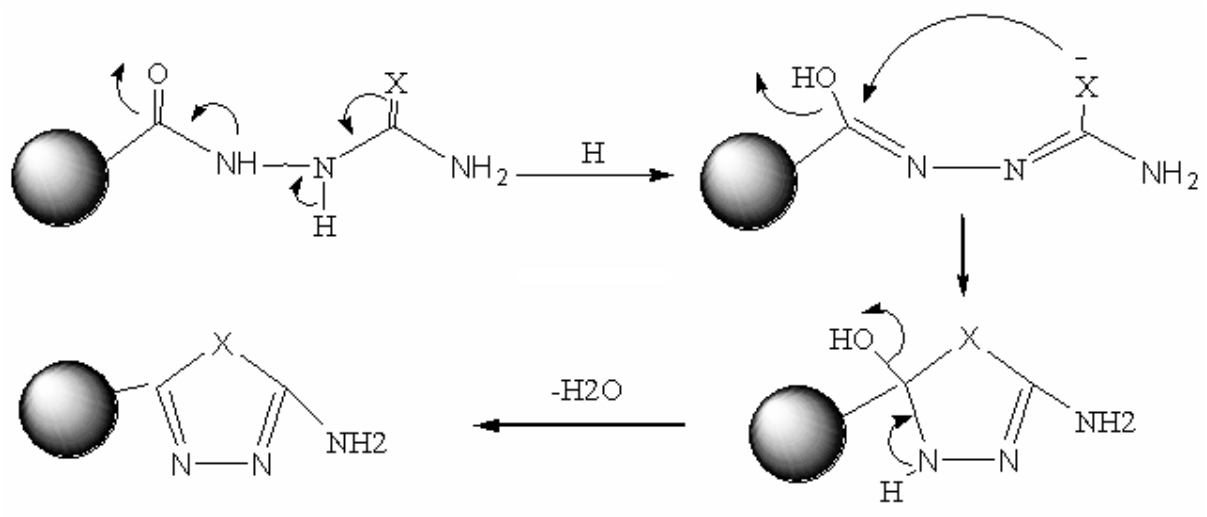
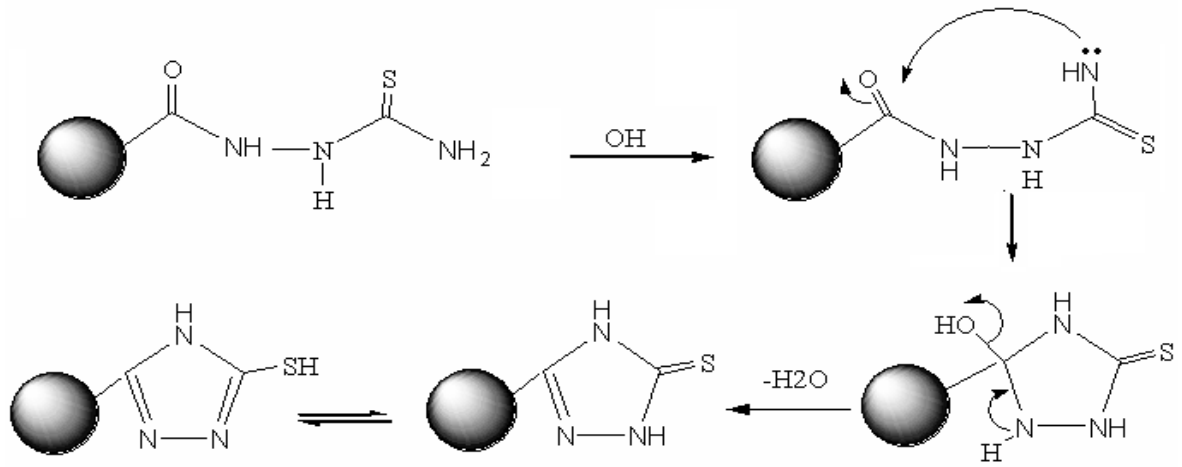


Scheme (1): syntheses of the prepared compounds



Scheme (2): Mechanism of preparation of thiazole and oxadiazole



Scheme (3): Mechanism of preparation triazole

Table 1. Characterization data of the compounds.

