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A convenient, rapid and eco-friendly synthesis of Chalcones 3-(Substitutedphenyl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide

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ABSTRACT

In this work, an attempt was made to synthesize chalcones 3-(Substitutedphenyl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide by condensation of Substitutedbenzaldehyde with *N*-(4*H*-1,2,4-triazol-4-yl)acetamide under basic conditions by using both conventional and Microwave synthesis methods. A simple condensation reaction of substiutedbenzaldehyde and *N*-(4*H*-1,2,4-triazol-4-yl)acetamide using Sodium hydroxide as a base was carried out for the study. The synthesized chalcone derivatives were characterized for FTIR, ^{1}H NMR, ^{13}C NMR and Mass spectral analysis. It was observed that complete conversion to chalcone occurred in 10-12 min by Microwave synthesis method and in 10 h by conventional method. Finally, it has been observed that chalcone synthesis using Microwave synthesis method is a Convenient, Rapid and Eco-Friendly Synthesis over conventional method. The reaction time has been brought down from hours to seconds with improved yield as compared to conventional heating.

Introduction

In the last few years Microwave-assisted Organic Synthesis (MAOS) has gained popularity as a non-conventional technique for rapid organic synthesis [1] and many researchers have described accelerated organic reactions, and a large number of papers have appeared proving the synthetic utility of Microwave in routine organic synthesis [2,3]. It is convenient, rapid, economical and eco-friendly and is believed to be a step towards green chemistry.

Chalcones come under an aromatic ketone that forms the central core for a variety of important biological compounds. Chalcone derivatives are very versatile as physiologically active compounds and substrates for the evaluation of various organic syntheses. Chalcone and their derivatives are of high interest materials due to their antioxidant, antibacterial, antifungal, antitumor and anti-inflammatory properties [4–7]. Chalcones are valuable intermediates in the synthesis of many active pharmaceutical drugs like biosynthesis of flavonoids [8] and Auwers synthesis of flavones [9]. Having such varied pharmacological activities, these molecules have attracted medicinal chemists and therefore several strategies have been developed to synthesize them.

Claisen–Schmidt condensation between acetophenone and benzaldehyde gives chalcone [7]. This reaction is catalyzed by acids and bases under homogeneous or heterogeneous conditions. Several researchers have reported the synthesis of chalcone by using different catalysts like zinc oxide [10], organolithium [11], KF–Al_2O_3 [12], modified phosphates [13], zeolites and hydrotalcites [14] to get a better specification of chalcone products with low by-products and higher yield. The preparation of these catalysts is again a difficult task and the cost involved to synthesize catalysts is also higher. Catalyst recovery and waste disposal problems are again creating environmental issues. Researchers have published several reports on use of Microwaves as a process intensification tool for numerous catalytic reactions as well as in homogeneous and in heterogeneous reactions and it has proved to be a clean tool for improving yields and decreasing reaction time.

The chemistry of 1,2,4-triazoles has also received considerable attention owing to their synthetic and effective biological importance. 1,2,4-Triazole moieties have been incorporated into a wide variety of therapeutically interesting drug candidates including anti-inflammatories, CNS stimulants, sedatives, antianxiety compounds, antimicrobial agents [15-17] and antimycoticones such as fluconazole, intraconazole, voriconazole [18,19]. There are marketed drugs containing the 1,2,4-triazole group, e.g.: Triazolam [20], Alprazolam [21], Etizolam [22] and Furacylin [23]. In addition to these important biological applications, 1,2,4-triazoles are also of great utility in preparative organic chemistry, for example in the presence of various reagents, undergo different types of reactions to yield other heterocyclic compounds.

Our literature survey revealed that Chalcone are yet to be explored with 1,2,4-triazole ring system. Herein we report the Chalcone derivatives by synthesizing a series of ten molecules (3a–j) using both Microwave and conventional synthesis method. The effect of microwaves on % yield and reaction time of synthesized chalcones has been studied to understand the role of microwaves (microwave energy) in the synthesis of chalcones. In this study, only the HY-ALI feature associated with 'B' ring of the Chalcone moiety was changed by keeping the basic skeleton intact.

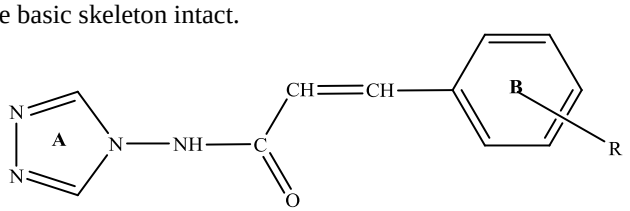

Figure 1. Schematic diagram of Chalcones 3a-j

Results and Discussion

The fundamental mechanism of microwave heating involves agitation of polar molecules or ions that oscillate under the

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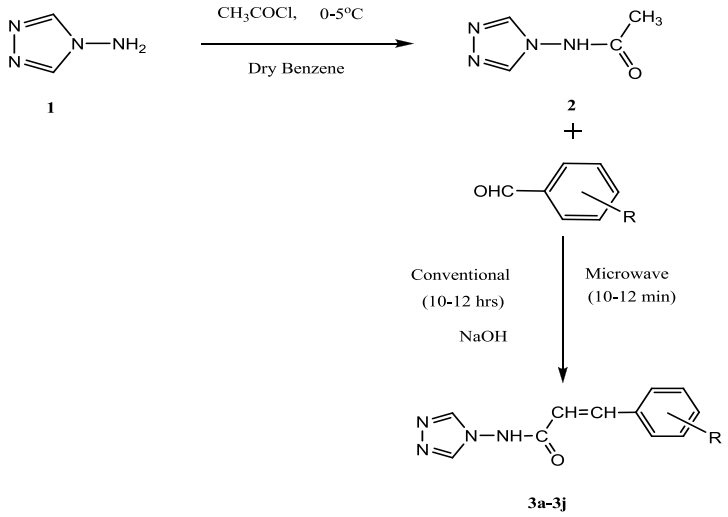
effect of an oscillating electric or magnetic field. In the presence of an oscillating field, particles try to orient themselves or be in phase with the field. However, the motion of these particles is restricted by resisting forces (inter-particle interaction and electric resistance), which restrict the motion of particles and generate random motion, producing heat. Since the response of various materials to microwave radiation is diverse, not all materials are amenable to microwave heating.

4-amino-4*H*-1,2,4-triazole (1) on acetylation with acetyl chloride and dry benzene gives N-(4*H*-1,2,4-triazol-4-yl)acetamide (2). This compound on reaction with substituted aldehydes in absolute ethanol and 2% NaOH afforded chalcones (3a-j). Formation of chalcones (3a-j) was evidenced by appearance of a ^{1}H NMR signal at δ 7.40-7.38 and δ 7.99-7.97 ppm, due to α and β carbon of chalcones (3a-j), and the observation of a IR band at 1691 cm^{-1} due to Carbonyl group confirmed the formation of chalcone. Similarly, in ^{13}C NMR spectra, signals were observed at δ 166.13 ppm, due to carbonyl group, at δ 138.48 ppm, due to β carbon and at δ 128.13 due to α carbon of Chalcones (3a-j). All the reactions under MWI were completed within 10.0-12.0 min., whereas similar reactions under conventional heating at refluxed temperature gave poor yields with comparatively longer reaction time periods (Table 2), demonstrating that the effect of microwave irradiation is not purely thermal. In fact, the microwave irradiation facilitates the polarization of the molecules under irradiation causing rapid reactions to occur. We are also studied for their optimization of results in terms of yield at various watts/powers and reaction time for microwave methods of chalcone derivatives and the results summarized in Tables 3 and 4. High yields of compounds 3a-j were obtained at 455 watt for 10.0 min under microwave irradiation.



3	R	3	R
a	-4-Cl	f	-2,4-Cl
b	-4-N(CH$_3$)$_2$	g	-3,4,5-OCH$_3$
c	-Naphthayl	h	-4-OCH$_3$
d	-3-OCH$_3$	i	-3-NO$_2$
e	-2-OH	j	-Br

Scheme 1. Synthesis of Chalcones 3a-j

Experimental Section

Melting points were determined in open capillary tubes and are uncorrected. All the chemicals and solvents used were of laboratory grade and solvents were purified by suitable methods. IR (Infrared spectrum) (KBr, cm^{-1}) were recorded on a Shimadzu-8400 FT-IR spectrometer using KBr disc, ^{1}H-NMR and ^{13}C-NMR spectra were recorded on a Brucker Avance II 400 NMR spectrometer using TMS as an internal standard (chemical shift in δ,ppm) in $CDCl_3$. The mass spectrum was recorded on Hewlett-Packard 5989, a Quadrapole Mass Spectrum. The homogeneity of the products was checked by TLC using Silica Gel GF_{254} (E.Merck) and the eluent system was a mixture of Acetone - Toluene in 2:8 proportions. The microwave assisted reactions are conducted in a "Scientific Microwave System"(Catalyst^{TM} Systems). whereby microwaves are generated by magnetron at a frequency of 2450 MHz having an output energy range of 140 to 700 Watts, P/L (1-10), % Power (20-100) and with an individual sensor for temperature control (fiber optic is used as an individual sensor for temperature control) with attachment of reflux condenser with constant stirring (thus avoiding the risk of high pressure development) and synthesis on preparative scales.

Figure 2. "Scientific Microwave System" (Catalyst^{TM} Systems)

General procedure for the preparation of *N*-(4*H*-1,2,4-triazol-4-yl)acetamide (2).

To a solution of 4-amino-4*H*-1,2,4-triazole **(1)** (0.01 mol) in dry benzene (50 mL), acetyl chloride (0.01 mol) was added drop by drop at 0–5 °C. The reaction mixture was stirred for 1 h and kept overnight. The reaction mixture was distilled off and then poured onto ice. The solid thus obtained was recrystallized from suitable solvent. Physical and analytical data are given in tables I.

General procedure for the preparation of 3-(substitutedphenyl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide (3a–3j).

a) Conventional method

A solution of *N*-(4*H*-1,2,4-triazol-4-yl)acetamide **(2)** (0.01 mol) in absolute ethanol (50 mL) is refluxed with various aromatic aldehydes in the presence of 2 % NaOH (5ml) for 10 h, concentrated, cooled and poured onto ice. The solids thus obtained were recrystallized from appropriate solvents. Physical, analytical and spectroscopic data of compounds are as follows, respectively.

b) Microwave method

A solution of *N*-(4*H*-1,2,4-triazol-4-yl)acetamide (2) (0.01 mol) in absolute ethanol (30 mL) and various aromatic aldehydes (0.01 mol) were taken and to it 5 mL of 2% NaOH solution was added. The reaction mixture was taken in round-bottomed flask placed in a microwave oven and irradiated for 10-12 min, at 7P/L, 65% Power & 455 watt and then concentrated, cooled and poured onto ice. The solids thus obtained were filtered, dried and recrystallized from appropriate solvents.

3-(4-chlorophenyl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide (3a).

White powder, Yield 74%, m.p.159^{o}C; TLC (Acetone : Toluene, 2:8). IR: (KBr, cm^{-1}) 3426 (N-H), 3123 (Ar C-H stretch), 2925 and 2852 (C-H stretch), 1691 (NH-C=O), 1653 (CH=CH of –Carbonyl-CH=CH-), 1595 (C=N in triazole ring), 1513 (C=C of aromatic ring), 762 (C-Cl); ^{1}HNMR (400 MHz, $CDCl_3$) δ/ppm: 8.31 (ss, 1H, N-H), 7.40-7.38 (d, 1H, -CO-CH=), 7.99-7.97 (d, 1H, =CH-Ar), 7.49-7.47 (d, 2H, Ar-H), 7.84-7.81 (d, 2H, Ar-H), 8.80 (ss, 2H, -CH=N in triazole ring); ^{13}CNMR (400 MHz, DMSO) δ/ppm: δ 166.13 (-NH-C=0), δ 156.03 (triazole ring C), δ 138.48 (=C-Ar), δ 130.73; 130.55; 129.56; 128.88 (Aromatic C), 128.13 (Carbonyl-C=); MS m/z (%): 249 (54) [M^{+}], 68 (100) [M+H^{+}]; Elemental Analysis Calculated: C: 53.13; H: 3.65; N: 22.53; Found: C: 53.10; H: 3.60; N: 22.45.

