Synthesis and Characterization of Some New Heterocyclic Compounds From Chalcone Derivatives

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الخلاصة

تم تحضير مشتقات جالكون عن طريق تفاعل بعض مشتقات البنزلديهايد مع مشتقات الثايوفين، النواتج بأمكانها التفاعل مع اليوريا، الثايوريا، أورثو فنلين ثنائي الامين و4،2- ثنائي نايترو فنيل هيدرازين، لأعطاء مشتقات حلقية غير متجانسة من الاوكسازين، الثيازين، الديازبين و البيرازول على التوالي. تمت متابعة التفاعل الكيميائي بأستخدام طريقة كروماتوغرافيا الطبقة الرقيقة. تم تشخيص تراكيب المركبات الحلقية غير المتجانسة الجديدة على أساس نقاط أنصهارها والقياسات الطيفية المتمثلة ب FT-IR, ^{13}CNMR و^{1}HNMR (لبعض منها).

الكلمات المفتاحية:- جالكون، أوكسازين، ثيازين، ديازبين و بيرازول.

Abstract

Chalcone derivatives were synthesized by reaction of some benzaldehyde derivatives with thiophene derivatives, The products were allowed to react with urea, thiourea, o-phenylendiamine and 2,4-dinitrophenylhydrazine, to give the heterocyclic derivatives of oxazine, thiazine, diazepine and pyrazole respectively. The chemical reaction were followed up by using thin layer chromatographic method. The characterization of newly synthesized heterocyclic compounds were established on the basis of their melting points and the spectral data like FT-IR, ^{13}CNMR and ^{1}HMNR (for some of them).

Key words:- *Chalcone, Oxazine, Thiazine, Diazepine and Pyrazole.*

Introduction

Chalcone and its derivatives are known for a wide spectrum of applications in the field of pharmaceuticals and other industries [1, 2]. For example, they have been found to exhibit various biological functions such as antioxidant, anti-inflammatory, analgesic, anticancer, antitubercular, antitumor, antimicrobial, and antihyperglycemic agents [3-6]. Chemically, chalcone consists of two aromatic rings separated by α,β-unsaturated carbonyl compound, and it is a versatile intermediate for synthesizing heterocyclic molecules [7, 8]. A series of chalcones

of 2-acetyl naphthalene and substituted aryl aldehydes were synthesized and evaluated for antimicrobial activity [9]. Chalcones (1,3-diaryl-2-propen-1-ones) are important intermediates for the synthesis of compounds such as flavonoids, isoflavonoids and their derivatives [10].Synthesis of chalcones and their derivatives were reported to have potential anti-inflammatory activity [11]. A series of chalcones were prepared by Claisen-Schmidt condensation of appropriate acetophenones with appropriate aromatic aldehydes in the presence of aqueous solution of potassium hydroxide and ethanol at room temperature [12]. Heterocyclic compound have been synthesized is due to potential biological and industrial application [13]. A series of new substituted diazepine derivatives have been synthesized by condensing substituted phenyl cyano ester with semicarbazide and thiosemicarbazide [14]. Oxazine derivatives are an important class of heterocycles, which has attracted much synthetic interest due to their wide range of biological activities. Oxazine is a heterocyclic compound can be formally derived from benzene, and its reduction products, by suitable substitution of carbon (and hydrogen) atoms by nitrogen and oxygen [15]. Pyrazol belongs to the family of azoles ,five membered heterocyclic [16], pyrazol ring is a prominent structural motif found in numerous pharmaceutically active compounds ,pyrazol important biological active such antiinflammatory, postmenopausal and osteoporosis [17]. Varity of thiazine derivatives have been reported claiming diversified biological profile like antimicrobial and anti-tuberculosis[18].

Experimental

Most chemicals were purchased from Fluke and BDH Chemical Ltd. General: uncorrected melting points were determined using Electrothermal melting point apparatus (Electro thermal , melting point , 9300 –U.K). Infrared spectra were recorded on (Shimadzu FTIR–prestige Fourier transform infrared Spectrophotometer). Carbon Nuclear Magnetic Resonance (^{13}C-NMR) and Proton Nuclear Magnetic Resonance (^{1}H-NMR) spectra were recorded with Brucker (300 MHz) using tetramethylsilane (TMS) as an internal standard, and DMSO as a solvent in Shahid Beheshti University of Medical Sciences and Health Services, Tehran,Iran.

1.Preparation of 3,3'-(2-chloro-1,4-phenylene)bis(1-(thiophen-2-yl)prop-2-en-1-one) [19]**. (B)** To 1-(thiophen-2-yl)ethanone (1.26 gm, 0.01 mole) a 2-chloroterephthalaldehyde (0.842

gm, 0.005 mole) was added in ethanol absolute (50 ml) and Sodium hydroxide (10%). The mixture was stirred for 11 hrs at room temperature using magnetic stirrer, the reaction was cold. Filtered and neutralized with dil.HCl. The reaction was monitored by T.L.C (2ml methanol:4ml benzene) and the solvent was evaporated and the precipitation was recrystallized from absolute Ethanol to give 3,3'-(2-chloro-1,4-phenylene)bis(1-(thiophen-2-yl)prop-2-en-1-one).

2.Preparation of 4,4'-(2-chloro-1,4-phenylene)bis(6-(thiophen-2-yl)-6H-1,3-oxazin-2-amine) (B1) [20]

A mixture of chalcone (B) (0.384 gm ,0.001 mol) , urea (0.12 gm,0.002 mol) were dissolved in(40 ml) absolute ethanol and sodium hydroxide (10 %) was refluxed 8 hrs, then pourd into 400ml cold water with continuous stirring for an hour and then kept in refrigerator for 24 hrs. The product was filtered ,washed and recrystallized by absolute ethanol and monitored by T.L.C (1.5ml methanol:3.5ml benzene).

3.Preparation of 4,4'-(2-chloro-1,4-phenylene)bis(6-(thiophen-2-yl)-6H-1,3-thiazin-2-amine) (B2) [21]

A mixture of chalcone (B) (0.384 gm ,0.001 mol) ,thiourea (0.152 gm,0.002 mol) were dissolved in(40 ml) absolute ethanol and sodium hydroxide (10 %) was refluxed 9 hrs, then pourd into 400ml cold water with continuous stirring for an hour and then kept in refrigerator for 24 hrs. The product was filtered ,washed and recrystallized by absolute ethanol and monitored by T.L.C (2ml methanol:4ml benzene).

4.Preparatio of (2Z,4E)-4-(2-chloro-4-((2Z)-2-(thiophen-2-yl)-1H-benzo[b][1,4]diazepin-4-yl)phenyl)-2-(thiophen-2-yl)-1H-benzo[b][1,4]diazepine[22]**.(B3)**

A mixture of chalcone (B) (0.192 gm ,0.0005 mol), o-phenylendiamine (0.108 gm,0.001mol) were dissolved in(50 ml) absolute ethanol and sodium hydroxide (10 %) was refluxed 7 hrs, then pourd into 400ml cold water with continuous stirring for an hour and then kept in refrigerator for 24 hrs. The product was filtered ,washed and recrystallized by absolute ethanol and monitored by T.L.C (2ml methanol:4ml benzene).

5.Preparation of 3,3'-(2-chloro-1,4-phenylene)bis(1-(2,4-dinitrophenyl)-5-(thiophen-2-yl)-4,5-dihydro-1H-pyrazole) [23]**. (B4)**

A mixture of chalcones (B) (0.384 gm,0.001 mol),2,4-dinitrophenylhydrazine (0.396 gm,0.002 mol) in absolute ethanol 40 ml refluxed for 10 hrs. Then the mixture was concentrated and allowed to cooled , the resulting solid was filtered and recrystallized from absolute ethanol and monitored by T.L.C (2ml methanol:4ml benzene).

6.Preparation of 3-(4-hydroxyphenyl)-1-(thiophen-2-yl)prop-2-en-1-one[19]**. (C)**

To 1-(thiophen-2-yl) ethanone (0.63 gm, 0.005 mole) a 4-hydroxybenzaldehyde (0.61gm, 0.005 mole) was added in ethanol absolute (30 ml) and Sodium hydroxide (10%). The mixture was stirred for 7 hrs at room temperature using magnetic stirrer, the reaction was cold. Filtered and neutralized with dil.HCl. The reaction was monitored by T.L.C (1ml methanol:4ml benzene)and the solvent was evaporated and the precipitation was recrystallized from absolute Ethanol to give 3-(4-hydroxyphenyl)-1-(thiophen-2-yl)prop-2-en-1-one.

7.Preparation of 4-(2-amino-6-(thiophen-2-yl)-6H-1,3-oxazin-4-yl)phenol[20]**. (C1)**

A mixture of chalcone (C) (0.115 gm ,0.0005 mol) , urea (0.03 gm,0.0005 mol) were dissolved in(30 ml) absolute ethanol and sodium hydroxide (10 %) was refluxed 10 hrs, then pourd into 400ml cold water with continuous stirring for an hour and then kept in refrigerator for 24 hrs. The product was filtered ,washed and recrystallized by absolute ethanol and monitored by T.L.C (2ml methanol:4ml benzene).