Com. No.	Substituted				Molecular formula Molecular weigh	M.P	Yield %	R_f
	R1	R2	R3	R4				
1	HCOO	H	CH_3	CH_3	$C_{21}H_{21}O_5N_3S$ (428)	180 Dec.	60	0.43
2	NO_2	H	H	H	$C_{18}H_{16}O_5N_4S$ (400)	140	80	0.74
3	CH_3	H	Br	H	$C_{19}H_{18}O_3N_3SBr$ (448)	Syrup	50	0.46
4	NO_2	H	C_2H_5	H	$C_{20}H_{20}O_5N_4S$ (428)	Syrup	66	0.69
5	NO_2	H	NO_2	H	$C_{18}H_{15}O_7N_5S$ (445)	165 Dec.	65	0.66
6	NO_2	H	NH_2	H	$C_{18}H_{17}O_5N_5S$ (415)	Syrup	60	0.61
7	Cl	NO_2	CH_3	CH_3	$C_{20}H_{19}O_5N_4SCl$ (462)	Syrup	80	0.78
8	Cl	NO_2	H	H	$C_{18}H_{15}O_5N_4SCl$ (434)	Syrup	75	0.56
9	Cl	NO_2	NH_2	H	$C_{18}H_{16}O_5N_4SCl$ (435)	Syrup	83	0.79
10	Cl	NO_2	NO_2	H	$C_{18}H_{14}O_7N_5SCl$ (479)	Syrup	60	0.86
11	Cl	NO_2	Br	H	$C_{18}H_{14}O_5N_4SBrCl$ (513)	Syrup	90	0.88
12	NO_2	NO_2	H	H	$C_{18}H_{15}O_7N_5S$ (445)	Syrup	70	0.66
13	NO_2	NO_2	C_2H_5	H	$C_{20}H_{19}O_7N_5S$ (473)	Syrup	85	0.53
14	NO_2	NO_2	NO_2	H	$C_{18}H_{14}O_9N_6S$ (490)	Syrup	94	0.85
15	NO_2	NO_2	Br	H	$C_{18}H_{14}O_7N_5SBr$ (533)	Syrup	75	0.77
16	NO_2	NO_2	NH_2	H	$C_{18}H_{16}O_7N_6S$ (460)	Syrup	50	0.36
17	NO_2	NO_2	CH_3	CH_3	$C_{20}H_{19}O_7N_5S$ (473)	Syrup	65	0.51
18	CH_3	H	Br	H	$C_{18}H_{17}O_2N_6S_2Br$ (492)	170	63	0.72
19	NO_2	H	H	H	$C_{17}H_{15}O_4N_7S_2$ (445)	150	55	0.74

Com. No.	Substituted				Molecular formula Molecular weigh	M.P	Yield %	R_f
	R1	R2	R3	R4				
20	NO_2	H	C_2H_5	H	$C_{19}H_{19}O_4N_7S_2$ (473)	Syrup	78	0.52
21	Cl	NO_2	CH_3	CH_3	$C_{19}H_{18}O_4N_7S_2Cl$ (507)	Syrup	60	0.73
22	Cl	NO_2	NH_2	H	$C_{17}H_{15}O_4N_8S_2Cl$ (480)	Syrup	65	0.54
23	NO_2	H	NO_2	H	$C_{17}H_{14}O_6N_8S_2$ (490)	Syrup	80	0.65
24	Cl	NO_2	Br	H	$C_{17}H_{13}O_4N_7S_2Br$ (522)	Syrup	70	0.55
25	NO_2	NO_2	H	H	$C_{17}H_{14}O_6N_8S_2$ (490)	Syrup	75	0.61
26	CH_3	H	Br	H	$C_{18}H_{17}O_3N_6SBr$ (476)	Syrup	65	0.73
27	NO_2	H	H	H	$C_{17}H_{15}O_5N_7S$ (429)	Syrup	50	0.57
28	NO_2	H	C_2H_5	H	$C_{19}H_{19}O_5N_7S$ (457)	Syrup	75	0.69
29	Cl	NO_2	CH_3	CH_3	$C_{19}H_{18}O_5N_7SCl$ (491)	Syrup	60	0.43
30	Cl	NO_2	NH_2	H	$C_{17}H_{15}O_5N_7SCl$ (464)	Syrup	65	0.83
31	NO_2	H	NO_2	H	$C_{17}H_{14}O_7N_8S$ (474)	Syrup	80	0.92
32	Cl	NO_2	Br	H	$C_{17}H_{13}O_5N_7SBrCl$ (541)	Syrup	75	0.81
33	NO_2	NO_2	H	H	$C_{17}H_{14}O_7N_8S$ (474)	Syrup	85	0.78
34	CH_3	H	Br	H	$C_{18}H_{15}O_2N_6SBr$ (458)	Syrup	65	0.62
35	NO_2	H	H	H	$C_{17}H_{13}O_4N_7S$ (411)	Syrup	70	0.74
36	NO_2	H	C_2H_5	H	$C_{19}H_{17}O_4N_7S$ (439)	Syrup	85	0.59
37	Cl	NO_2	CH_3	CH_3	$C_{19}H_{16}O_4N_7SCl$ (473)	Syrup	55	0.77
38	Cl	NO_2	NH_2	H	$C_{17}H_{13}O_4N_8SCl$ (460)	Syrup	65	0.81