3-(4-(dimethylamino)phenyl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide (3b).

Orange crystals, Yield 70%, m.p. 184^{o}C; TLC (Acetone: Toluene, (2:8). IR: (KBr, cm^{-1}) 3416 (N-H), 3090 (Ar CH stretch), 2915 and 2830 (C-H stretch), 1643 (NH-C=O), 1595 (C=N in triazole ring), 1540 (C=C of aromatic ring), 1330 (C–N); ^{1}H NMR (400 MHz, $CDCl_3$) δ/ppm: 8.39 (ss, 1H, N-H), 6.71-6.70 (d, 1H, -CO-CH=), 7.68 (d, 1H, =CH-Ar), 6.72 (d, 2H, Ar-H), 7.67-7.66 (d, 2H, Ar-H), 8.53 (ss, 2H, -CH=N in triazole ring), 3.08 (ss, 6H, N-$(CH_3)_2$); ^{13}CNMR (400 MHz, DMSO) δ/ppm: δ 164.88 (-NH-C=0), δ 138.88 (triazole ring C), δ 135.58 (=C-Ar), δ 152.18; 129.63, 128.50; 127.86 (Aromatic C), 129.01 (Carbonyl-C=), δ 39.78; 39.57 (-N$(CH_3)_2$; MS m/z (%): 258 (52.5) [M^{+}], 68 (100) [M+H^{+}]; Elemental Analysis Calculated: C: 60.69; H: 5.88; N: 27.22; Found: C: 60.61; H: 5.83; N: 27.15.

3-(naphthalen-1-yl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide (3c).

Yellow crystals, Yield 69%, m.p. 225^{o}C; TLC (Acetone : Toluene, 2:8). IR: (KBr, cm^{-1}) 3458 (N-H), 3070 (Ar C-H stretch), 2930 and 2840 (C-H stretch), 1664 (NH-C=O), 1625 (CH=CH of –Carbonyl-CH=CH-), 1580 (C=N in triazole ring), 1535 (C=C of aromatic ring); ^{1}HNMR (400 MHz, $CDCl_3$) δ/ppm: 8.21 (ss, 1H, N-H), 6.44-6.41 (d, 1H, -CO-CH=), 7.58-7.54 (d, 1H, =CH-Ar), 7.59-7.98 (m, 7H, Ar-H of Naphthalenyl), 8.90 (ss, 2H, -CH=N in triazole ring); ^{13}CNMR (400 MHz, DMSO) δ/ppm: δ 168.72 (-NH-C=0), δ 146.26 (triazole ring C), δ 143.96 (=C-Ar), δ 133.64; 132.00; 128.82; 128.34; 126.95; 126.32; 126.30; 124.22; 122.96 (Aromatic C), 120.01 (Carbonyl-C=); MS m/z (%): 265 (48.2) [M^{+}], 68 (100) [M+H^{+}]; Elemental Analysis Calculated: C: 68.17; H: 4.58; N: 21.20; Found: C: 68.08; H: 4.54; N: 21.12.

3-(3-methoxyphenyl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide (3d).

Greenish powder, Yield 72%, m.p. 126^{o}C; TLC (Acetone : Toluene, 2:8). IR: (KBr, cm^{-1}) 3440 (N-H), 3110 (Ar C-H stretch), 2985 and 2838 (C-H stretch), 1671 (NH-C=O), 1635 (CH=CH of –Carbonyl-CH=CH-), 1587 (C=N in triazole ring), 1510 (C=C of aromatic ring); ^{1}HNMR (400 MHz, $CDCl_3$) δ/ppm: 8.12 (ss, 1H, N-H), 6.48-6.44 (d, 1H, -CO-CH=), 7.59-7.57 (d, 1H, =CH-Ar), 7.69-7.54 (m, 4H, Ar-H), 3.63 (ss, 3H, -O-CH_3), 8.48 (ss, 2H, -CH=N in triazole ring); ^{13}CNMR (400 MHz, DMSO) δ/ppm: δ 164.78 (-NH-C=0), δ 148.88 (triazole ring C), δ 142.48 (=C-Ar), δ 160.32; 134.22; 129.65, 120.82; 115.36; 113.24 (Aromatic C), 119.52 (Carbonyl-C=), δ 57.68 (-O-CH_3); MS m/z (%): 245 (51.6) [M^{+}], 68 (100) [M+H^{+}]; Elemental Analysis Calculated: C: 59.01; H: 4.95; N: 22.94; Found: C: 58.95; H: 4.89; N: 22.82.

3-(2-hydroxyphenyl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide (3e).

Yellowish powder, Yield 65%, m.p. 196^{o}C; TLC (Acetone : Toluene, 2:8). IR: (KBr, cm^{-1}) 3435 (N-H), 3020 (Ar C-H stretch), 2925 and 2830 (C-H stretch), 1631 (NH-C=O), 1614 (CH=CH of –Carbonyl-CH=CH-), 1575 (C=N in triazole ring), 1535 (C=C of aromatic ring); ^{1}HNMR (400 MHz, $CDCl_3$) δ/ppm: 8.27 (ss, 1H, N-H), 6.40-6.38 (d, 1H, -CO-CH=), 7.79-7.77 (d, 1H, =CH-Ar), 6.44-7.76 (m, 4H, Ar-H), 8.70 (ss, 2H, -CH=N in triazole ring), 5.40 (ss, 1H, Aromatic C-OH); ^{13}CNMR (400 MHz, DMSO) δ/ppm: δ 162.52 (-NH-C=0), δ 148.61 (triazole ring C), δ 145.38 (=C-Ar), δ 157.27; 129.32; 128.96; 122.63, 121.20; 117.64 (Aromatic C), 116.18 (Carbonyl-C=); MS m/z (%): 231 (56.2) [M^{+}], 68 (100) [M+H^{+}]; Elemental Analysis Calculated: C: 57.39; H: 4.38; N: 24.34; Found: C: 57.45; H: 4.31; N: 24.42.

3-(2,4-dichlorophenyl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide (3f).

Grey powder, Yield 76%, m.p. 154^{o}C; TLC (Acetone : Toluene, 2:8). IR: (KBr, cm^{-1}) 3435 (N-H), 3046 (Ar C-H stretch), 2920 and 2845 (C-H stretch), 1652 (NH-C=O), 1610 (CH=CH of –Carbonyl-CH=CH-), 1530 (C=N in triazole ring), 1505 (C=C of aromatic ring); ^{1}HNMR (400 MHz, $CDCl_3$) δ/ppm: 8.1021 (ss, 1H, N-H), 6.6938-6.6133 (d, 1H, -CO-CH=), 8.4906-8.4691 (d, 1H, =CH-Ar), 8.0904-8.0690 (d, 1H, Ar-H), 7.5188-7.4973 (d, 1H, Ar-H), 7.6521 (ss, 1H, Ar-H), 9.1690 (ss, 2H, -CH=N in triazole ring); ^{13}CNMR (400 MHz, DMSO) δ/ppm: δ 164.16 (-NH-C=0), δ 138.88 (triazole ring C), δ 137.65 (=C-Ar), δ 135.58; 133.68; 129.63; 129.01; 128.59; 127.86 (Aromatic C), 119.53 (Carbonyl-C=); MS m/z (%): 284 and 285 (40.1)(18.3) [M^{+}][M^{+1}], 68 (100) [M+H^{+}]; Elemental Analysis Calculated: C: 46.67; H: 2.85; N: 19.79; Found: C: 46.54; H: 2.78; N: 19.64.

***N*-(4*H*-1,2,4-triazol-4-yl)-3-(3,4,5-trimethoxyphenyl)acrylamide (3g).**

White powder, Yield 69%, m.p. 162^{o}C; TLC (Acetone : Toluene, 2:8). IR: (KBr, cm^{-1}) 3415 (N-H), 3026 (Ar C-H stretch), 2928 and 2830 (C-H stretch), 1684 (NH-C=O), 1615 (CH=CH of –Carbonyl-CH=CH-), 1525 (C=N in triazole ring), 1513 (C=C of aromatic ring); ^{1}HNMR (400 MHz, $CDCl_3$) δ/ppm: 8.1811 (ss, 1H, N-H), 6.4951-6.4532 (d, 1H, -CO-CH=), 7.4806-7.4610 (d, 1H, =CH-Ar), 6.1904-6.1690 (d, 2H, Ar-H), 3.3415 (ss, 9H, -O-CH_3), 8.2645 (ss, 2H, -CH=N in triazole ring); ^{13}CNMR (400 MHz, DMSO) δ/ppm: δ 168.26 (-NH-C=0), δ 148.08 (triazole ring C), δ 141.65 (=C-Ar), δ 153.85; 138.23; 130.36; 103.81 (Aromatic C), 118.35 (Carbonyl-C=), 56.32 (-OCH_3); MS m/z (%): 305 (30.8) [M^{+}], 68 (100) [M+H^{+}]; Elemental Analysis Calculated: C: 55.26; H: 5.30; N: 18.41; Found: C: 55.12; H: 5.26; N: 18.38.

3-(4-methoxyphenyl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide (3h).

Greenish powder, Yield 77%, m.p. 116^{o}C; TLC (Acetone : Toluene, 2:8). IR: (KBr, cm^{-1}) 3429 (N-H), 3086 (Ar C-H stretch), 2938 and 2856 (C-H stretch), 167 8 (NH-C=O), 1612 (CH=CH of –Carbonyl-CH=CH-), 1518 (C=N in triazole ring),

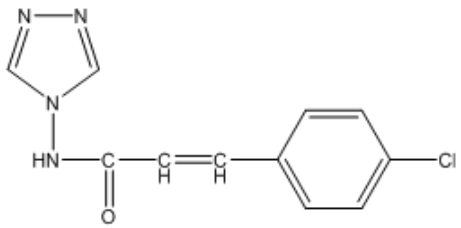
^{1}HNMR (400 MHz, $CDCl_3$) δ/ppm: 8.1239 (ss, 1H, N-H), 6.4329-6.4117 (d, 1H, -CO-CH=), 7.5960-7.5621 (d, 1H, =CH-Ar), 6.9604-6.9490 (d, 2H, Ar-H), 7.6440-7.6209 (d, 2H, Ar-H), 3.8415 (ss, 3H, -O-CH_3), 8.4645 (ss, 2H, -CH=N in triazole ring); ^{13}CNMR (400 MHz, DMSO) δ/ppm: δ 166.62 (-NH-C=0), δ 146.80 (triazole ring C), δ 140.52 (=C-Ar), δ 159.51; 130.36; 122.69; 113.18 (Aromatic C), 119.95 (Carbonyl-C=), 56.43 (-OCH_3); MS m/z (%): 245 (46.7) [M^+], 68 (100) [$M+H^+$]; Elemental Analysis Calculated: C: 59.01; H: 4.95; N: 22.94; Found: C: 58.92; H: 4.87; N: 22.99.