8.Preparation of4-(2-amino-4-(thiophen-2-yl)-6H-1,3-thiazin-6-yl)phenol[21]**. (C2)**

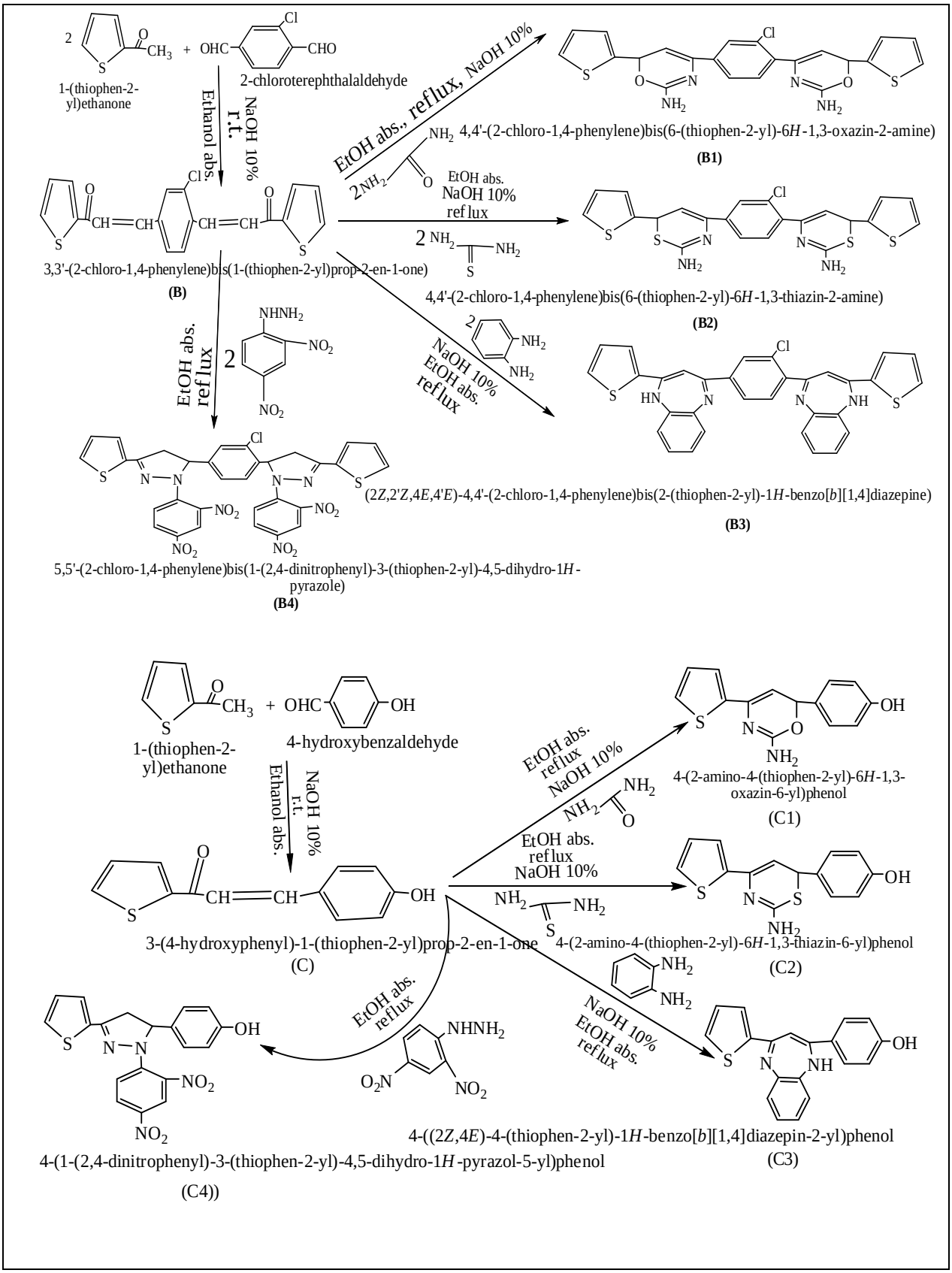
A mixture of chalcone (C) (0.23 gm ,0.001 mol) , thiourea (0.076 gm,0.001 mol) were dissolved in(40 ml) absolute ethanol and sodium hydroxide (10 %) was refluxed 10 hrs, then pourd into 400ml cold water with continuous stirring for an hour and then kept in refrigerator for 24 hrs. The product was filtered ,washed and recrystallized by absolute ethanol and monitored by T.L.C (2ml methanol:4ml benzene).

9.Preparation of 4-((2Z,4E)-2-(thiophen-2-yl)-1H-benzo[b][1,4]diazepin-4-yl)phenol[22].(C3)

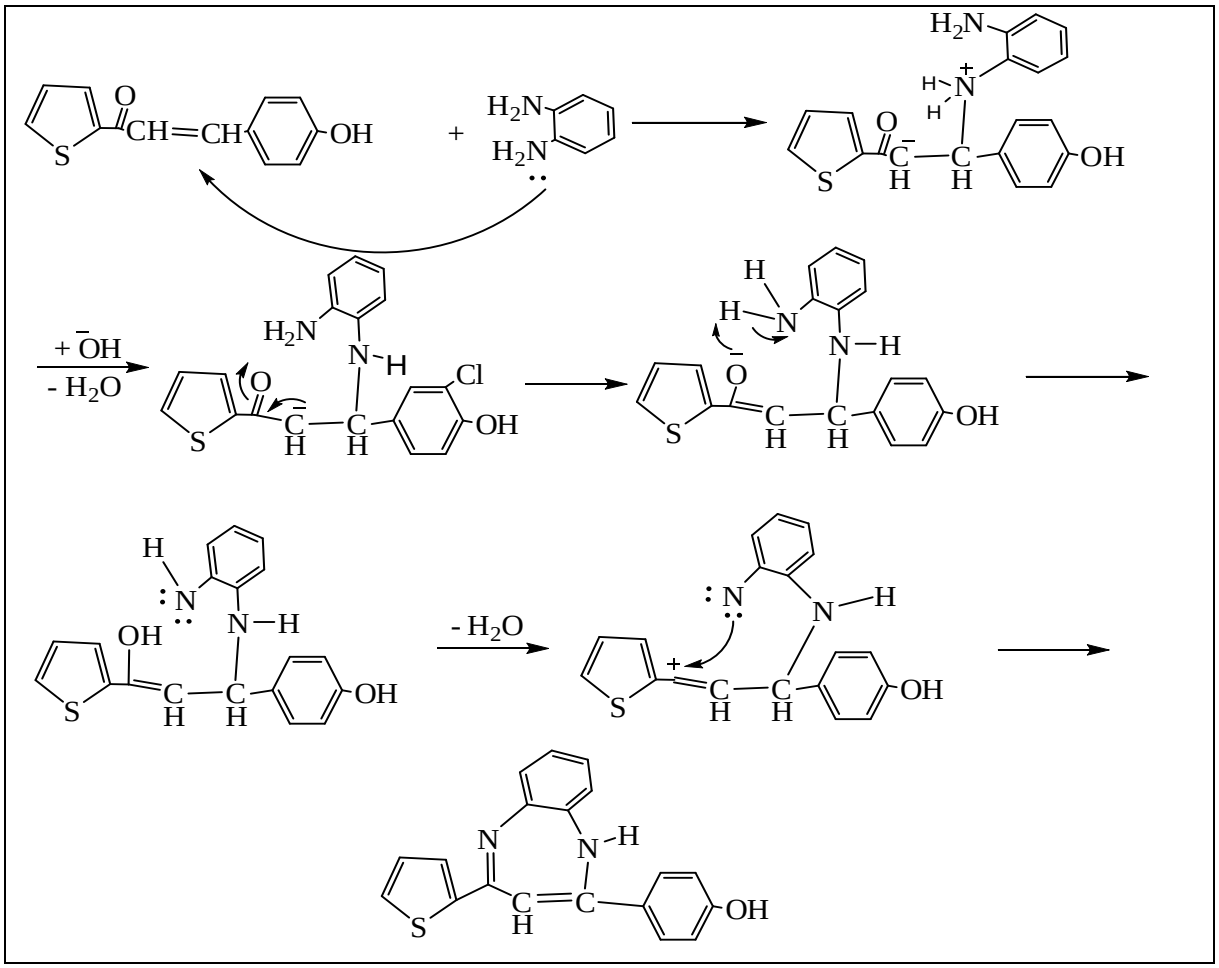
A mixture of chalcone (C) (0.115 gm ,0.0005 mol), o-phenylendiamine (0.054 gm,0.0005mol) were dissolved in(40 ml) absolute ethanol and sodium hydroxide (10 %) was refluxed 11 hrs, then pourd into 400ml cold water with continuous stirring for an hour and then kept in refrigerator for 24 hrs. The product was filtered ,washed and recrystallized by absolute ethanol and monitored by T.L.C (1ml methanol:4ml benzene).

10.Preparation of 4-(1-(2,4-dinitrophenyl)-5-(thiophen-2-yl)-4,5-dihydro-1H-pyrazol-3-yl)phenol[23]. (C4)

A mixture of chalcones (C) (0.115 gm,0.0005 mol), 2,4-dinitrophenylhydrazine (0.099 gm,0.0005 mol) in absolute ethanol 30 ml refluxed for 10 hrs. Then the mixture was concentrated and allowed to cooled , the resulting solid was filtered and recrystallized from absolute ethanol and monitored by T.L.C (1ml methanol:4ml benzene).



Scheme 1: Synthesis new compounds



Scheme 2: Mechanism of compound diazepine (C3)

Results And Discussion

The derivatives were prepared following the reaction sequences in **scheme 1**. All prepared derivatives (B- C4) have been characterized by spectroscopic method such as (FT-IR, ^{13}C-NMR&^{1}H-NMR) spectra. Reaction 1-(thiophen-yl) ethanone with 2-chloroterephthaldehyde to afford compound (B) indicated by appearance the carbonyl chalcone C=O stretching at 1660 cm^{-1} **Table 1**. ^{1}H-NMR (DMSO-*d6*) of compound (B): 6.44 (s,1H,CH ethylene), 8.13& 8.53 (protons of thiophene ring), 7.16-7.92 (protons of benzene ring) **Table 2**. ^{13}C-NMR (DMSO-*d6*) of compound (B): 186.85 (1C,C=O of chalcone), 148.16 (1C,CH=CH ethylene), 116.18 (1C,CH, ethylene), 116.86-136.05 (carbon of benzene and thiophene ring) **Table 2**. The chalcone (B) condensation with urea afford oxazine derivative (B1), FT-IR show the disappearance C=O chalcone and appearance NH_2 two bands symmetric and asymmetric at 3211 & 3348 cm^{-1}, C=N at 1624 cm^{-1} **Table 1**. The chalcone (B) condensation with thiourea afford thiazine derivative (B2), FT-IR show the disappearance C=O chalcone and appearance