Com. No.	Substituted				Molecular formula Molecular weigh	M.P	Yield %	R_f
	R1	R2	R3	R4				
39	NO_2	H	NO_2	H	$C_{17}H_{12}O_6N_8S$ (456)	Syrup	80	0.66
40	Cl	NO_2	Br	H	$C_{17}H_{11}O_4N_7SBrCl$ (524)	Syrup	73	0.38
41	NO_2	NO_2	H	H	$C_{17}H_{12}O_6N_8S$ (456)	Syrup	88	0.65
42	CH_3	H	Br	H	$C_{18}H_{15}N_6S_2OBr$ (484)	Syrup	70	0.71
43	NO_2	H	H	H	$C_{17}H_{16}N_7S_2O_3$ (430)	210 Dec.	66	0.61
44	NO_2	H	C_2H_5	H	$C_{19}H_{17}N_7S_2O_3$ (455)	Syrup	75	0.95
45	Cl	NO_2	NH_2	H	$C_{17}H_{13}N_8S_2O_3Cl$ (476)	Syrup	80	0.49
46	Cl	NO_2	Br	H	$C_{17}H_{11}N_7S_2O_3Cl$ Br (549)	Syrup	85	0.84
47	NO_2	NO_2	H	H	$C_{17}H_{12}N_8S_2O_5$ (472)	Syrup	60	0.61
48	NO_2	H	NO_2	H	$C_{17}H_{12}N_8S_2O_5$ (472)	Syrup	65	0.95
49	Cl	NO_2	CH_3	CH_3	$C_{19}H_{16}N_7S_2O_3Cl$ (489)	Syrup	63	0.67
50	NO_2	H	H	H	$C_{17}H_{13}N_7S_2O_3$ (427)	Syrup	50	0.54
51	NO_2	H	C_2H_5	H	$C_{19}H_{17}N_7S_2O_3$ (455)	Syrup	70	0.93
52	Cl	NO_2	CH_3	CH_3	$C_{19}H_{16}N_7S_2O_3Cl$ (489)	Syrup	63	0.81
53	NO_2	H	NO_2	H	$C_{17}H_{12}N_8S_2O_5$ (472)	Syrup	75	0.65
54	NO_2	NO_2	H	H	$C_{17}H_{12}N_8S_2O_5$ (444)	Syrup	60	0.83
55	CH_3	H	Br	H	$C_{18}H_{15}N_6S_2OBr$ (484)	Syrup	77	0.76
56	Cl	NO_2	NH_2	H	$C_{17}H_{13}N_8S_2O_3Cl$ (476)	Syrup	50	0.64
57	Cl	NO_2	Br	H	$C_{17}H_{11}N_7S_2O_3ClBr$ (549)	Syrup	77	0.89

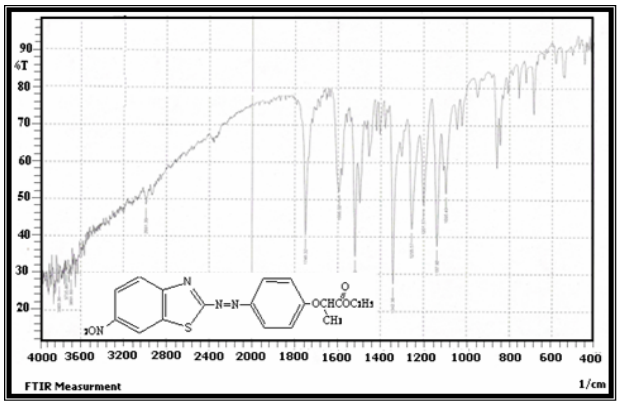
Table 2. Spectroscopic data for the prepared compounds