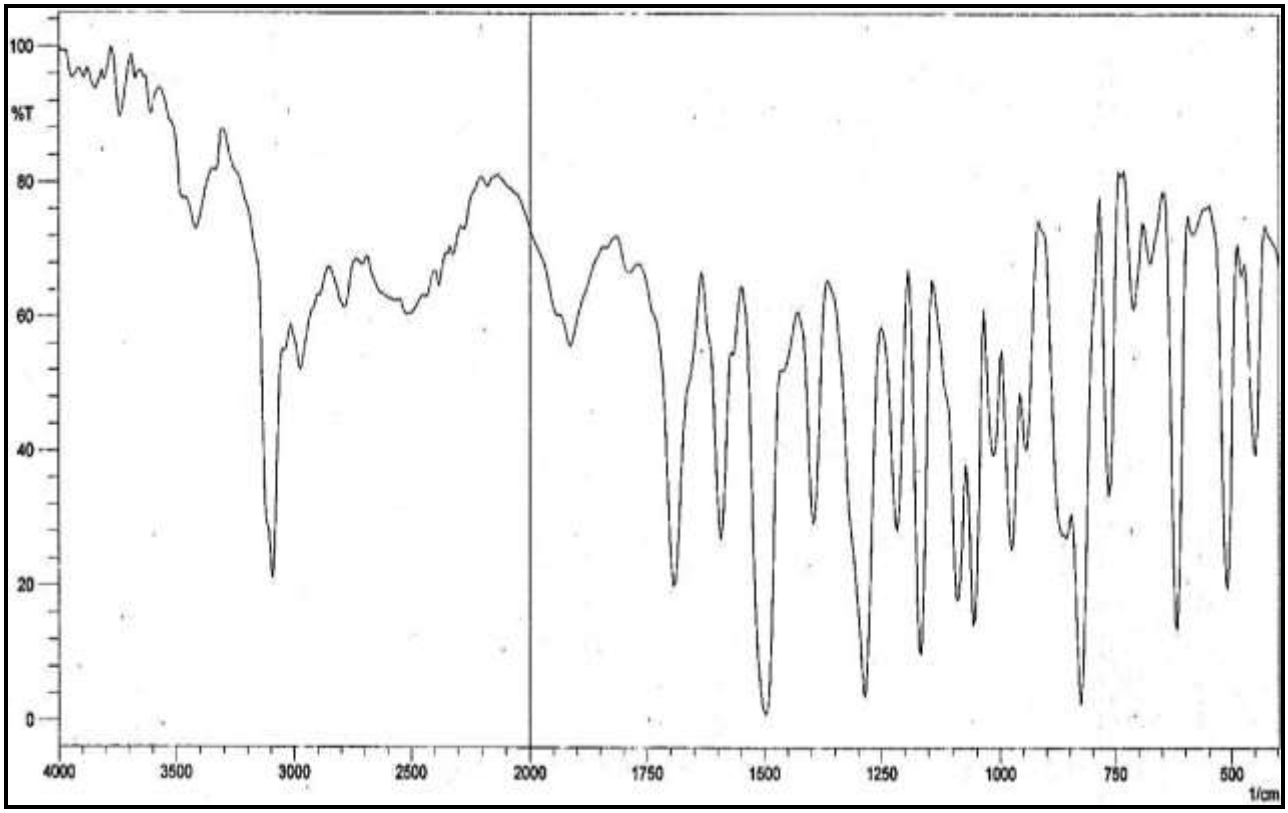
3-(3-nitrophenyl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide (3i).

White powder, Yield 68%, m.p. 151^{o}C; TLC (Acetone : Toluene, 2:8). IR: (KBr, cm^{-1}) 3414 (N-H), 3056 (Ar C-H stretch), 2924 and 2858 (C-H stretch), 1679 (NH-C=O), 1613 (CH=CH of –Carbonyl-CH=CH-), 1524 (C=N in triazole ring), 1528 (C=C of aromatic ring); ^{1}HNMR (400 MHz, $CDCl_3$) δ/ppm: 8.1149 (ss, 1H, N-H), 7.8111-7.7912 (d, 1H, -CO-CH=), 8.0568-8.0310 (d, 1H, =CH-Ar), 8.2805-8.2611 (d, 1H, Ar-H [-C-CH-CH-]), 8.4067-8.3811 (Multiplet, 1H, Ar-H [-CH-CH-CH-]), 8.7099 (d, 1H, Ar-H [-CH-CH-C-NO_2]), 9.0848 (ss, 2H, -CH=N in triazole ring), 9.3248 (ss, 1H, Ar-H [-C-CH-C-]); ^{13}CNMR (400 MHz, DMSO) δ/ppm: δ 169.15 (-NH-C=0), δ 138.68 (triazole ring C), δ 135.64 (=C-Ar), δ 148.05; 133.84; 130.29; 125.97; 123.15; 122.66 (Aromatic C), 116.65 (Carbonyl-C=); MS m/z (%): 260 (38.9) [M^+], 68 (100) [$M+H^+$]; Elemental Analysis Calculated: C: 50.97; H: 3.50; N: 27.02; Found: C: 50.89; H: 3.38; N: 26.95.

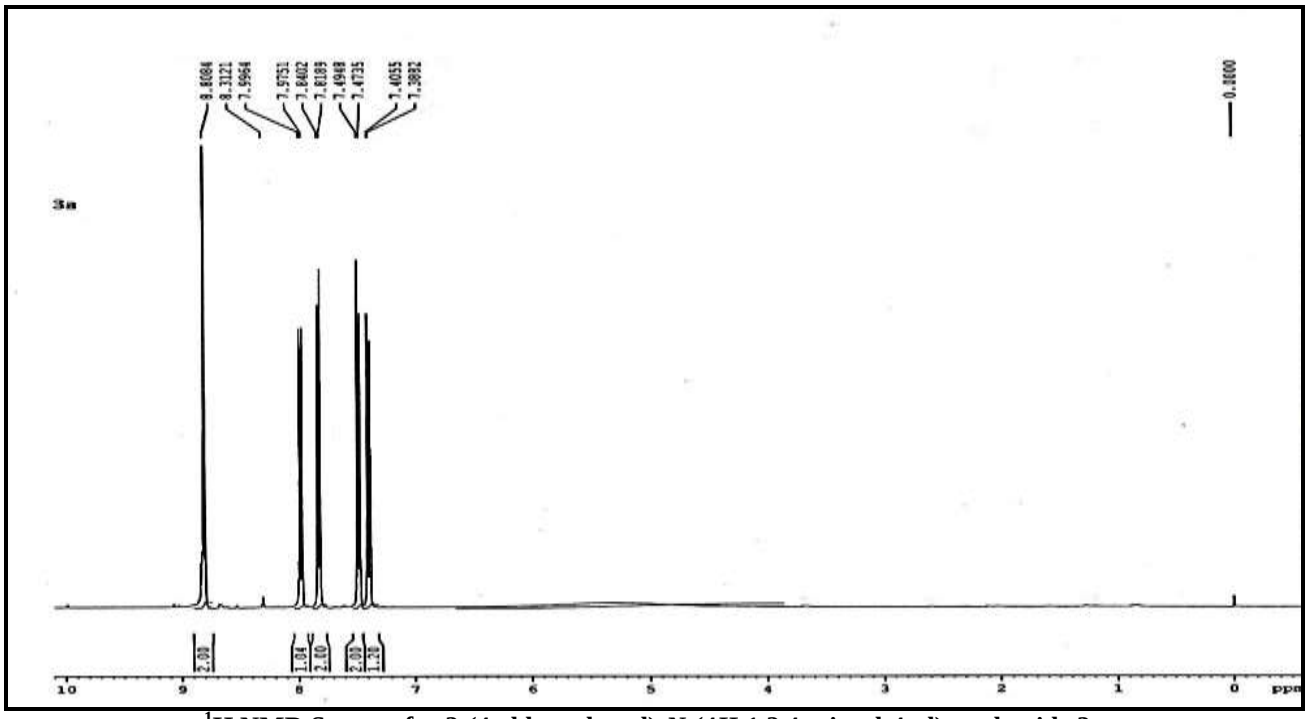
3-(4-bromophenyl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide (3j).

Brown powder, Yield 74%, m.p. 218^{o}C; TLC (Acetone : Toluene, 2:8). IR: (KBr, cm^{-1}) 3426 (N-H), 3095 (Ar C-H stretch), 2938 and 2813 (C-H stretch), 1695 (NH-C=O), 1619 (CH=CH of –Carbonyl-CH=CH-), 1523 (C=N in triazole ring), 1502 (C=C of aromatic ring); ^{1}HNMR (400 MHz, $CDCl_3$) δ/ppm: 8.0213 (ss, 1H, N-H), 6.4834-6.4612 (d, 1H, -CO-CH=), 7.4562-7.4323 (d, 1H, =CH-Ar), 7.6305-7.6198 (d, 2H, Ar-H), 7.5531-7.5374 (d, 2H, Ar-H), 8.2884 (ss, 2H, -CH=N in triazole ring); ^{13}CNMR (400 MHz, DMSO) δ/ppm: δ 162.28 (-NH-C=0), δ 144.63 (triazole ring C), δ 142.57 (=C-Ar), δ 134.87; 131.43; 128.98; 122.74 (Aromatic C), 120.98 (Carbonyl-C=); MS m/z (%): 294 (59.2) [M^+], 68 (100) [$M+H^+$]; Elemental Analysis Calculated: C: 45.07; H: 3.09; N: 19.11; Found: C: 45.01; H: 3.02; N: 18.98.

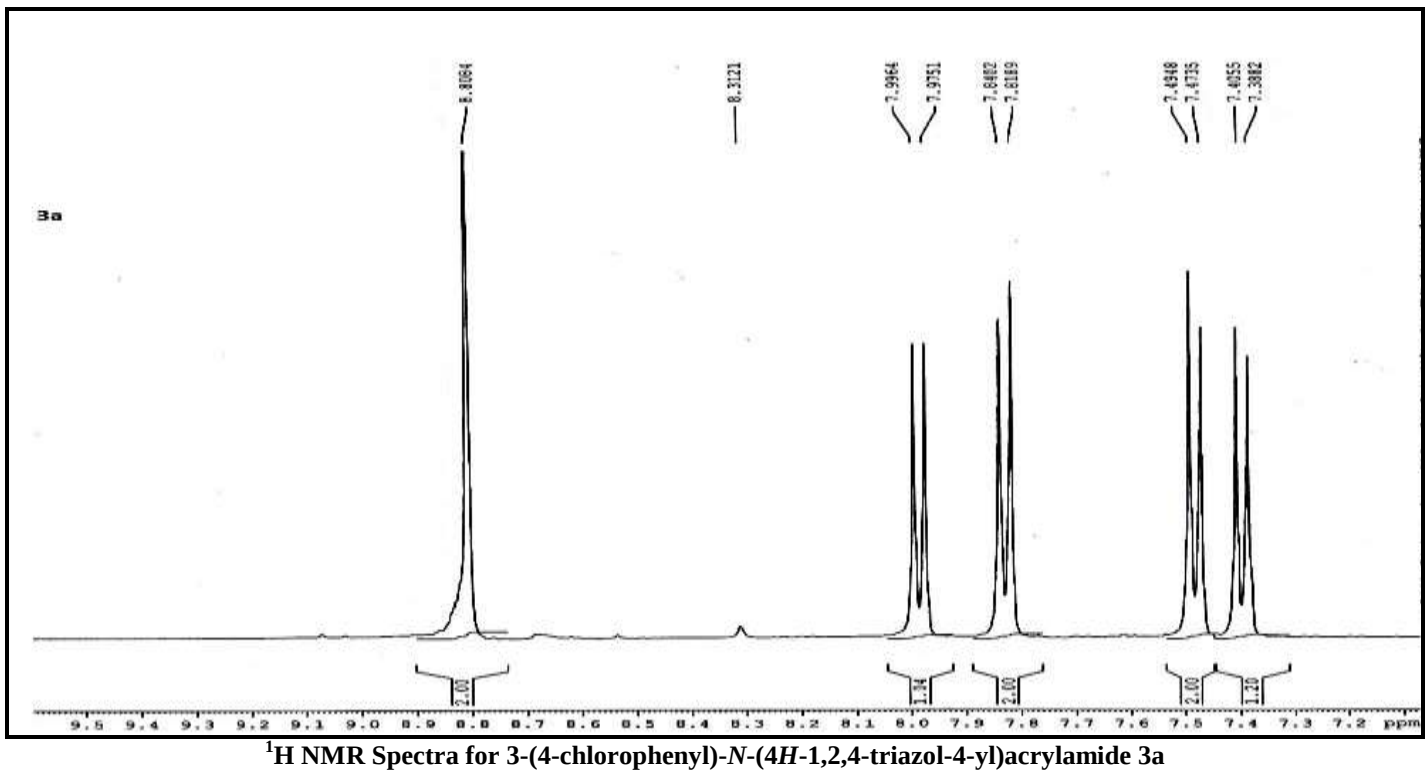




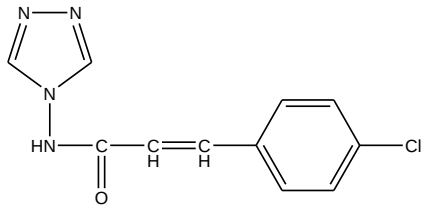
Infrared Spectra for 3-(4-chlorophenyl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide 3a