NH_2 two bands symmetric and asymmetric at 3178 & 3278 cm^{-1} **Table 1**. ^{1}H-NMR (DMSO-*d6*) of compound (B2): 8.3 (s, 2H, NH_2), 4.2 (s, 1H,CH methine), 6.5 (s, 1H, CH=), 6.8- 7.7 (protons of thiophene & benzene ring) **Table 2**. ^{13}C-NMR (DMSO-*d6*) of compound (B2) : 165.89 (1C, 1- imine), 148.43 (1C, 1-ethylene), 139.28 (1C, 2-thiophene), 116.22-138.59 (carbon of benzene and thiophene ring) **Table 2**. The chalcone (B) condensation with o-phenylendiamine afford diazepine derivative (B3), FT-IR show disappearance of C=O chalcone and appearance NH band at 3381 cm^{-1} and C=N at 1624 cm^{-1} **Table 1**. ^{1}H-NMR (DMSO –*d6*) of compound (B3): 5.3 (s, 1H, NH arom.), 6.3(s, 1H, 1-ethylene), 6.4- 8.3 (protons of aromatic and thiophene ring) **Table 2**. The chalcone (B) reacts with 2,4-dinitrophenylhydrazine to afford pyrazole derivative (B4), FT-IR show the disappearance C=O chalcone and appearance the pyrazole ring N-N at 1282 cm^{-1} and band C=N at 1639 cm^{-1} **Table 1**. ^{1}H-NMR (DMSO-*d6*) of compound (B4): 3.5 (s, 2H, methylene), 3.9 (s, 1H, methine), 6.9-8.8 (protons of benzene and thiophene ring) **Table 2**. ^{13}C-NMR (DMSO-*d6*) of compound (B4): 48.39 (1C, aliph. methylene), 55.69 (1C,aliph.), 158.43 (1C, C=N, 1-imine), 107.98-146.57 (carbon of aromatic ring) **Table 2**. Reaction 1-(thiophen-yl) ethanone with 4-Hydroxybenzaldehyde to afford compound (C) indicated by appearance the carbonyl chalcone C=O stretching at 1670 cm^{-1} and OH at 3201 cm^{-1} **Table 1**. ^{1}H-NMR (DMSO-*d6*) of compound (C): 9.7 (s, 1H, OH arom.), 6.6 (s, 1H, 1-ethylene), 7.7 (s,1H, 1-ethylene), 6.9 7.4 (proton of benzene ring), 8.2 & 8.3 (protons of thiophene ring) **Table 2**. ^{13}C-NMR (DMSO-*d6*) of compound (C): 184.9 (1C, C=O chalcone), 162.8 (1C,C-OH), 149.9 (1C, =CH, 1-ethylene), 120.4 (1C, CH=, ethylene), 130.6- 132.5 (carbon of thiophene ring), 116.2- 127.8 (carbon of benzene ring) **Table 2**. The chalcone (C) condensation with urea afford oxazine derivative (C1), FT-IR show the disappearance carbonyl chalcone and appearance NH_2 two bands symmetric and asymmetric at 3315 & 3369 cm^{-1} and appearance OH at 3481 cm^{-1} **Table 1**. ^{1}H-NMR (DMSO-*d6*) of compound (C1): 9.7 (s, 1H, OH arom.), 5.1 (s, 2H, NH_2 amine), 5.6 (s, 1H,methine), 6.6 (s, 1H, 1-ethylene), 6.7 & 6.8 (protons of benzene ring), 6.9- 7.75 (protons of thiophene ring) **Table 2**. ^{13}C-NMR (DMSO-*d6*) of compound (C1): 165.2 (1C,C=N, 1-imine), 162.5 (1C, C-OH, 1-benzene), 78.1 (1C, C-O, aliph.), 146.5 (1C, 1-ethylene), 127.3 (1C, 1-ethylene), 116.3 & 128.7 (carbon of benzene ring), 132.4 & 134.2 (carbon of thiophene ring) **Table 2**. The chalcone (C) condensation with thiourea afford thiazine derivative (C2), FT-IR show the disappearance carbonyl chalcone and appearance NH_2 two bands symmetric and asymmetric at 3192 & 3360 cm^{-1} and appearance OH at 3406 cm^{-1} **Table 1**. The chalcone (C)

condensation with o-phenylendiamine afford diazepine derivative (C3), FT-IR show disappearance of C=O chalcone and appearance of NH bands at 3196 cm^{-1} , C=N at 1629 cm^{-1} and OH at 3383 cm^{-1} **Table 1**. ^{1}H-NMR (DMSO-*d6*) of compound (C3): 9.6 (s, 1H, OH arom.), 5.4 (s, 1H, NH arom.), 6.5 (s, 1H, 1-ethylene), 6.7- 7.2 (proton of benzene ring), 7.7-8.5 (protons of thiophene ring) **Table 2**. ^{13}C-NMR (DMSO-*d6*) of compound (C3): 162.9 (1C,C=N, 1-imine), 155.3 (1C, C-OH, 1-benzene), 141.1 (1C, 1-ethylene), 72.3 (1C, 1-ethylene), 113.9- 125.8 (carbon of benzene ring), 130.9-140.2 (carbon of thiophene ring) **Table 2**. The chalcone (C) reacts with 2,4- dinitrophenylhydrazine to afford pyrazole derivative (C4), FT-IR show the disappearance C=O chalcone and appearance the pyrazole ring N-N at 1273 cm^{-1} , C=N band at 1616 cm^{-1},OH band at 3450 cm^{-1} **Table 1**. ^{1}H-NMR (DMSO-*d6*) of compound (C4): 10.01 (s, 1H, OH arom.), 3.4 & 3.7 (s, 2H, methylene), 5.09 (s, 1H,methine), 8.3- 8.8 (protons of benzene ring contain NO_2), 6.9 &7.6 (protons of benzene), 8.00 & 8.03 (protons of thiophene ring) **Table 2**.

Table 1: Physical Properties and Spectral Data of Compounds

NO.	Molecular formula	Molecular weight	Yield %	M.P C°	R. f	FT-IR(KBr) *ν* cm^{-1}
B	$C_{20}H_{13}ClO_2S_2$	385	79	220-222	0.69	3007 C-H arom., 2933 C-H aliph., 1660 C=O, 1612 C=C alkene, 1577 C=C arom., 759 C-Cl, 659 C-S
B1	$C_{22}H_{17}ClN_4O_2S_2$	469	69	210-212	0.73	3034 C-H arom., 2981 C-H aliph., 3211 & 3348 NH_2, 1624 C=N, 1591 C=C alkene, 1480 C=C arom., 1249 C-O, 1141 C-N, 759 C-Cl, 667 C-S
B2	$C_{22}H_{17}ClN_4S_4$	501	78	212-214	0.75	3059 C-H arom., 2981 C-H aliph., 3178 & 3278 NH_2, 1616 C=N, 1581 C=C alkene, 1471 C=C arom., 1083 C-N, 731 C-Cl, 632 C-S
B3	$C_{32}H_{21}ClN_4S_2$	561.12	68	190-192	0.62	3057 C-H arom., 2978 C-H aliph., 3381 NH, 1624 C=N, 1583 C=C alkene, 1571 C=C arom., 1141 C-N, 756 C-Cl, 689 C-S
B4	$C_{32}H_{21}ClN_8O_8S_2$	745	66	170-172	0.71	3007 C-H arom., 2970 C-H aliph., 1639 C=N, 1575 C=C arom., 1516 & 1323 NO_2, 1282 N-N, 1222 C-N, 744 C-Cl, 632 C-S

C	$C_{13}H_{10}O_2S$	230	80	205- 207	0.75	3059 C-H arom., 2970 C-H aliph., 3201 OH, 1670 C=O, 1597 C=C alkene, 1560 C=C arom., 1288 C-O, 669 C-S
C1	$C_{14}H_{12}N_2O_2S$	272	85	140- 142	0.66	3034 C-H arom., 2920 C-H aliph., 3481 OH, 3315 & 3369 NH_2, 1600 C=N, 1585 C=C alkene, 1516 C=C arom., 1230 C-O cyclic, 1203 C-OH, 1139 C-N, 651 C-S
C2	$C_{14}H_{12}N_2O_2S_2$	288	69	202- 204	0.77	3084 C-H arom., 2929 C-H aliph., 3406 OH, 3192 & 3360 NH_2, 1622 C=N, 1589 C=C alkene, 1546 C=C arom., 1246 C-OH, 1157 C-N, 671 C-S
C3	$C_{14}H_{14}N_2OS$	318	76	175- 177	0.73	3057 C-H arom., 3034 C-H aliph., 3383 OH, 3196 NH, 1629 C=N, 1608 C=C alkene, 1591 C=C arom., 1224 C-OH, 1178 C-N, 614 C-S
C4	$C_{19}H_{14}N_4O_5S$	410	64	180- 182	0.62	3007 C-H arom., 2920 C-H aliph., 3450 OH, 1616 C=N, 1585 C=C arom., 1330 & 1510 NO_2, 1273 N-N, 1305 C-OH, 1132 C-N, 638 C-S

Table 2: Chemical Shift ^{1}H-NMR & ^{13}C-NMR Spectra

NO.	^{1}H- NMR(DMSO-*d6*) δ ppm	^{13}C- NMR(DMSO-*d6*) δ ppm
B	6.44 (s,1H,CH ethylene), 8.13& 8.53 (protons of thiophene ring), 7.16-7.92 (protons of benzene ring)	186.85 (1C,C=O of chalcone), 148.16 (1C,CH=CH ethylene), 116.18 (1C,CH, ethylene), 116.86-136.05 (carbon of benzene and thiophene ring)
B2	8.3 (s, 2H, NH2), 4.2 (s, 1H,CH methine), 6.5 (s, 1H, CH=), 6.8- 7.7 (protons of thiophene & benzene ring)	165.89 (1C, 1- imine), 148.43 (1C,1-ethylene), 139.28 (1C, 2-thiophene), 116.22- 138.59 (carbon of benzene and thiophene ring)
B3	5.3(s,1H, NH arom.), 6.3(s, 1H, 1-ethylene), 6.4-8.3 (protons of aromatic and thiophene ring)	----------
B4	3.5 (s, 2H, methylene), 3.9 (s, 1H, methine), 6.9-8.8 (protons of benzene and thiophene ring)	48.39 (1C, aliph. methylene), 55.69 (1C,aliph.), 158.43 (1C, C=N, 1-imine), 107.98- 146.57 (carbon of aromatic ring)
C	9.7 (s, 1H, OH arom.), 6.6 (s, 1H, 1-ethylene), 7.7 (s,1H, 1-ethylene), 6.9 7.4 (protons of benzene ring), 8.2 & 8.3 (protons of thiophene ring)	184.9 (1C, C=O chalcone), 162.8 (1C,C-OH), 149.9 (1C, =CH, 1-ethylene), 120.4 (1C, CH=, 1ethylene), 130.6- 132.5 (carbon of thiophene ring), 116.2- 127.8 (carbon of benzene ring)