Com. No.	Spectroscopic data
1	IR (KBr, cm^{-1}): 1250, 1745, 1575, 3309-3249, 1789. UV (□□$_{max}$ / nm ethanol):212 .
2	IR (KBr, cm^{-1}): 1590, 1530,1338, 1765, 1253. UV (□□$_{max}$ / nm ethanol): 258,377: ^{1}H-NMR (DMSO)□□1.12□□ t,3H^{1}, CH_3);□□2.51(m 2H^{2}, CH_2);□□1.73□□m,1H^{3},-CH);□□2.88□□d, 3H^{4} , CH_3) ;□□□□□□□d,1H^{5},HAr.);□□7.65(d, 1H^{6}, H Ar.);□□8.1□d, 1H^{7}, H Ar.);□□8.24□□d, 1H^{8}, H Ar.); □7.94□□s, 1H^{9}, H Ar.): ^{13}C-NMR 115-144, 170, 65.
3	IR (KBr, cm^{-1}) 840,1615,1744,1253: UV (□□$_{max}$/ nm ethanol):227, 328.
4	IR (KBr, cm^{-1}):1233,1760,1593,1533,1361,2943.UV (□□$_{max}$/ nm ethanol):382.
5	IR (KBr, cm^{-1}): 1255, 1601, 1549, 1367, 1743.
6	IR (KBr,cm^{-1}):1235, 1759, 1599, 1549, 1367,3382.
7	IR (KBr, cm^{-1}):1244, 1740, 1513, 1551, 1368, 845:UV (□□$_{max}$ / nm ethanol):211, 352.
8	IR (KBr, cm^{-1}):1230, 1750, 1593, 1553, 1363,640: UV (□□$_{max}$ / nm ethanol):232,274.
9	IR (KBr, cm^{-1}):1220, 1765, 1579, 1559, 1365: UV (□□$_{max}$ / nm ethanol):207, 268.
10	IR (KBr, cm^{-1}): 1270,1735, 1605,1563, 1375 :UV (□□$_{max}$ / nm ethanol):234,261.
11	IR (KBr, cm^{-1}):1264, 1740, 1593, 1538, 1353,657, 877: UV (□□$_{max}$ / nm ethanol) 263, 342.
12	IR (KBr, cm^{-1}):1253, 1744, 1605, 1536, 1355: UV (□□$_{max}$ / nm ethanol): 257, 225.
13	IR (KBr, cm^{-1}):1230, 1761, 1575, 1567, 1344, 659:UV (□□$_{max}$ / nm ethanol) 254, 325.
14	IR (KBr, cm^{-1}):1255, 1757, 1604, 1550, 1340: UV (□□$_{max}$ / nm ethanol): 221, 345.
15	IR (KBr, cm^{-1}):1251, 1745, 1600, 1545, 1364.
16	IR (KBr, cm^{-1}):1245, 1765, 1601, 1537, 1324, 1432.
17	IR (KBr, cm^{-1}):1253, 1749, 1605, 1557, 1334, 640, 1433.
18	IR (KBr, cm^{-1}):1712, 1605, 1105, 3210, 832,2932.