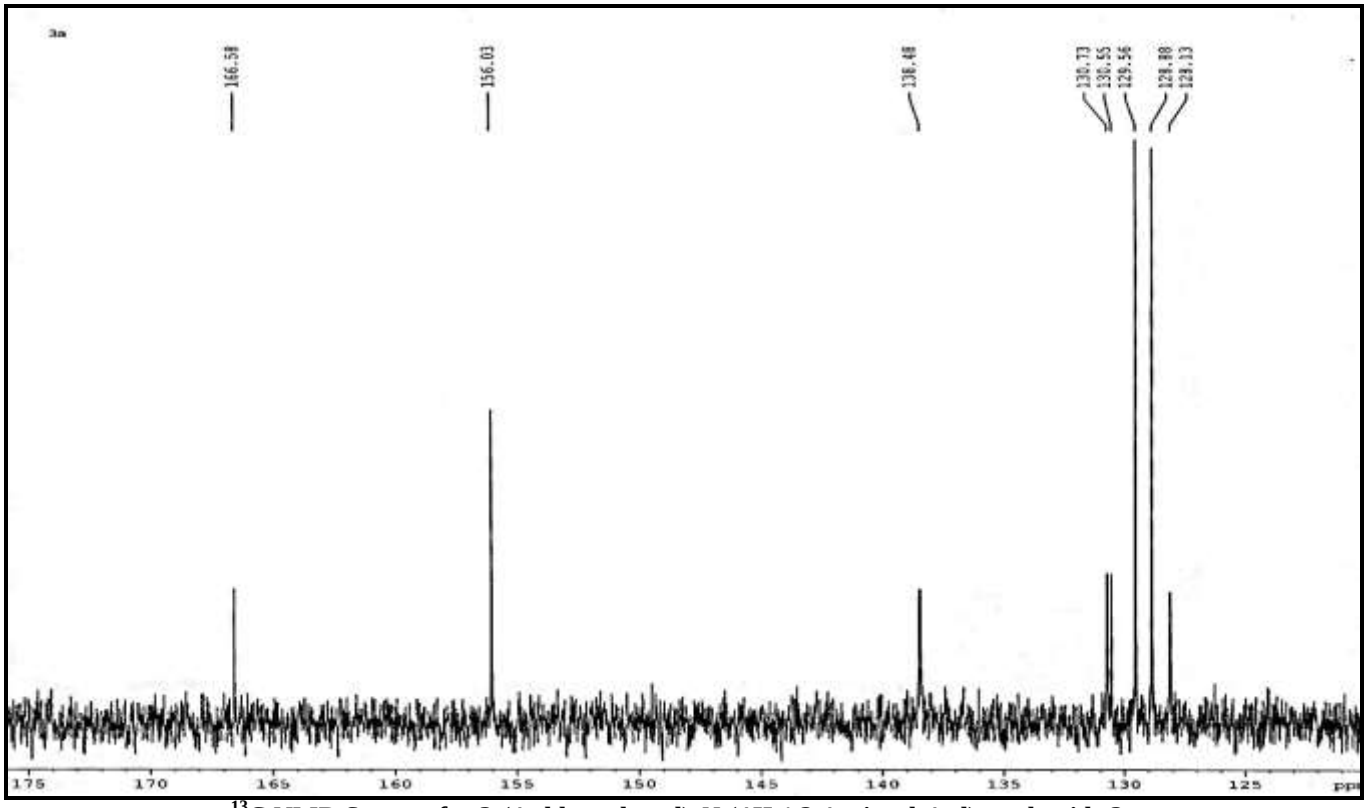


[1]H NMR Spectra for 3-(4-chlorophenyl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide 3a

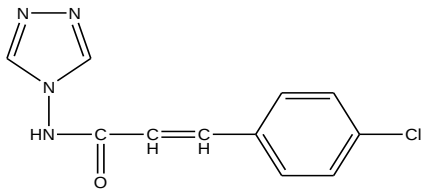


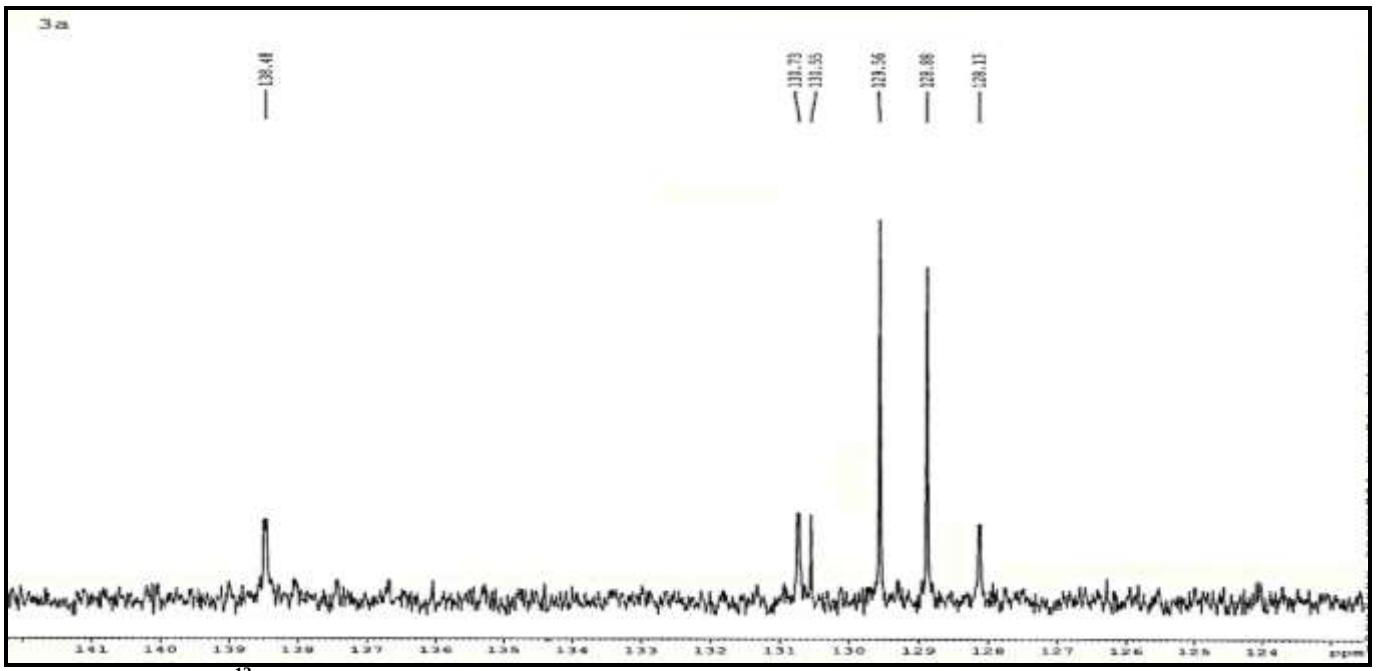
[1]H NMR Spectra for 3-(4-chlorophenyl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide 3a



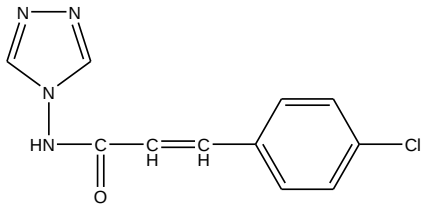


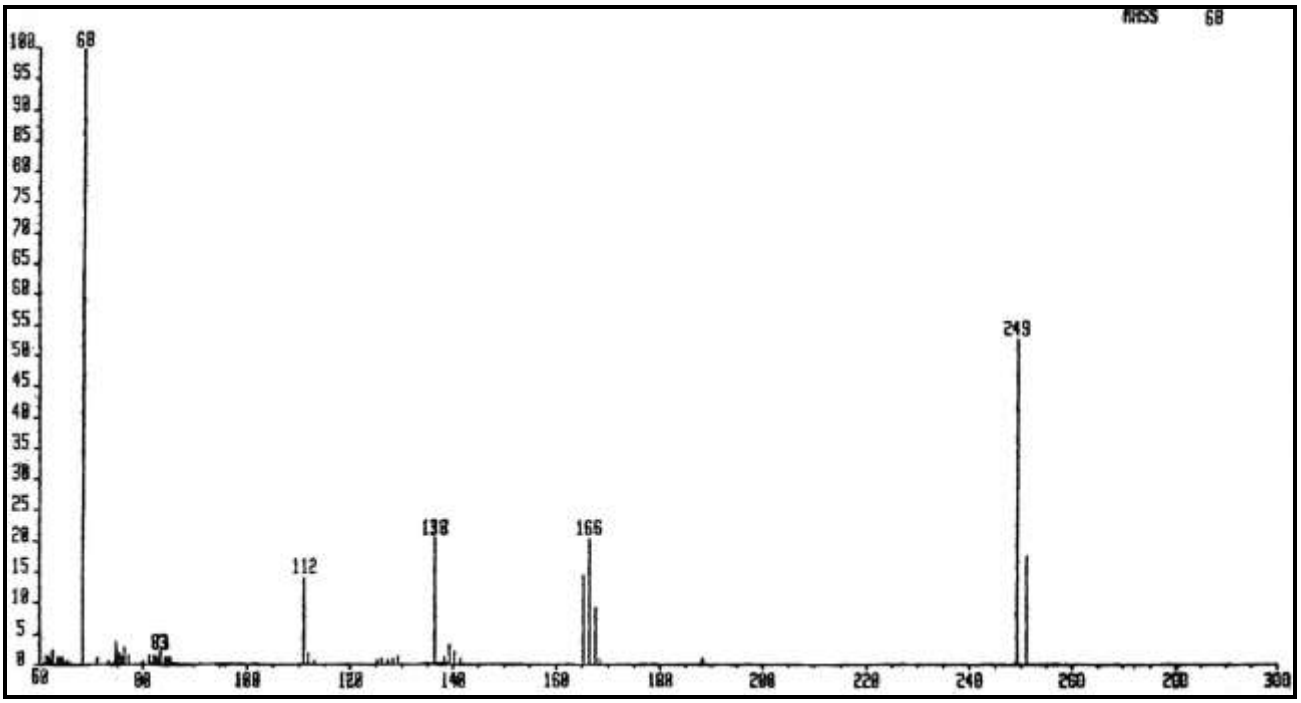
^{13}C NMR Spectra for 3-(4-chlorophenyl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide 3a