C1	9.7 (s, 1H, OH arom.), 5.1 (s, 2H, NH2 amine), 5.6 (s, 1H,methine), 6.6 (s, 1H, 1-ethylene), 6.7 & 6.8 (protons of benzene ring), 6.9- 7.75 (protons of thiophene ring)	165.2 (1C,C=N, 1-imine), 162.5 (1C, C-OH, 1-benzene), 78.1 (1C, C-O, aliph.), 146.5 (1C, 1-ethylene), 127.3 (1C, 1-ethylene), 116.3 & 128.7 (carbon of benzene ring), 132.4 & 134.2 (carbon of thiophene ring)
C3	9.6 (s, 1H, OH arom.), 5.4 (s, 1H, NH arom.), 6.5 (s, 1H, 1-ethylene), 6.7- 7.2 (protons of benzene ring), 7.7- 8.5 (protons of thiophene ring)	162.9 (1C,C=N, 1-imine), 155.3 (1C, C-OH, 1-benzene), 141.1 (1C, 1-ethylene), 72.3 (1C, 1-ethylene), 113.9- 125.8 (carbon of benzene ring), 130.9-140.2 (carbon of thiophene ring)
C4	10.01 (s, 1H, OH arom.), 3.4 & 3.7 (s, 2H, methylene), 5.09 (s, 1H,methine), 8.3- 8.8 (protons of benzene ring contain NO2), 6.9 &7.6 (protons of benzene), 8.00 & 8.0 (protons of thiophene ring)	----------