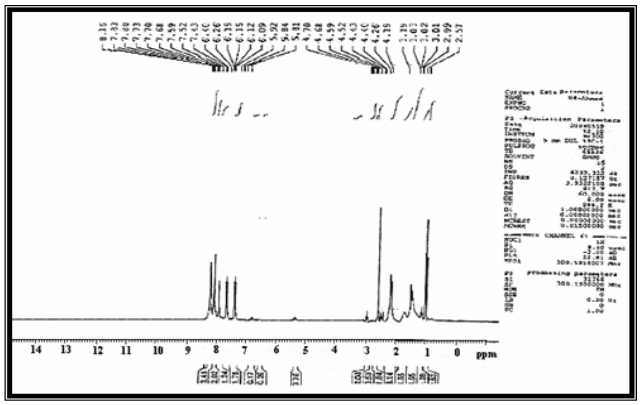
Com. No.	Spectroscopic data
19	IR (KBr, cm^{-1}):1563,1375, 3301, 1115, 1595, 1710 :UV (□□$_{max}$ / nm ethanol): 252,372.
20	380,267IR (KBr, cm^{-1}): 1560,1371,2944,3322,1118,1600,1711:UV (□□$_{max}$ / nm ethanol):
21	IR (KBr, cm^{-1}): 1555,1365,642,2935,3310,1153,1583,1696: UV (□□$_{max}$ / nm ethanol)231,393.
22	IR (KBr, cm^{-1}): 1550,1356,645,3356,3230,1113,1585,1699:UV (□□$_{max}$ / nm ethanol)207,268.
23	IR (KBr, cm^{-1}): 1561,1363,3329,1110,1600,1700: UV (□□$_{max}$ / nm ethanol)235,255.
24	IR (KBr, cm^{-1}): 863,1563,1375,3301,1172,1596,1712: UV (□□$_{max}$ / nm ethanol) 233,403.
25	IR (KBr, cm^{-1}): 1566,1353,3336,1139,1609,1698: UV (□□$_{max}$ / nm ethanol) 237, 337.
26	321,229 IR (KBr, cm^{-1}):1680, 1583, 1659, 3355, 2992: UV (□□$_{max}$ / nm ethanol)
27	IR (KBr, cm^{-1}):1586, 1600, 3350, 1534, 1338, 1688: UV (□□$_{max}$ / nm ethanol)251, 371.
28	IR (KBr, cm^{-1}):1582, 1601, 3405, 1690, 1536,1335, 2989: UV (□□$_{max}$ / nm ethanol)227, 380.
29	IR (KBr, cm^{-1}): 1566, 1603, 3352, 1710, 655, 2995 UV (□□$_{max}$ / nm ethanol)232, 391.
30	IR (KBr, cm^{-1}):1569, 1602, 3322, 1701 650, 1553,1327: UV (□□$_{max}$ / nm ethanol) 217.5, 258.5
31	IR (KBr, cm^{-1}):1550, 1633, 3390, 1690, 1542, 1333 :UV (□□$_{max}$ / nm ethanol)291, 352.
32	IR (KBr, cm^{-1}):1563, 1610, 3381 1715, 649, 827:UV (□□$_{max}$ / nm ethanol)231, 413.
33	IR (KBr, cm^{-1}):1549, 1600, 3355, 1722, 1532, 1344:UV (□□$_{max}$ / nm ethanol)243, 278.
34	IR (KBr, cm^{-1}):3331, 1583, 1601, 1255, 2997, 843:UV (□□$_{max}$ / nm ethanol)243, 371.
35	IR (KBr, cm^{-1}):3310, 1589, 1600, 1250 1554, 1345: UV (□□$_{max}$ / nm ethanol)257,381:^{1}H-NMR(DMSO)□□4.23(s,2H^{1},NH_2);□□2.48(m 1H^{2},CH)□□□□□□□□□□d,3H^{3},CH_3);□□7.19□d,1H^{4},HAr.);□□7.26□d,1H^{5}, H Ar.);□□7.76□d, 1H^{6}, H Ar.);□□7.93□□d, 1H^{7}, H Ar.);□□7.69□□s,1H^{8},H Ar.): ^{13}C-NMR 120-125, 54,

Com. No.	Spectroscopic data
36	IR (KBr, cm^{-1}):3350, 1591, 1601, 1225: UV (□□$_{max}$ / nm ethanol)217, 303
37	IR (KBr, cm^{-1}):3373, 1566, 1625, 1234, 2998 , 655: UV (□□$_{max}$ / nm ethanol)210, 278.
38	IR (KBr, cm^{-1}):3305, 1595, 1620, 1235, 1562, 1356, 651: UV (□□$_{max}$ / nm ethanol)235, 262.
39	IR (KBr, cm^{-1})3315, 1600, 1615, 1310, 1553, 1341: UV (□□$_{max}$ / nm ethanol)235, 334.
40	IR (KBr, cm^{-1}):3335, 1612, 1635, 1258, 665, 844: UV (□□$_{max}$ / nm ethanol)238, 337.
41	IR (KBr, cm^{-1}):3326, 1600, 1625, 1250,1554, 1345.
42	IR (KBr, cm^{-1}):3331, 1583, 1601, 1255, 2997,843 .
43	IR (KBr, cm^{-1}):3310, 1589, 1600, 1250, 1554, 1345 :UV (□□$_{max}$ / nm ethanol)209,215:^{1}H-NMR(DMSO)□□6.23(s,1H^{1},NH);□□7.15(s,1H^{2},NH); □2.01□m,1H^{2}, CH); □1.23□d,3H^{3}, CH_3); □7.62□□d, 1H^{5}, H Ar.);□□7.81 □d, 1H^{6}, H Ar.);□□7.96□d,1H^{7} ,H Ar.) □8.31□d, 1H^{8}, H, Ar.);□□□□□□ □s, 1H^{9}, H Ar.): ^{13}C-NMR 115-135 , 60 , 165.25 .
44	IR (KBr, cm^{-1}):3350, 1591, 1601, 1225, 2988: UV (□□$_{max}$ / nm ethanol)206, 272.
45	IR (KBr, cm^{-1}):3373, 1566, 1625, 1234, 2998, 655: UV (□□$_{max}$ / nm ethanol):268, 342.
46	IR (KBr, cm^{-1}):3305, 1595, 1620, 1235, 1562, 1356, 651 :UV (□□$_{max}$ / nm ethanol):220, 282.
47	IR (KBr, cm^{-1}): 3315, 1600, 1615, 1310, 1553, 1341: UV (□□$_{max}$ / nm ethanol)218,271.
48	IR (KBr, cm^{-1}):3335, 1612, 1635, 1258,665, 844: UV (□□$_{max}$ / nm ethanol)295, 259.
50	IR (KBr, cm^{-1}):1231, 1567, 1626, 3335, 2915, 836 :UV (□□$_{max}$ / nm ethanol):370: ^{1}H-NMR(DMSO)□□4.35(s, 2H^{1}, NH_2);□□3.31 □m,1H^{2},-CH)□□□1.09□□d, 3H^{3}, CH_3);□□6.80 □d, 1H^{4}, H Ar.); □7.21□d, 1H^{5}, H Ar.);□□7.91 □d, 1H^{6}, H Ar.);□□8.13□d, 1H^{7}, H Ar.);□□7.01□s, 1H^{8}, H Ar.).^{13}C-NMR: 118-130, 63.
51	IR (KBr, cm^{-1}):1220, 1551, 1620, 3412, 1510, 1348 :UV (□□$_{max}$ / nm ethanol):215,380.
52	IR (KBr, cm^{-1}):1210,1583, 1625, 3410, 1588, 1339, 2910 : UV (□□$_{max}$ / nm ethanol):271, 392.