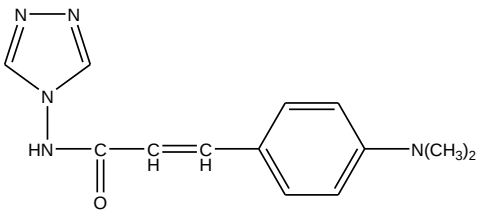


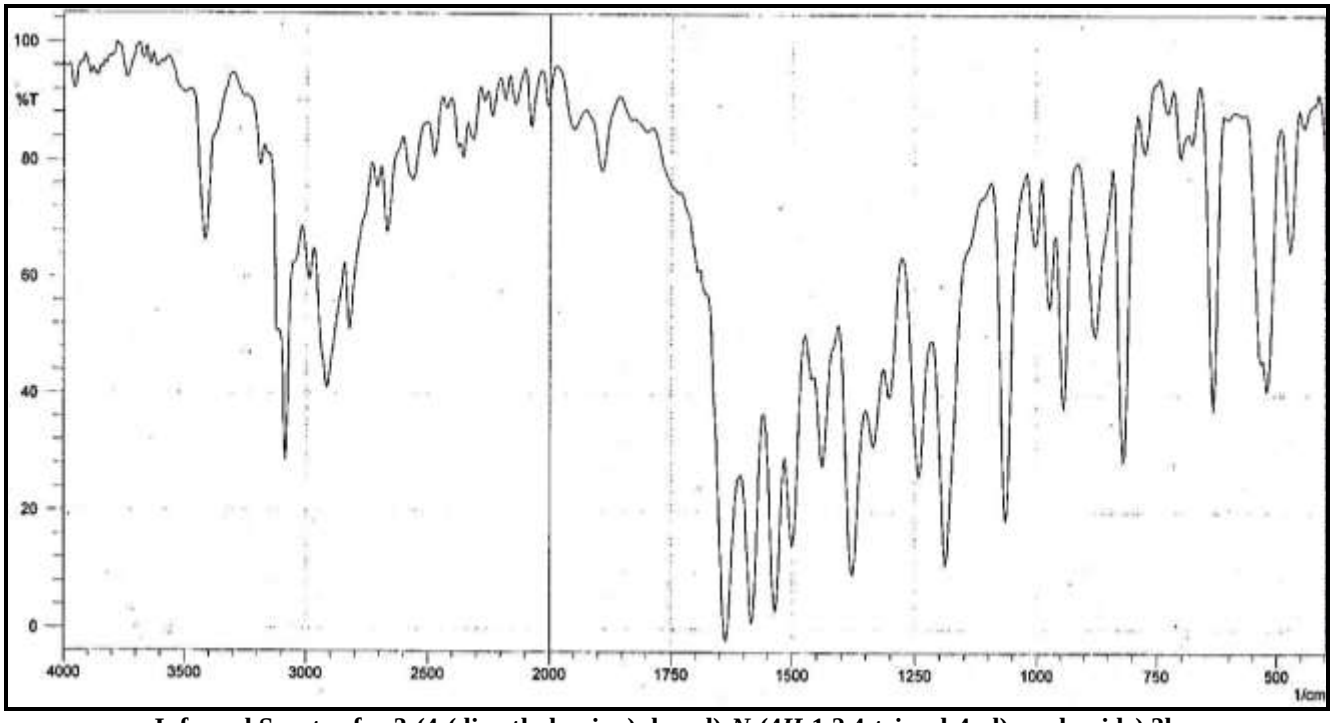
^{13}C NMR Spectra for 3-(4-chlorophenyl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide 3a



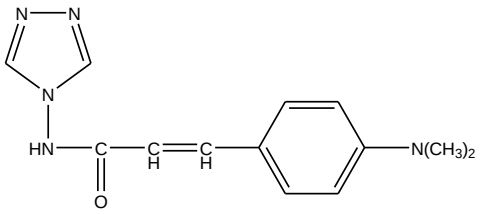


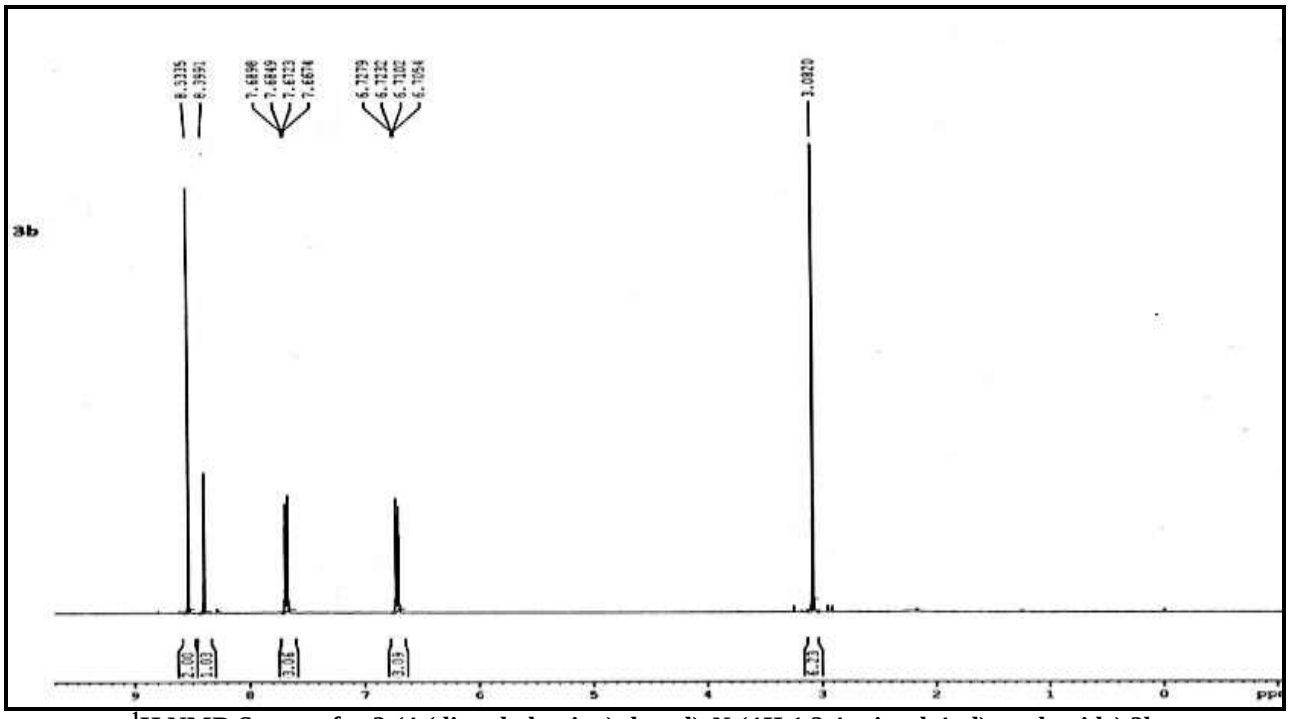
Mass Spectra for 3-(4-chlorophenyl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide 3a



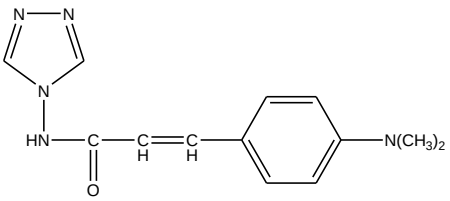


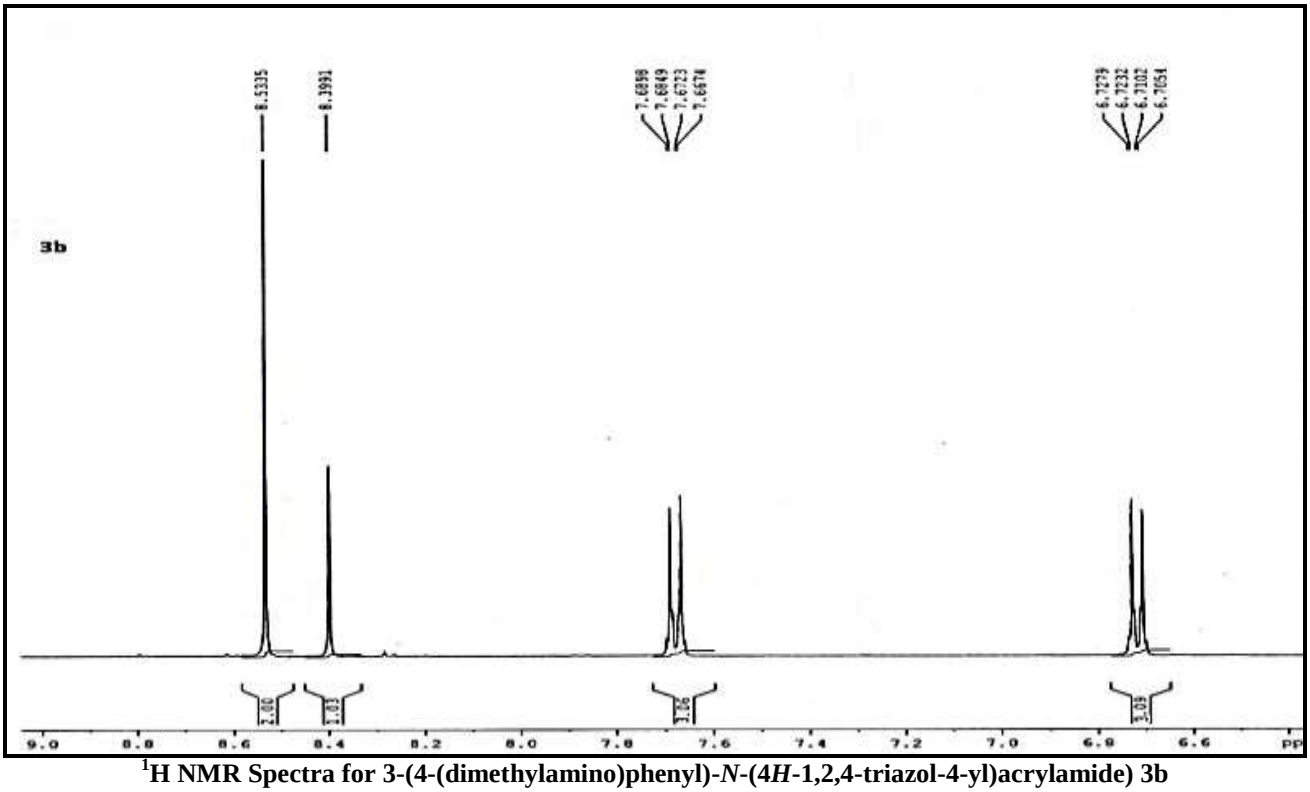
Infrared Spectra for 3-(4-(dimethylamino)phenyl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide) 3b



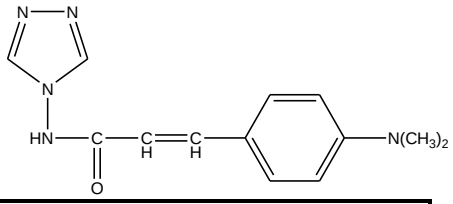


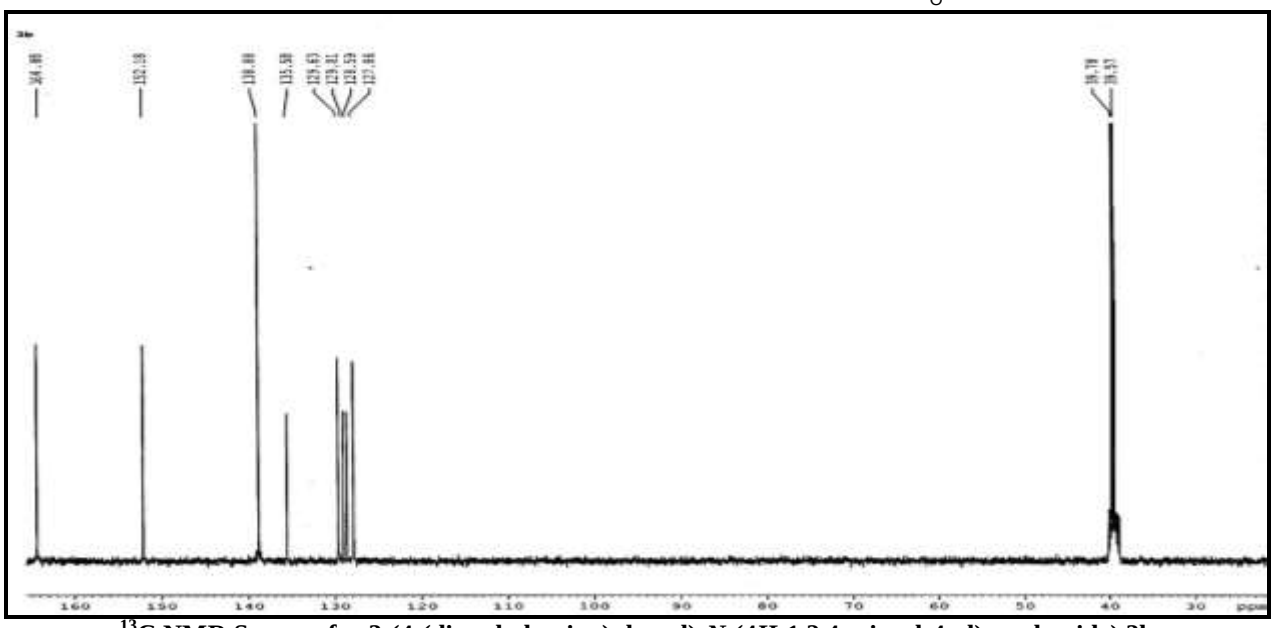
^{1}H NMR Spectra for 3-(4-(dimethylamino)phenyl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide) 3b