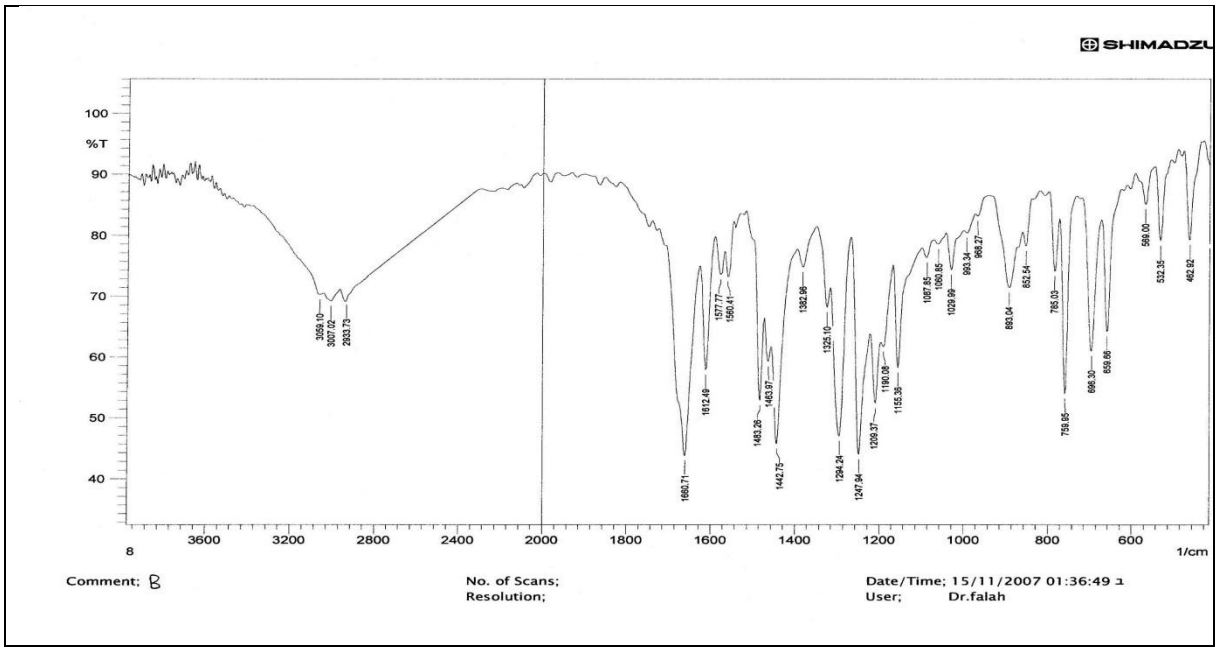


Fig. 1: FT-IR Spectrum of compound B

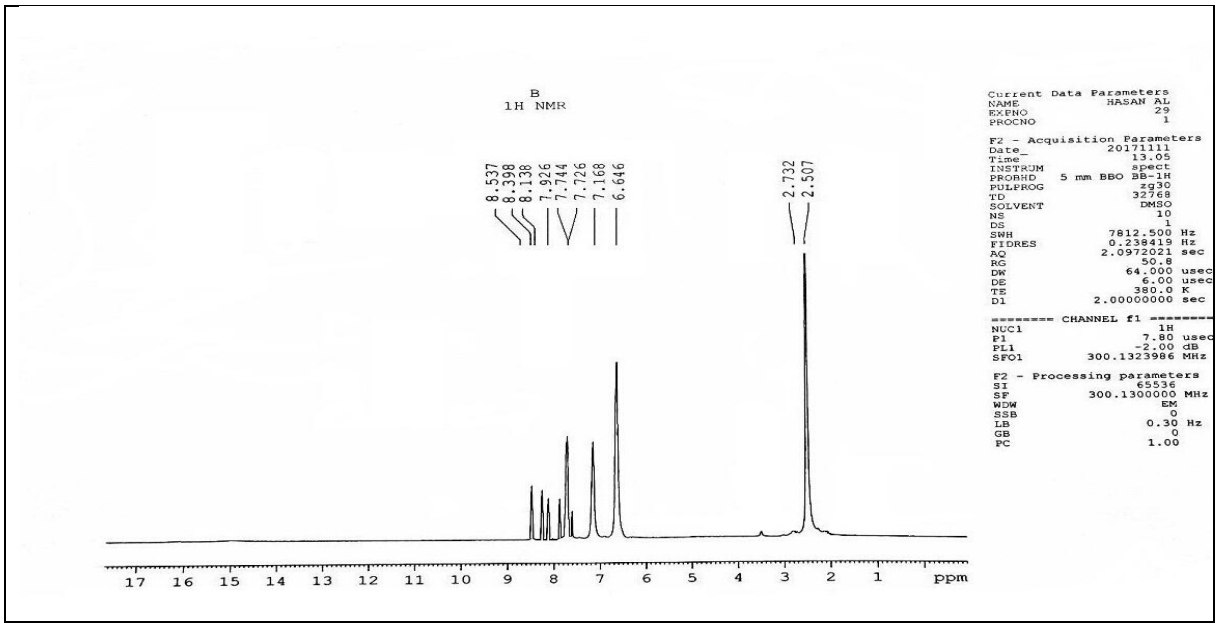


Fig. 2: ^{1}H-NMR Spectrum of compound B

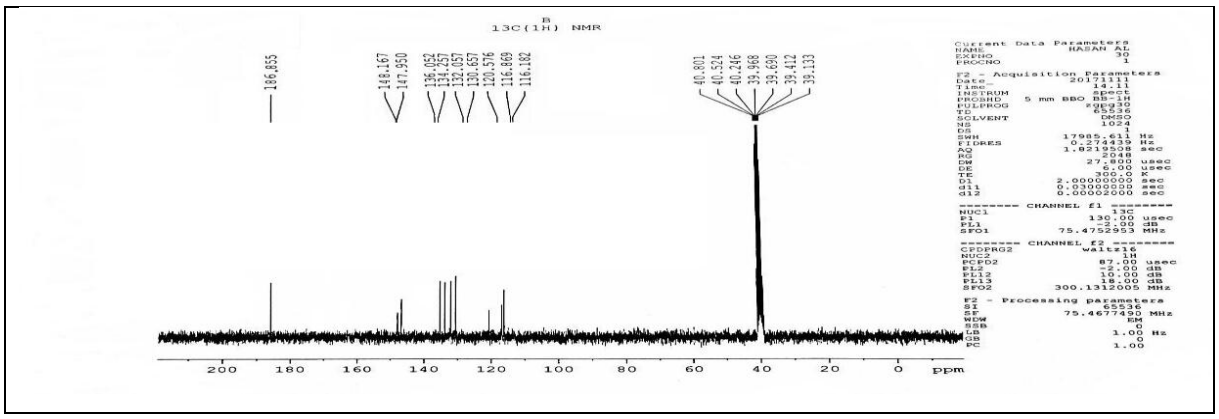


Fig.3: ^{13}C-NMR Spectrum of compound B

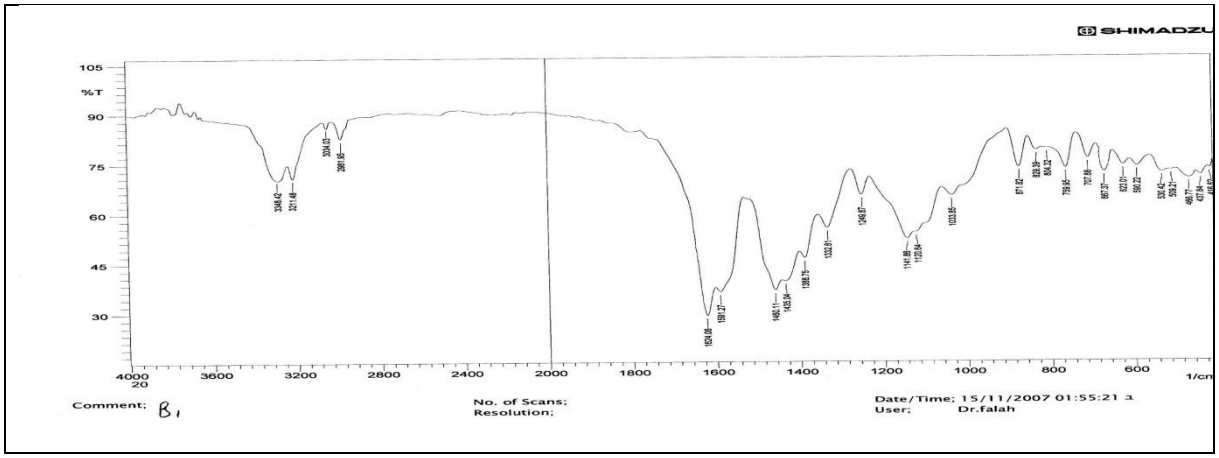


Fig. 4: FT-IR Spectrum of compound B1

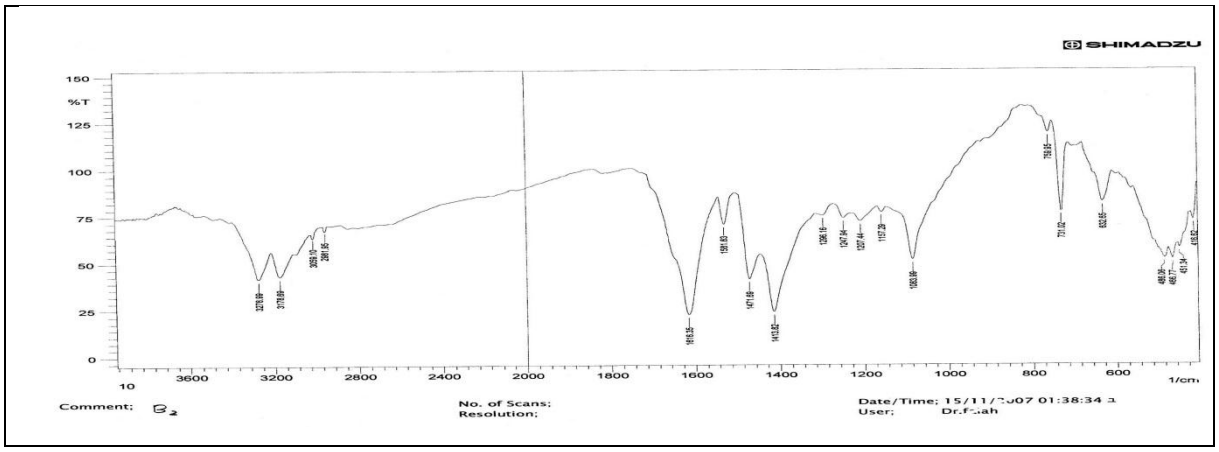


Fig. 5: FT-IR Spectrum of compound B2

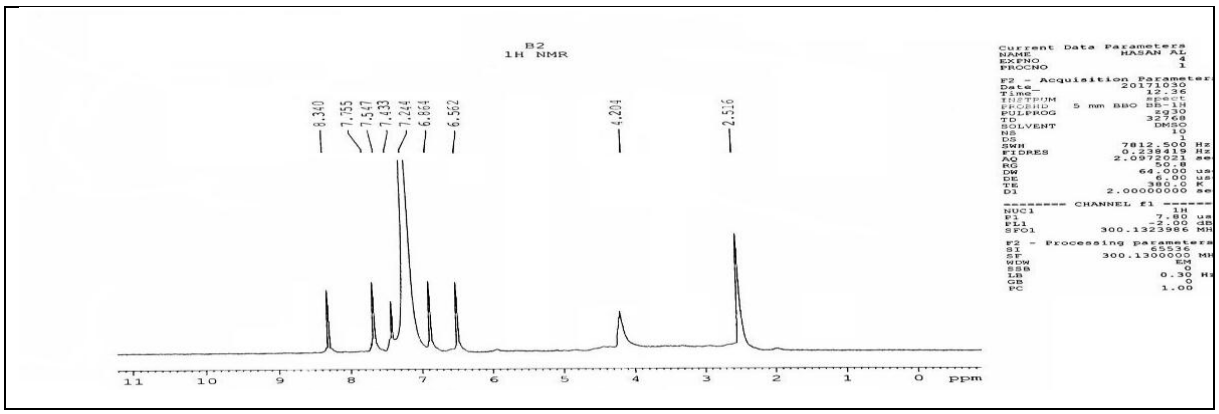


Fig. 6: ^{1}H-NMR Spectrum of compound B2