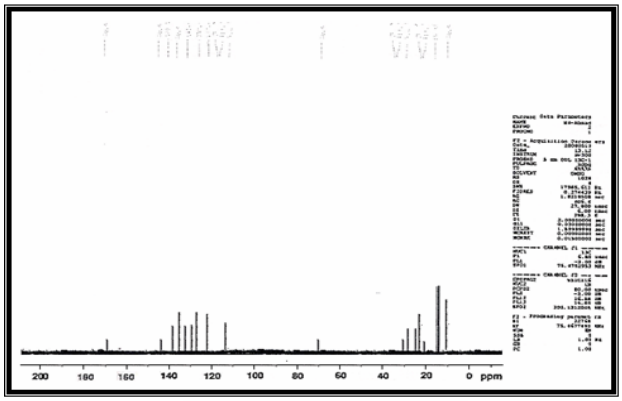
Com. No.	Spectroscopic data
53	IR (KBr, cm^{-1}): 1222, 1589, 1620, 3305, 1515, 1328, 635: UV (λ_{max} / nm ethanol)295, 353.
54	IR (KBr, cm^{-1}):1225, 1588, 1600, 3315, 1530, 1335, 848, 641: UV (λ_{max} / nm ethanol) 225, 237.
55	IR (KBr, cm^{-1}):1215, 1564, 1625, 3424, 1515, 1346 :UV (λ_{max} / nm ethanol)237, 325.
56	IR (KBr, cm^{-1}):1310, 1581, 1633, 3325, 1546, 1353 : UV (λ_{max} / nm ethanol): 212, 364.
57	IR (KBr, cm^{-1}):1322, 1590, 1608, 3315, 1553, 1374, 2913, 641 UV (λ_{max} / nm ethanol) 225, 303.



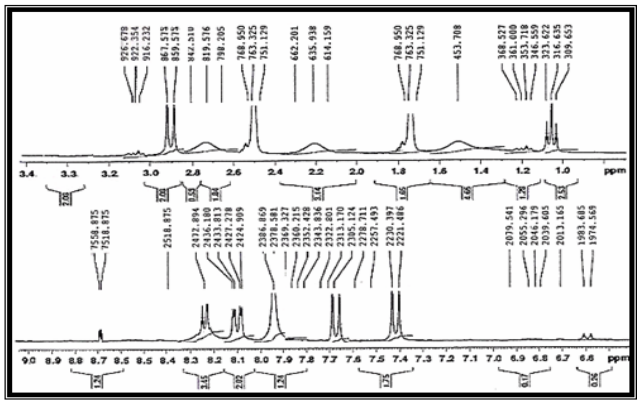
FT-IR spectra of compound (2)



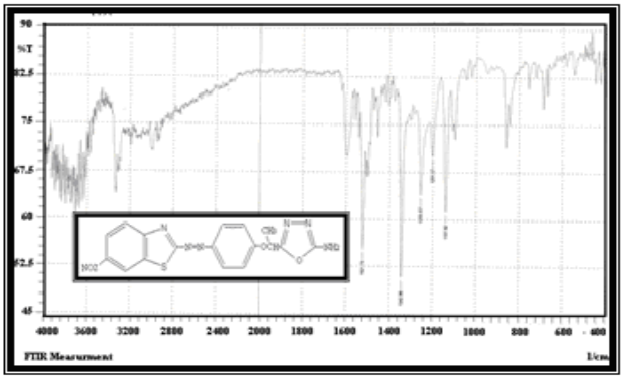
H-NMR spectra of compound (2)