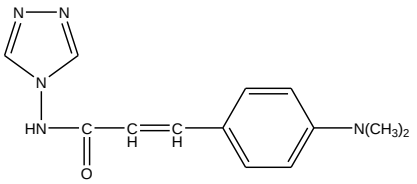


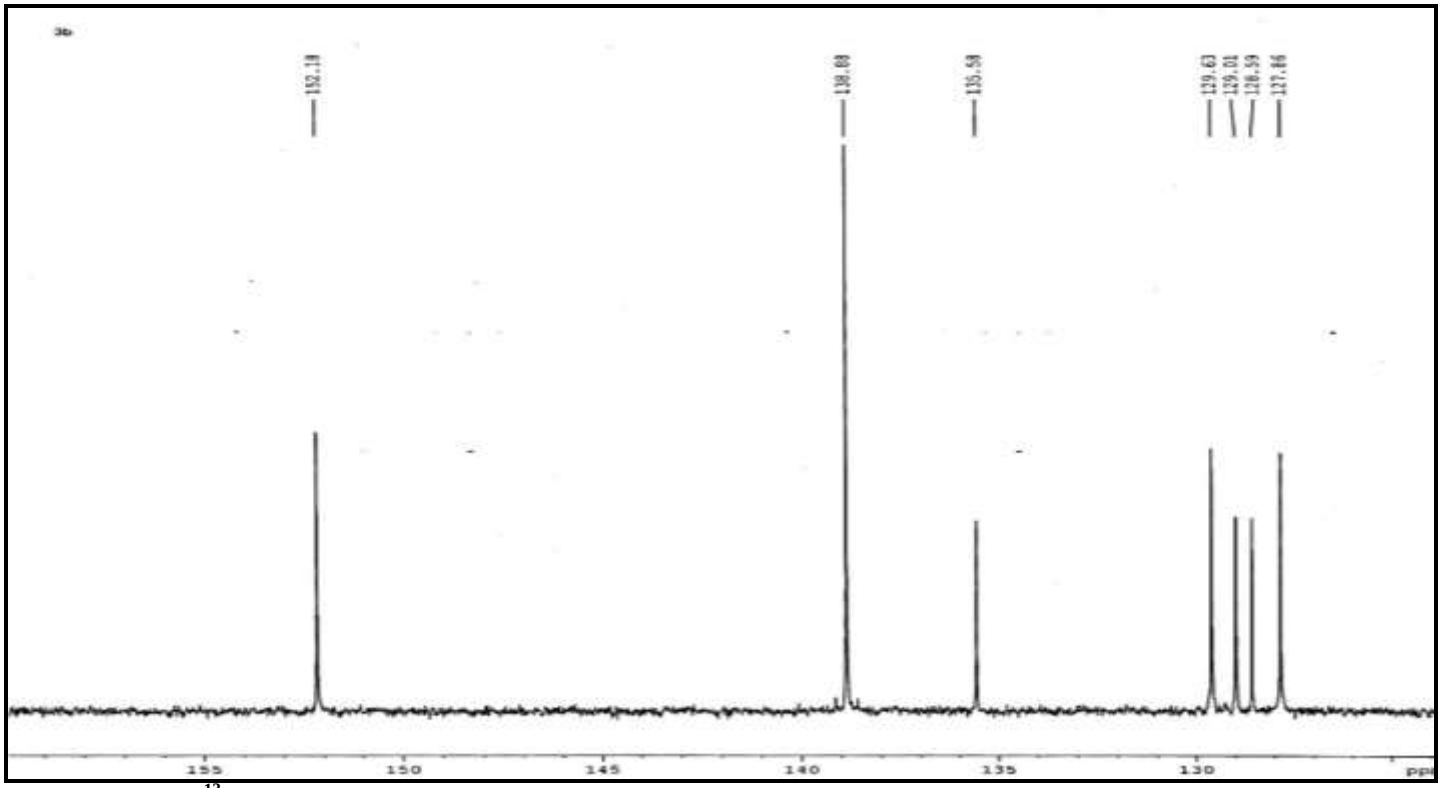
^{1}H NMR Spectra for 3-(4-(dimethylamino)phenyl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide) 3b



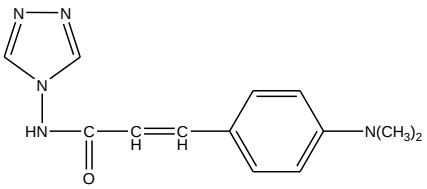


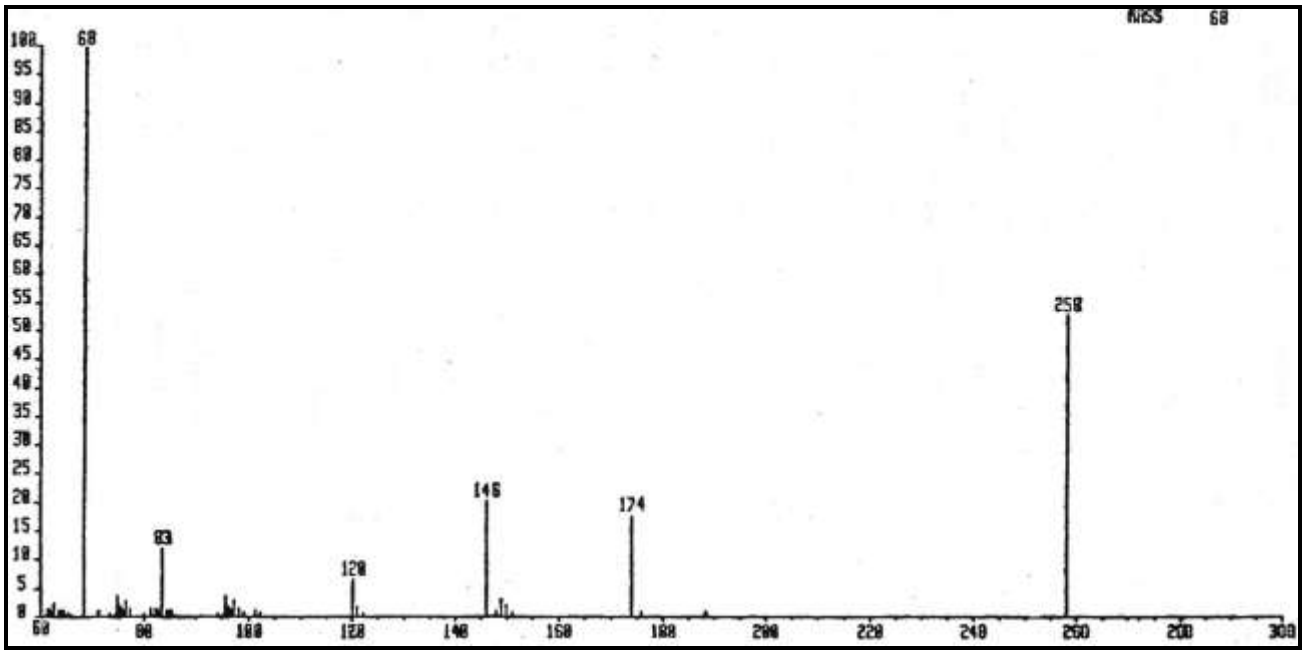
^{13}C NMR Spectra for 3-(4-(dimethylamino)phenyl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide) 3b



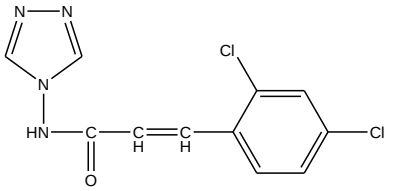


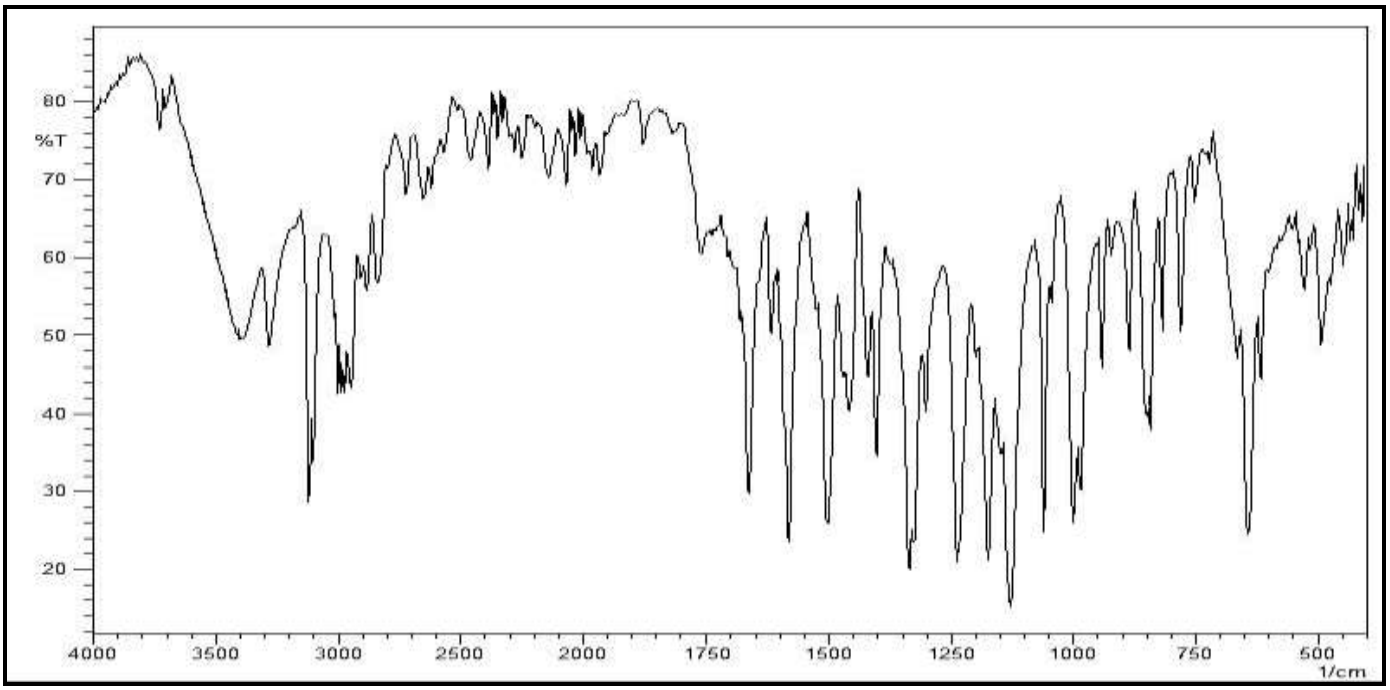
^{13}C NMR Spectra for 3-(4-(dimethylamino)phenyl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide) 3b