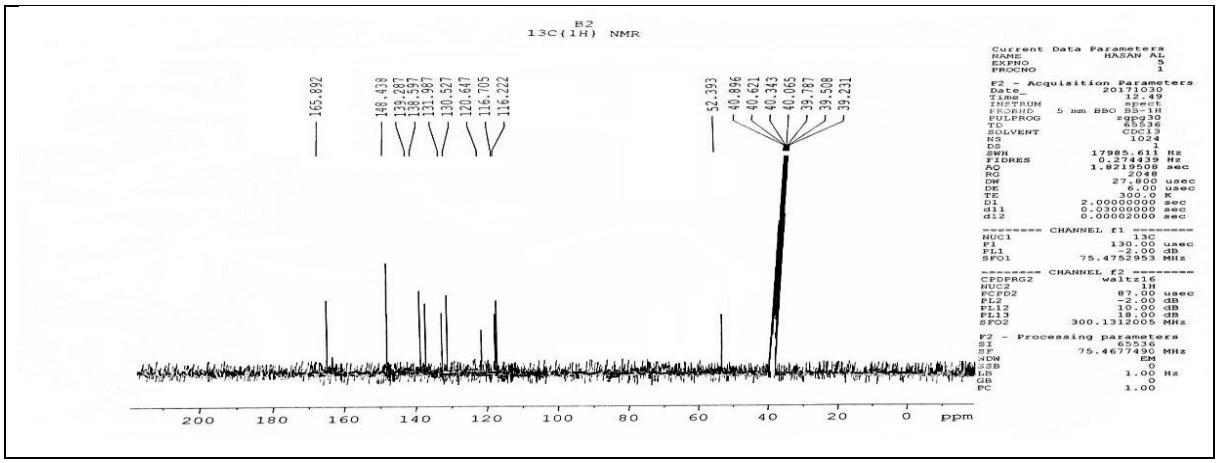


Fig.7: ^{13}C-NMR Spectrum of compound B2

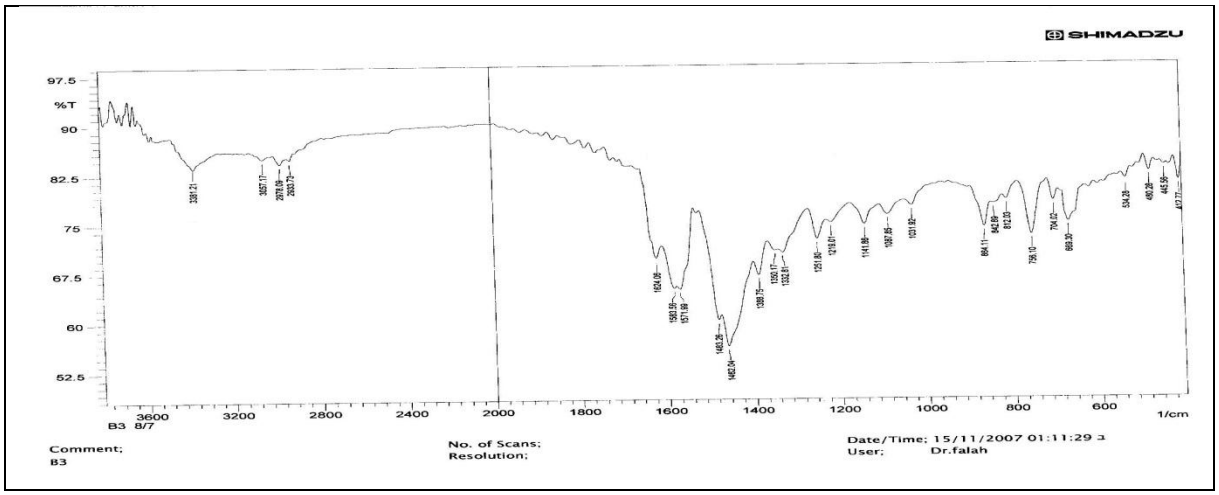


Fig. 8: FT-IR Spectrum of compound B3

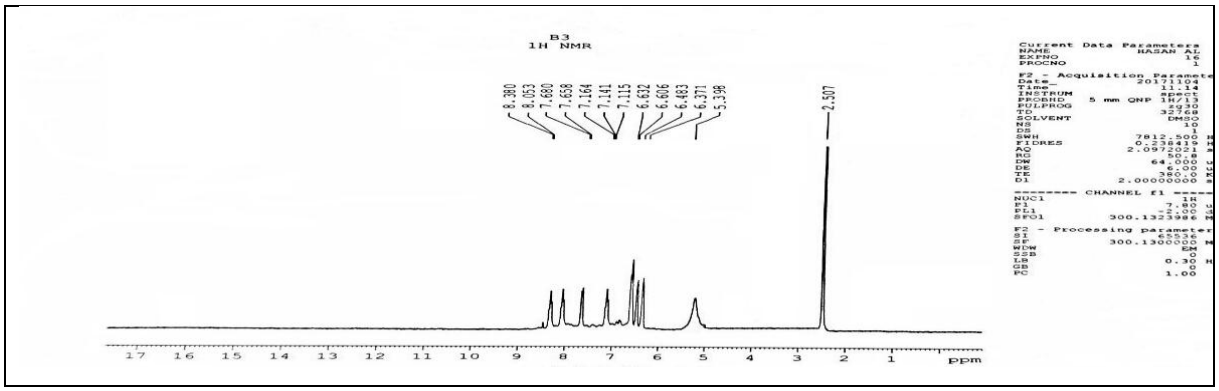


Fig. 9: ^{1}H-NMR Spectrum of compound B3

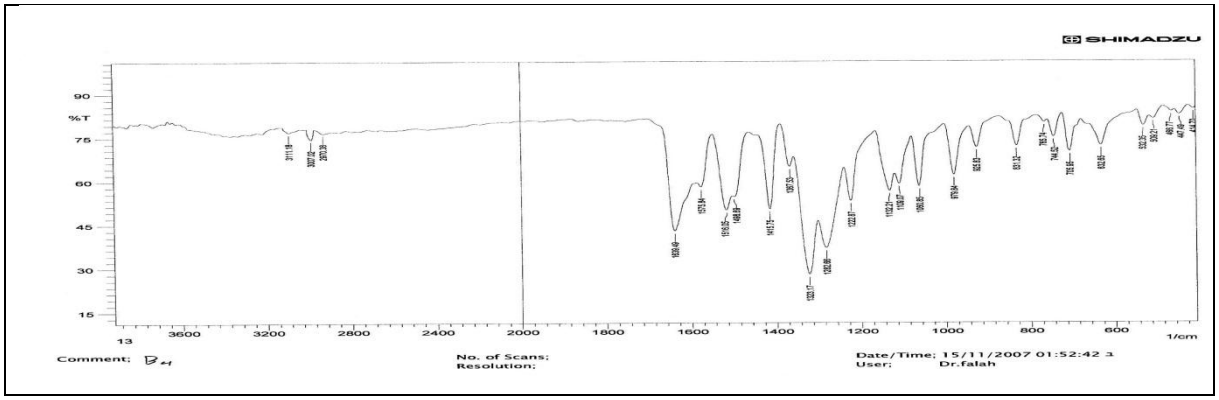


Fig. 10: FT-IR Spectrum of compound B4

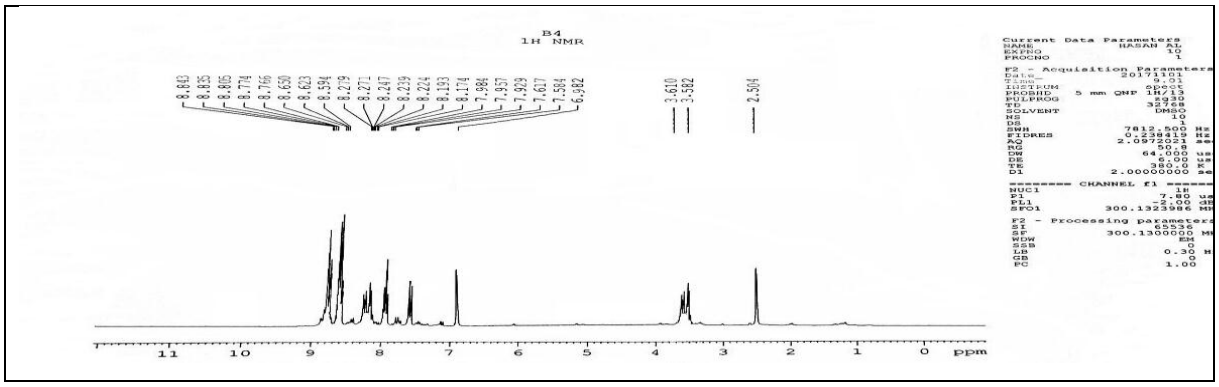


Fig. 11: ^{1}H-NMR Spectrum of compound B4

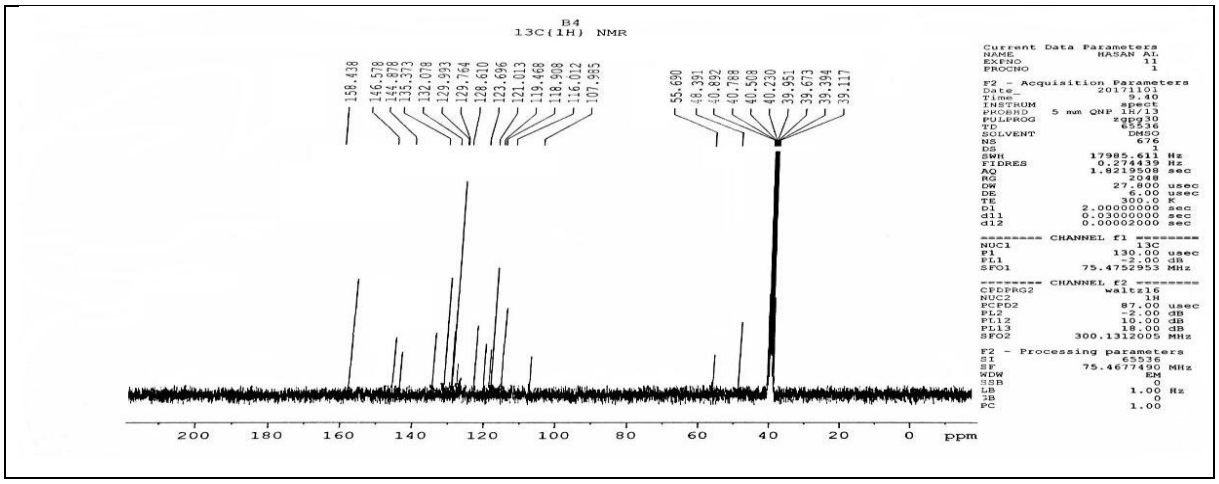


Fig. 12: ^{13}C-NMR Spectrum of compound B4

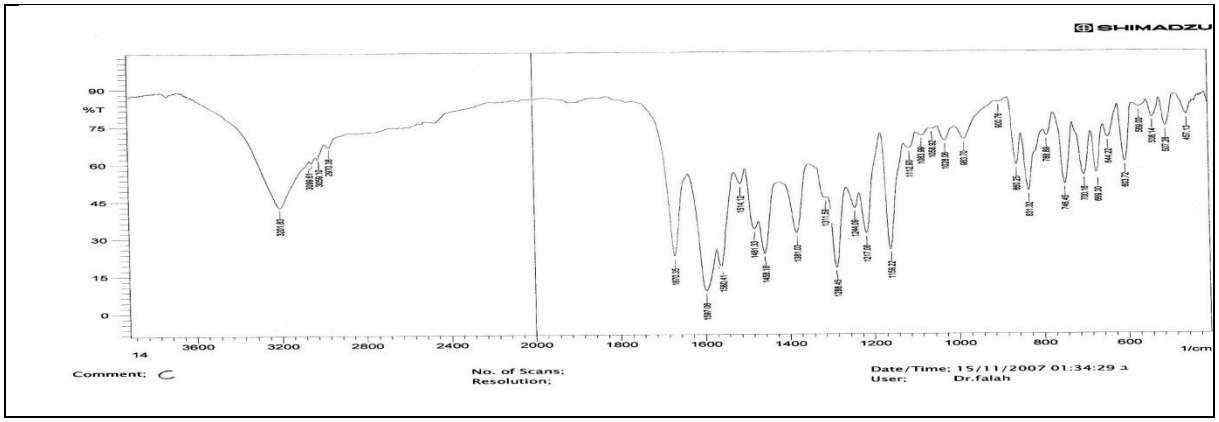