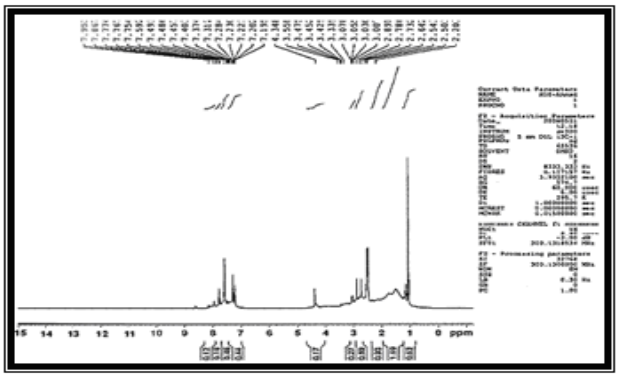
C-NMR spectra of compound (2)



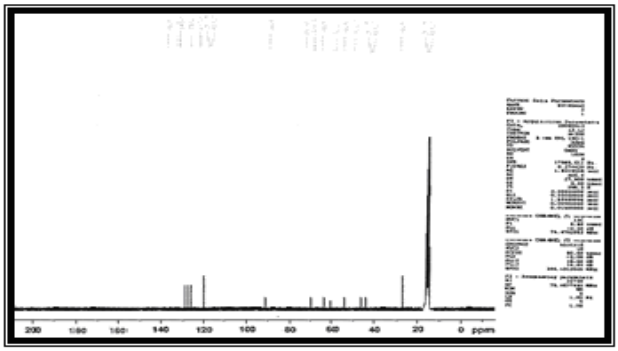
The expantion of H-NMR spectra



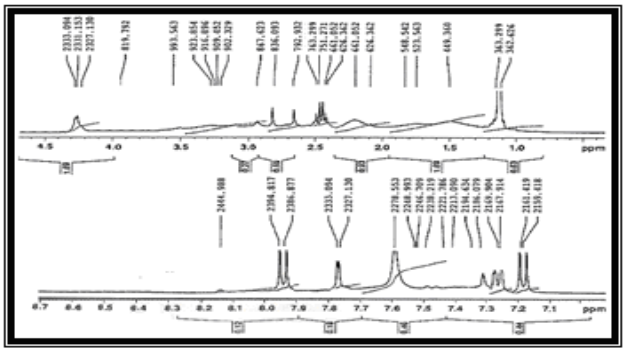
FT-IR spectra of compound (35)



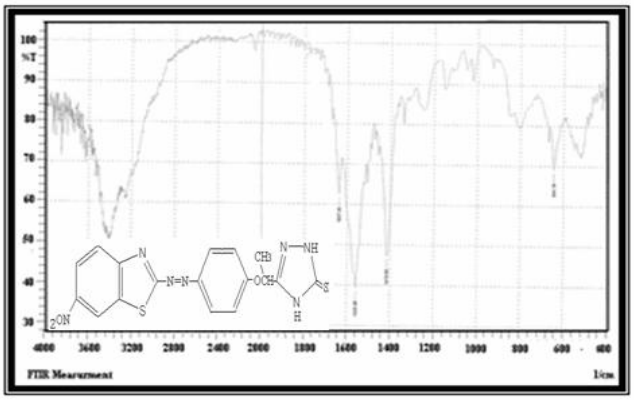
^{1}H-NMR spectra of compound (35)



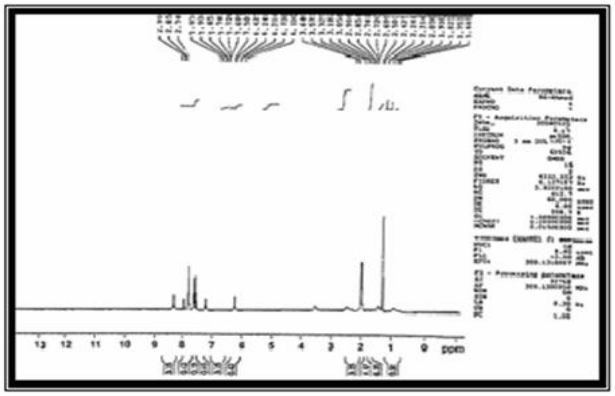
^{13}C-NMR spectra of compound (35)



The expantion of H-NMR spectra



FT-IR spectra of compound (43)