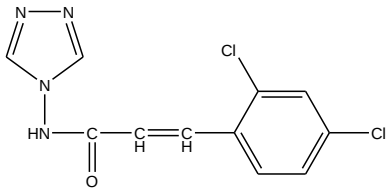


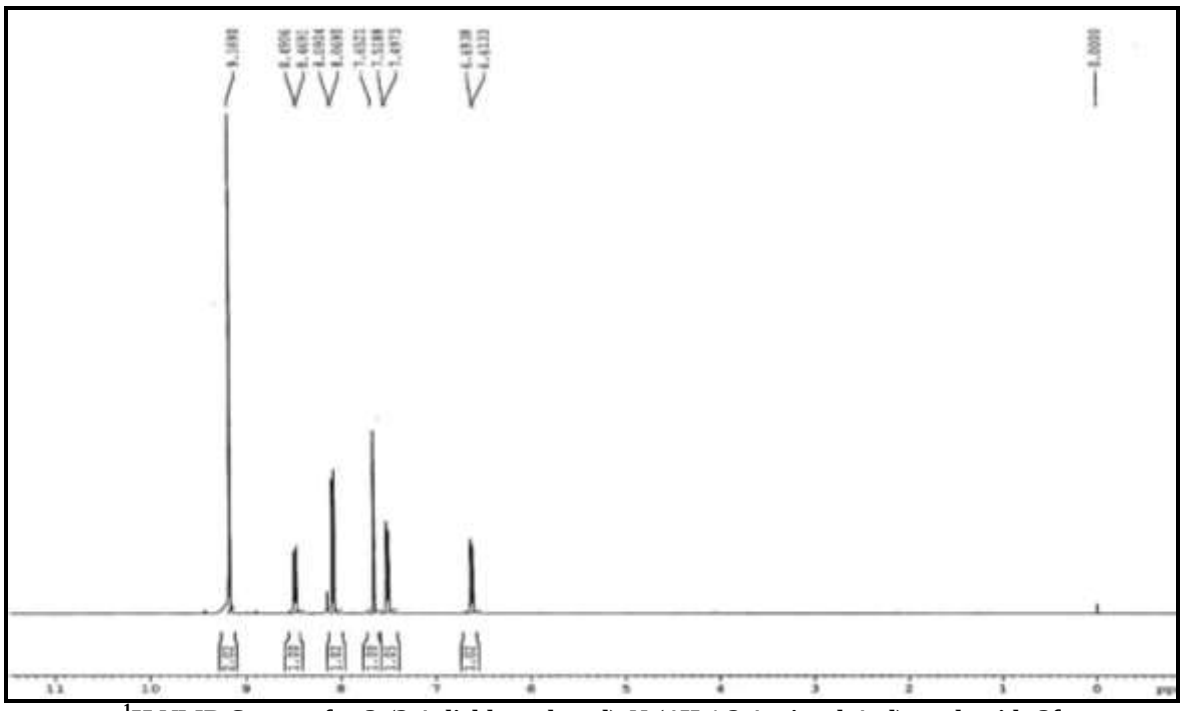
Mass Spectra for 3-(4-(dimethylamino)phenyl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide) 3b



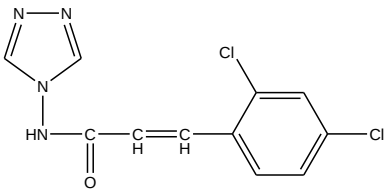


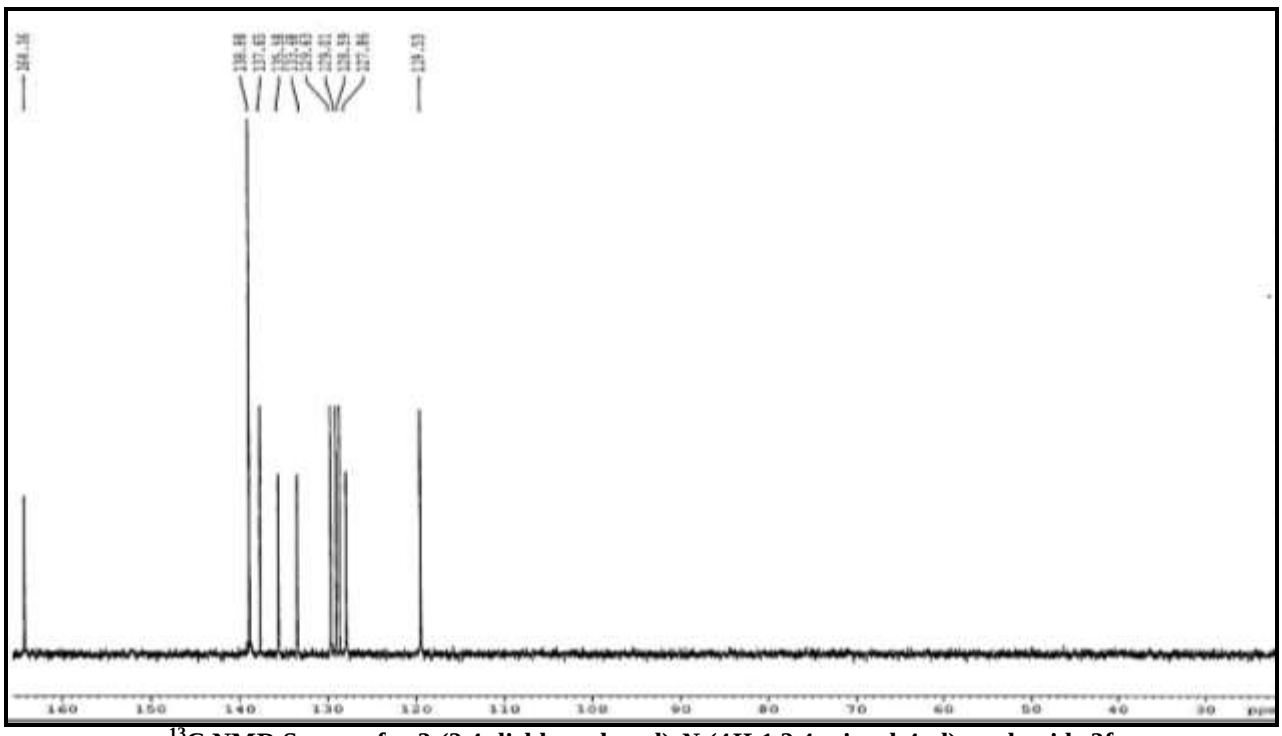
Infrared Spectra for 3-(2,4-dichlorophenyl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide 3f



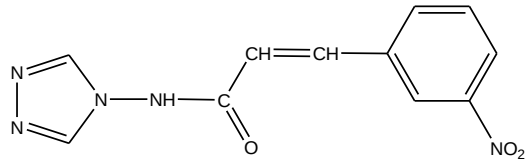


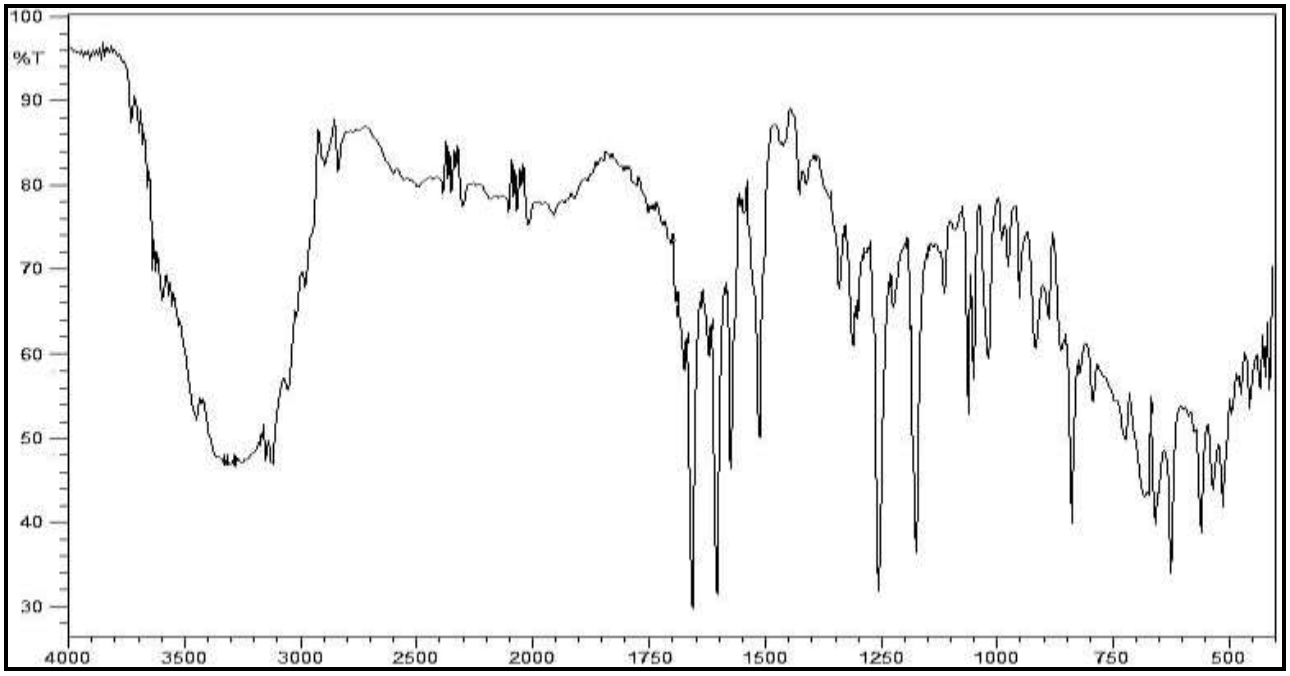
^{1}H NMR Spectra for 3-(2,4-dichlorophenyl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide 3f



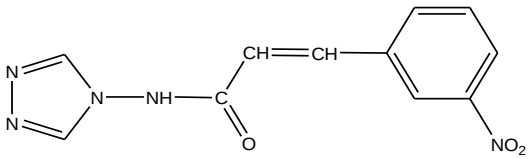


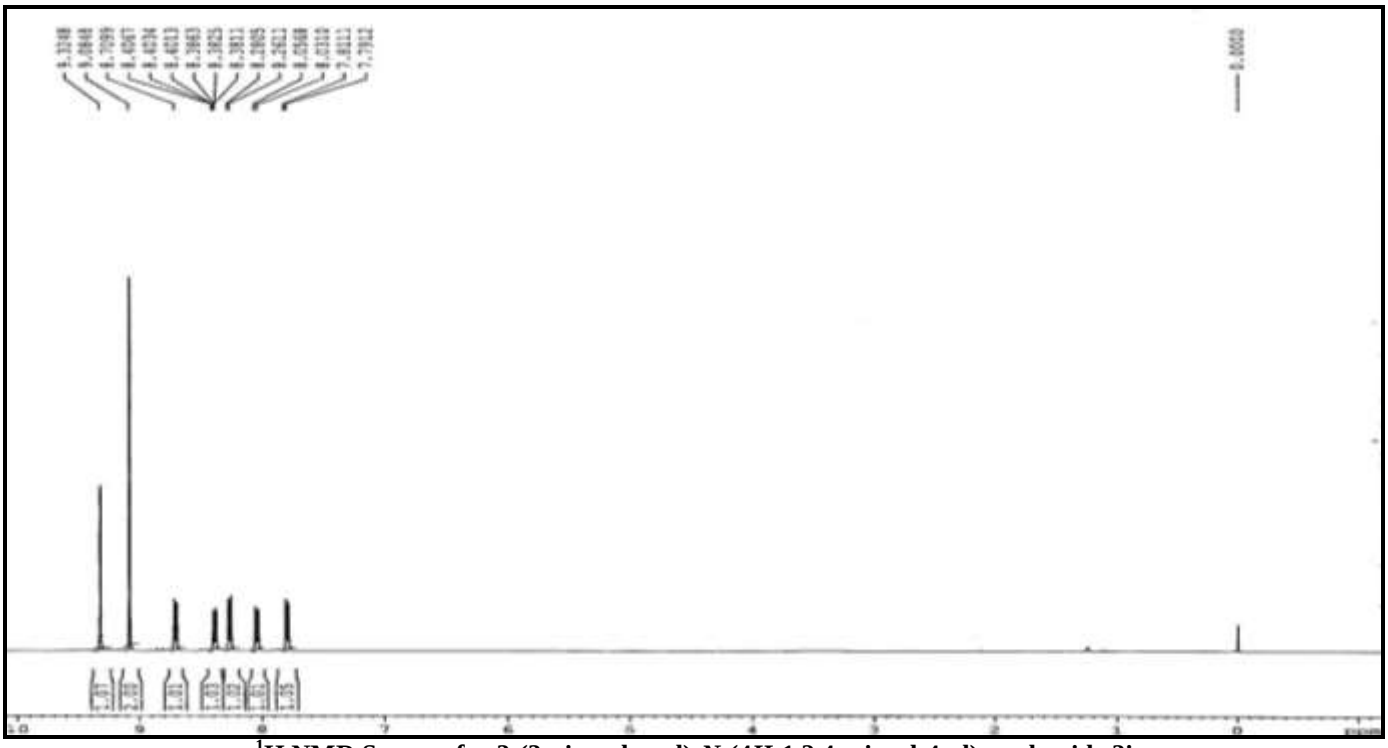
^{13}C NMR Spectra for 3-(2,4-dichlorophenyl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide 3f