Fig. 13: FT-IR Spectrum of compound C

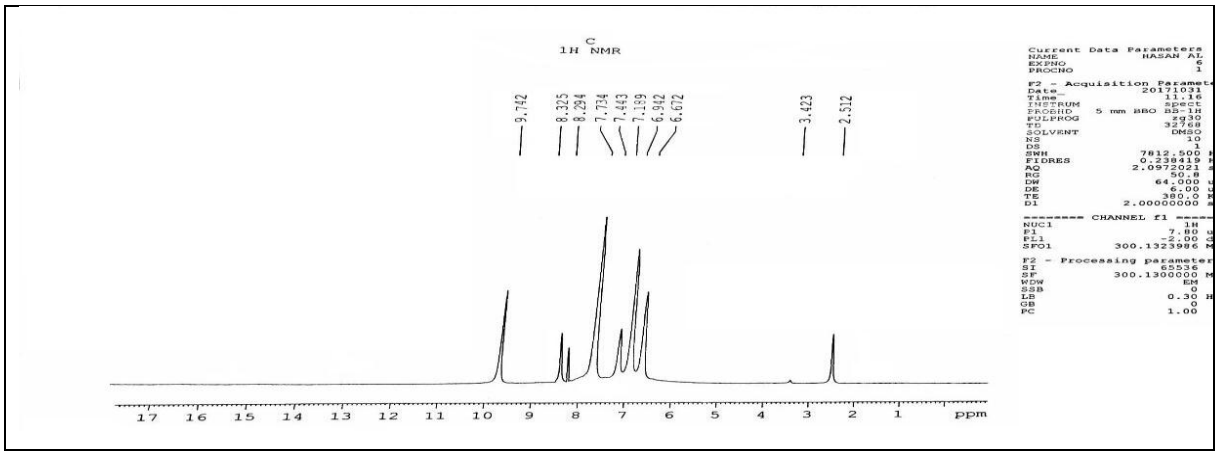


Fig. 14: ^{1}H-NMR Spectrum of compound C

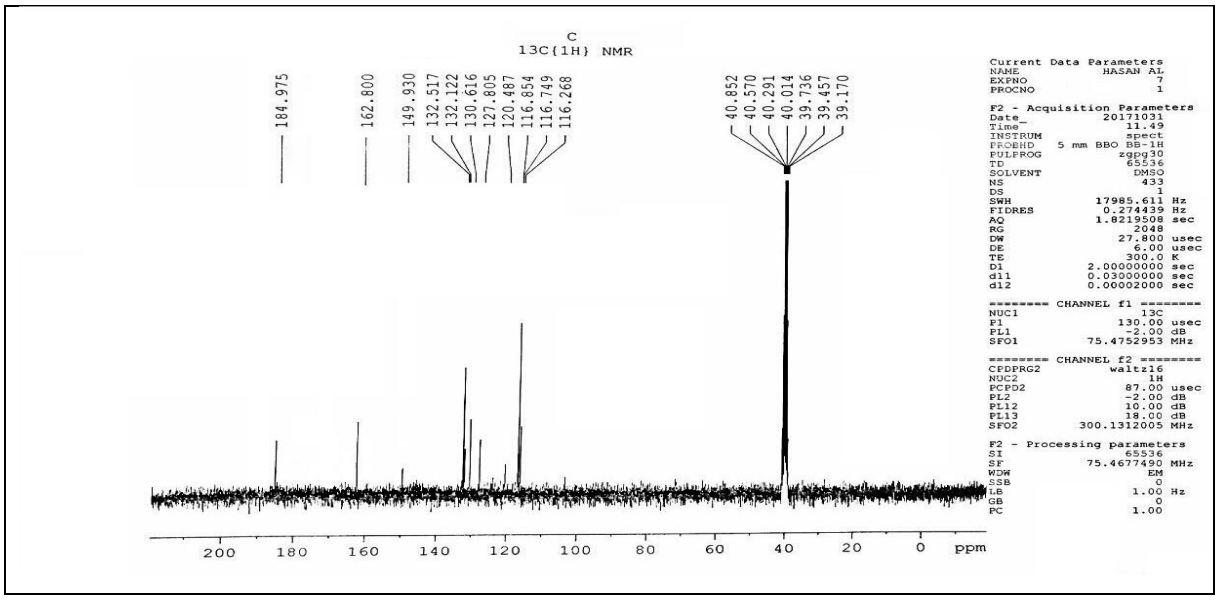


Fig. 15: ^{13}C-NMR Spectrum of compound C

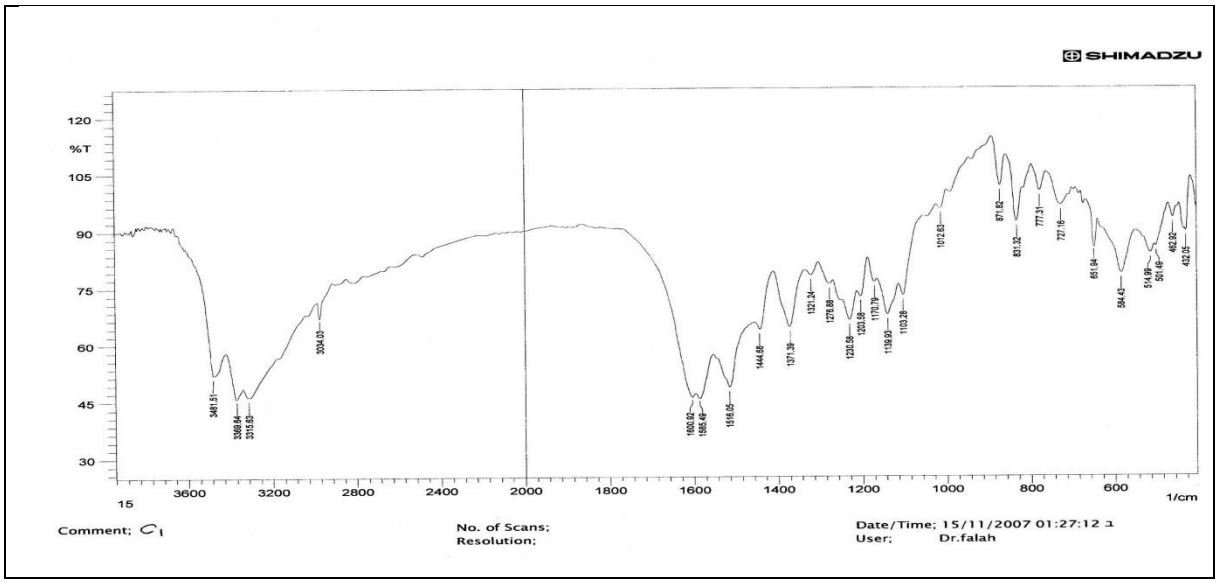


Fig. 16: FT-IR Spectrum of compound C1

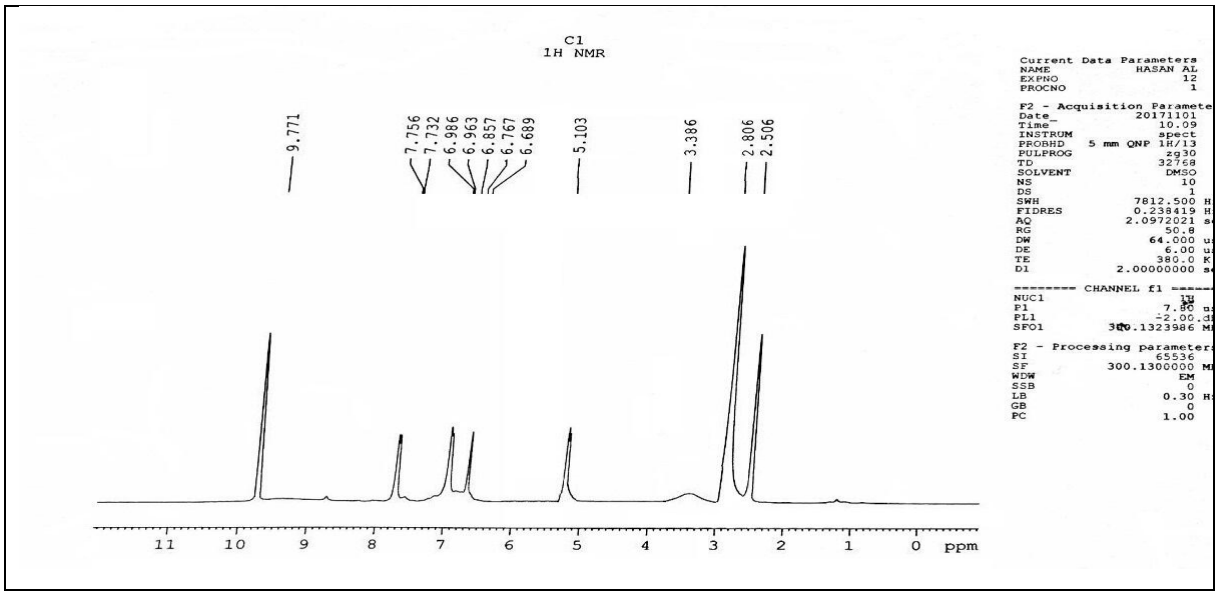


Fig. 17: ^{1}H-NMR Spectrum of compound C1

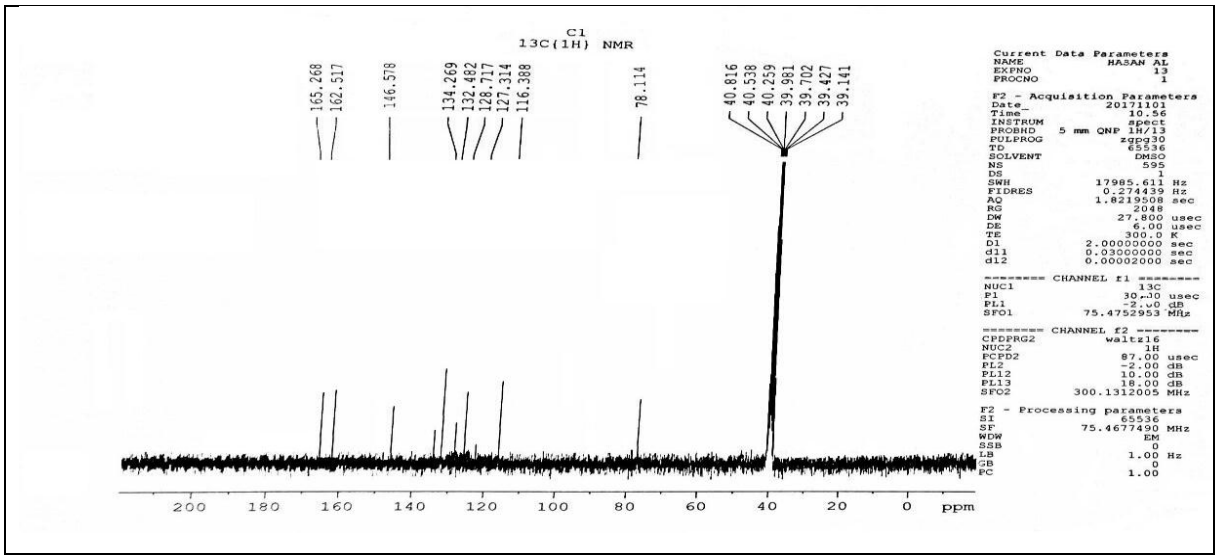


Fig. 18: ^{13}C-NMR Spectrum of compound C1

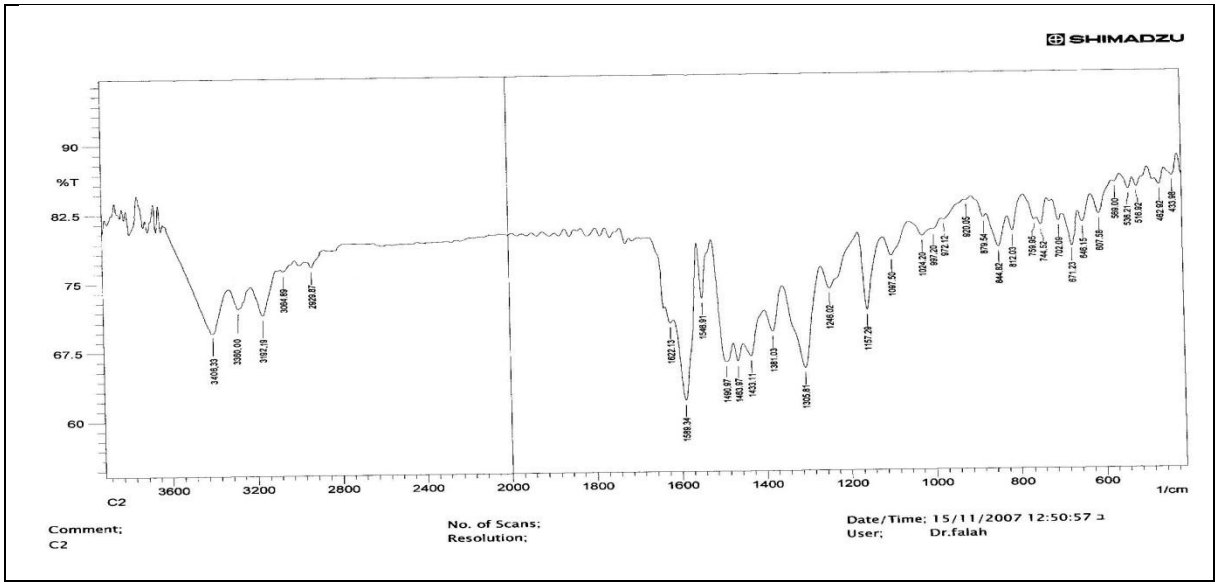