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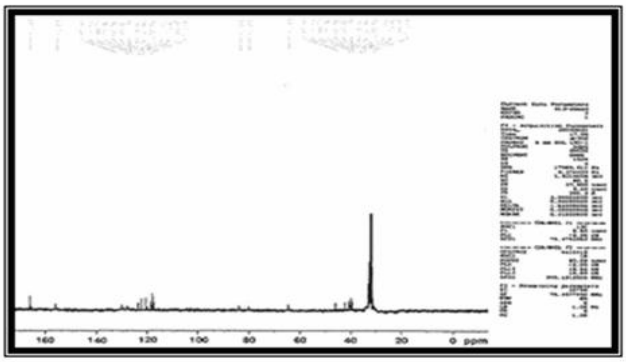
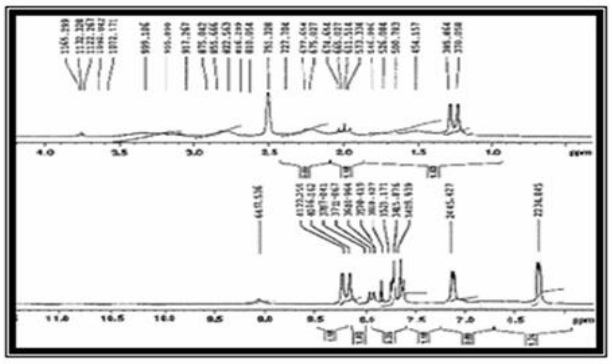
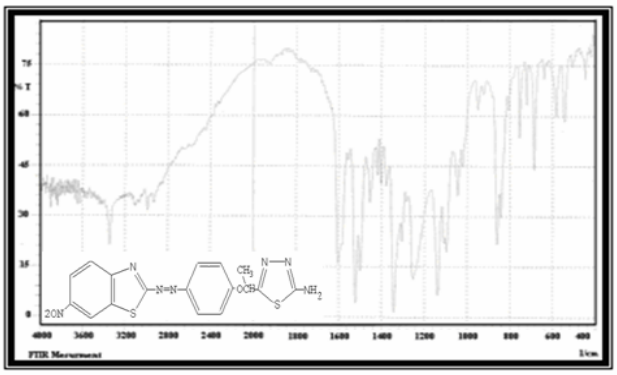


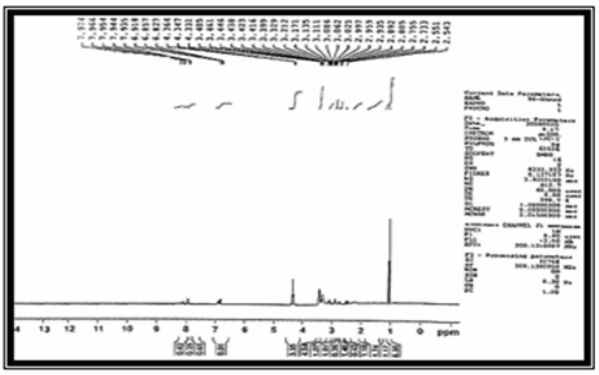
C-NMR spectra of compound (43)



The expantion of H-NMRspectra



FTIR spectra of compound (50)



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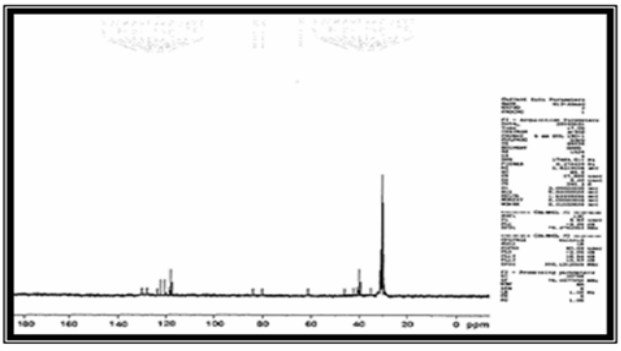
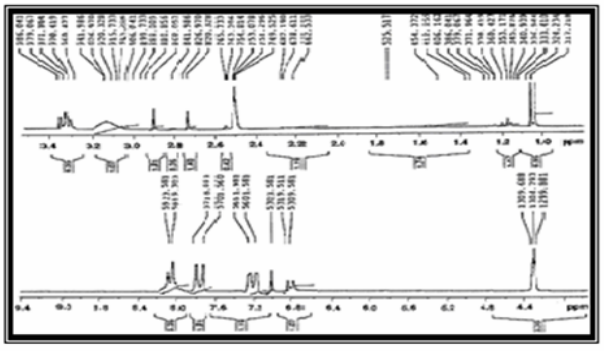


C-NMR spectra of compound (50)



The expantion of H-NMR spectra

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