Fig. 19: FT-IR Spectrum of compound C2

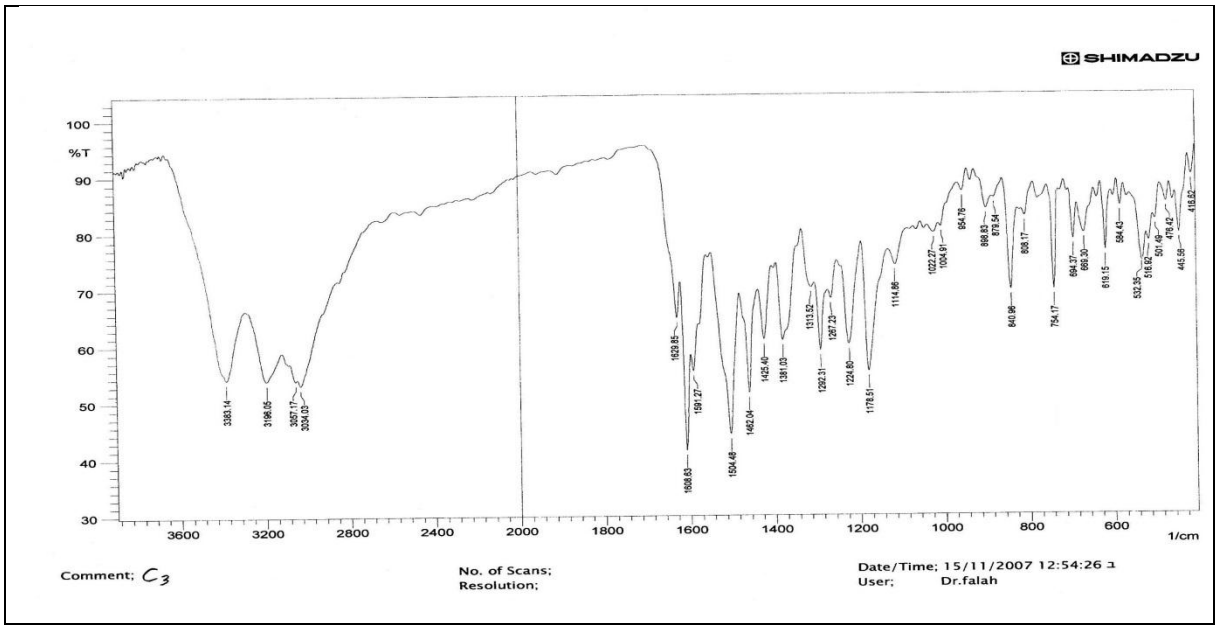


Fig. 20: FT-IR Spectrum of compound C3

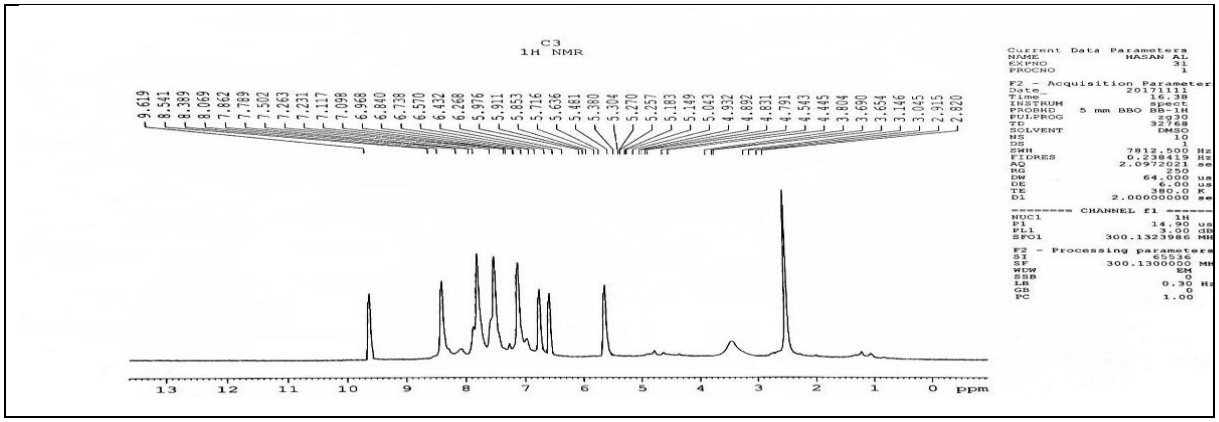


Fig. 21: ^{1}H-NMR Spectrum of compound C3

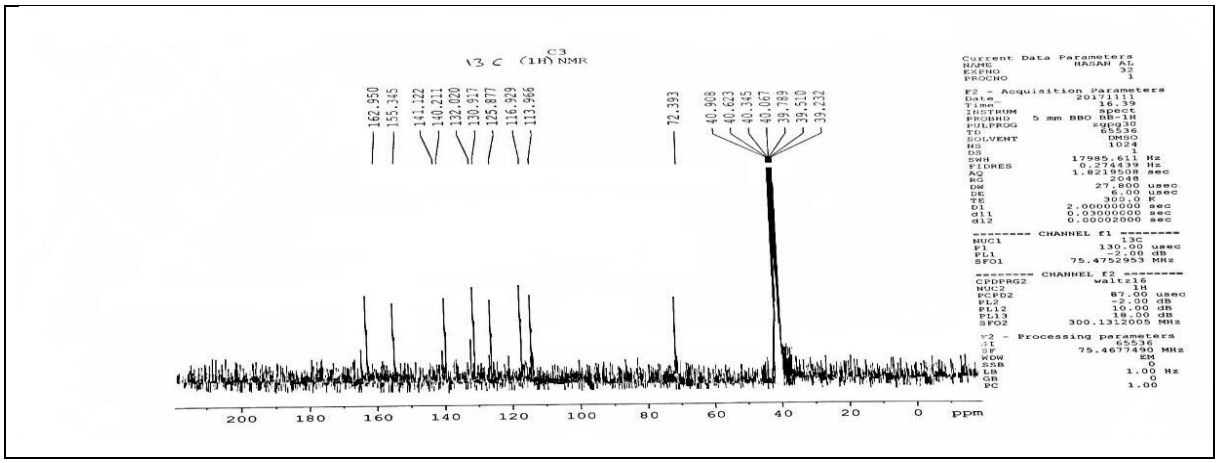


Fig. 22: ^{13}C-NMR Spectrum of compound C3

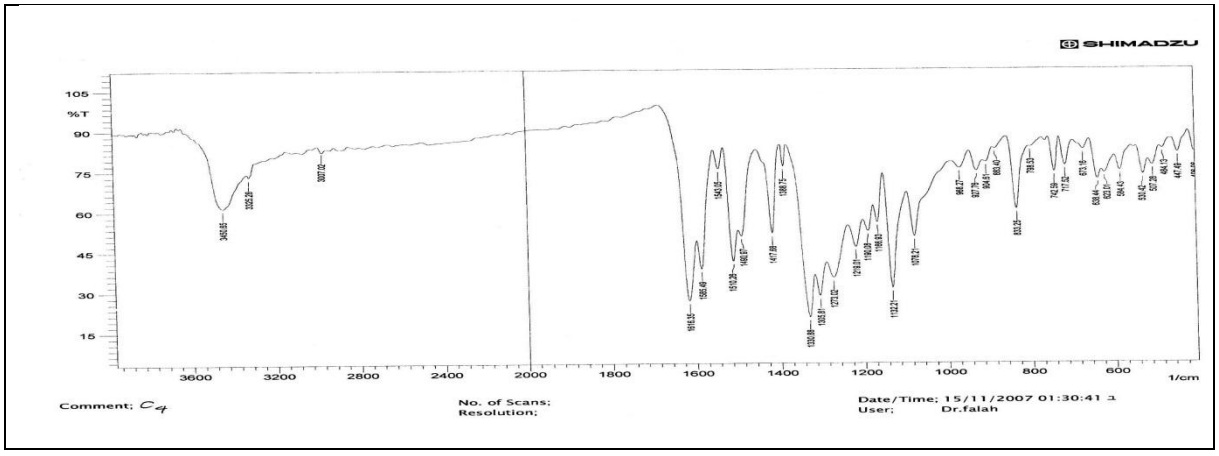


Fig. 23: FT-IR Spectrum of compound C4

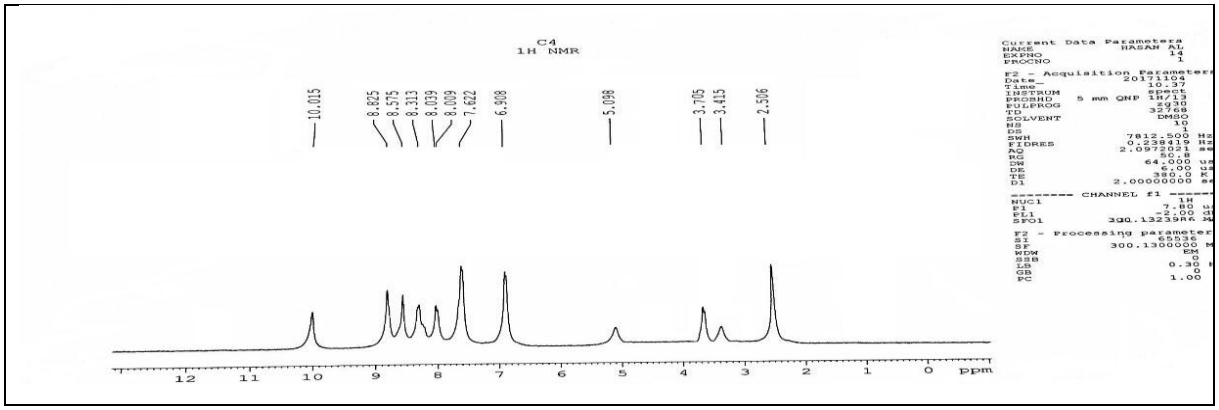


Fig. 24: ^{1}H-NMR Spectrum of compound C4

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