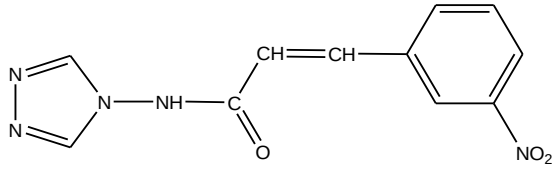


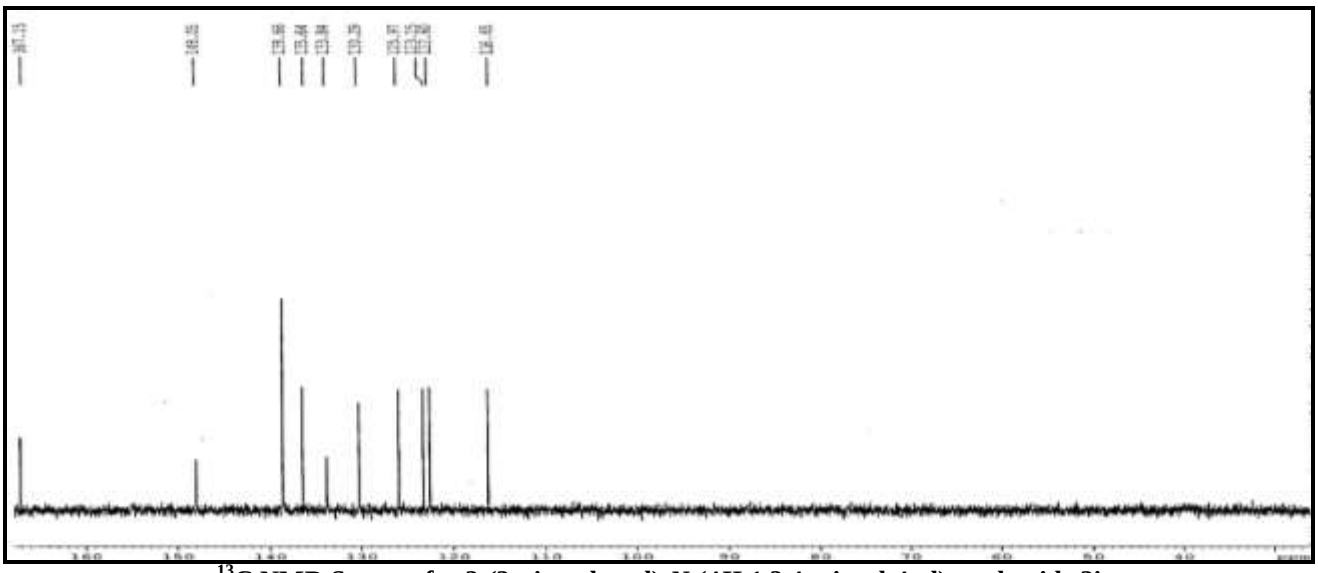
Infrared Spectra for 3-(3-nitrophenyl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide 3i





^{1}H NMR Spectra for 3-(3-nitrophenyl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide 3i





^{13}C NMR Spectra for 3-(3-nitrophenyl)-*N*-(4*H*-1,2,4-triazol-4-yl)acrylamide 3i

Table 1. Physical data of synthesized compounds 2, 3a-j

Compound	R	M.P. (°C)	Yield (%)	Mol. Formula	Mol. Weight	Recrystallization solvent*
2	-	155	84	$C_4H_6N_4O$	126.12	1
3a	-4-Cl	159	74	$C_{11}H_9ClN_4O$	248.67	1
3b	-4-$N(CH_3)_2$	186	70	$C_{13}H_{15}N_5O$	257.29	2
3c	-Naphthayl	225	69	$C_{15}H_{12}N_4O$	264.28	3
3d	-3-OCH_3	126	72	$C_{12}H_{12}N_4O_2$	244.25	1
3e	-2-OH	196	65	$C_{11}H_{10}N_4O_2$	230.22	2
3f	-2,4-Cl	154	76	$C_{11}H_8Cl_2N_4O$	283.11	1
3g	-3,4,5-OCH_3	162	69	$C_{14}H_{16}N_4O_4$	304.12	2
3h	-4-OCH_3	116	77	$C_{12}H_{12}N_4O_2$	244.10	1
3i	-3-NO_2	151	68	$C_{11}H_9N_5O_3$	259.22	3
3j	-4-Br	218	74	$C_{11}H_9BrN_4O$	293.12	2

*1. Methanol, 2. Ethanol, 3. Acetone

Table 2. Comparative Study in Terms of Yield and Reaction Period in the Presence of Various Powers and Temperatures for Conventional and Microwave Methods

Product	Substituents-R	Microwave Synthesis Method			Conventional Synthesis Method		
		Time (min.)	Power (watts)	Yield (%)[a]	Time (hr)	Temp.* (°C)	Yield (%)[b]
3a	-4-Cl	10.0	455	91	9.0	96	74
3b	-4-$N(CH_3)_2$	11.5	455	93	11.0	95	70
3c	-Naphthalene	10.5	455	92	10.0	96	69
3d	-3-OCH_3	10.0	455	89	9.5	92	72
3e	-2-OH	10.5	455	88	10.0	94	65
3f	-2,4-Cl	10.5	455	91	10.0	97	76
3g	-3,4,5-OCH_3	12.0	455	89	11.5	96	69
3h	-4-OCH_3	10.0	455	93	10.5	97	77
3i	-3-NO_2	12.0	455	91	11.5	94	68
3j	-4-Br	11.0	455	92	10.5	95	74

[a]Yield of isolated products of MWI method.
[b]Yield of isolated products of conventional method.
* = Reflux Temperature.

Table 3. The Effect of Microwave Irradiation Power[a]

Irradiation power (watts)	280	350	420	455
Yield (%)	80	84	87	91

[a]Irradiation time is 10.0 min.

Table 4. The Effect of Microwave Irradiation Time[a]

Irradiation time (min)	6	8	10	12	14
Yield (%)	85	87	91	88	83

[a]Irradiation power is 455 watts.

Acknowledgements

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References

1. Varma SR. Solvent-free organic syntheses using supported reagents and microwave irradiation. Green Chemistry. 1999; 1: 43-55.
2. Borah R, Kalita DJ, Sarma JC. Microwave promoted selective preparation of acetals and esters from aldehydes. Indian J. Chem. 2002; 41B: 1032-38.
3. Kidwai M, Dave B, Venkataramanan R. Microwave assisted solid phase synthesis of pyrimidine derivatives. Indian J. Chem. 2002; 41B: 2414-18.
4. Nielsen SF, Christensen SB, Cruziani G, Kharazmi A, Liljefors T. Antileishmanial chalcones: statistical design, synthesis, and three dimensional quantitative structure activity relationship analysis. J. Med. Chem. 1998; 41: 4819-32.
5. Liu M, Wilairat P, Go ML. Antimalarial alkoxylated and hydroxylated chalcones: structure_activity relationship analysis. J. Med. Chem. 2001; 44: 4443-52.
6. Rojas J, Dominguez JN, Charris JE, Lobo G, Paya M, Ferrandiz ML. Synthesis and inhibitory activity of dimethylamino-chalcone derivatives on the induction of nitric oxide synthase. Eur. J. Med. Chem. 2002; 37: 699-755.
7. Calvino V, Picallo M, Lopez-Peinado AJ, Martın-Aranda RM, Duran-Valle CJ. Ultrasound accelerated Claisen–Schmidt condensation: a green route to chalcones. Appl. Surf. Sci. 2006; 252: 6071-74.
8. Dixon RA, Paiva NL. Stress-induced phenylpropanoid metabolism. Plant cell. 1995; 7: 1085-97.
9. Li J. Name Reactions in Heterocyclic Chemistry. Wiley-Interscience Publication; 2005. p. 262-65.
10. Saravanamurugan S, Palanichamy M, Banumathi Arabindoo, Murugesan V. Solvent free synthesis of chalcone and flavanone over zinc oxide supported metal oxide catalysts. Catal. Commun. 2005; 6: 399-403.
11. Daskiewicz J B, Comte G, Barron D, Pietro AD, Thomasson F. Organolithium mediated synthesis of prenylchalcones as potential inhibitors of chemoresistance. Tetrahedron Lett. 1999; 40: 7095-98.
12. Li JT, Yang WZ, Wang ZX, Li SH, Li TS. Improved synthesis of chalcones under ultrasound irradiation. Ultrason. Sonochem. 2002; 9: 237-39.
13. Sebti S, Solhy A, Tahir R, Boulaajaj S, Mayoral JA, Fraile JM, Kossir A, Oumimoun H. Calcined sodium nitrate/natural phosphate: an extremely active catalyst for the easy synthesis of chalcones in heterogeneous media. Tetrahedron Lett. 2001; 42: 7953-55.
14. Climent MJ, Corma A, Iborra S, Primo J. Base catalysis for fine chemicals production: Claisen–Schmidt condensation on zeolites and hydrotalcites for the production of chalcones and flavanones of pharmaceutical. J. Catal. 1995; 151: 60-66.
15. Heindel ND, Reid JR. 4-Amino-3-mercapto-4H- 1,2,4-triazoles and propargyl aldehydes: A new route to 3-R-8-aryl-1,2,4-triazolo[3,4-b]-1,3,4-thiadiazepines. J. Heterocycl. Chem. 1980; 17(5): 1087-88.
16. Holla BS, Kalluraya B, Sridhar KR, Drake E, Thomas LM, Bhandary KK, Levine MS. Synthesis, structural characterization, crystallographic analysis and antibacterial properties of some nitrofuryl triazolo[3,4-b]-1,3,4-thiadiazines. Eur. J. Med. Chem. 1994; 29: 301-08.
17. Mathew V, Keshavayya J, Vidya VP, Acharya, Reddy BM. Heterocyclic system containing bridgehead nitrogen atom: synthesis and pharmacological activities of some substituted 1,2,4-triazolo[3,4-b]-1,3,4-thiadiazoles. Eur. J.Med. Chem. 2006; 41: 1048-58.
18. Budavari S, O'Neil M, Smith A, Heckelman P, Obenchain J. The Merck Index an Encyclopedia of Chemicals, Drugs and Biologicals. 12th ed., Merck Co. Inc.: Rahway, NJ, USA; 1996.
19. Haber J. Present status and perspectives on antimycoties with systematic effects. Cas. Lek. Cesk. 2001; 140: 596-604.
20. Brucato A, Coppola A, Gianguzza S, Provenzano PM. Triazolam: characteristics of its depressive action. Boll. Soc. Ital. Biol. Sper. 1978; 54: 1051-57.
21. Coffen DL, Fryer RI. Proces for preparing triazolo benzodiazepines. U.S. Patent: 3,849,434, 1974.
22. Shiroki M, Tahara T, Araki, K. Jap. Patent: 75100096, 1975.
23. Povelitsa FD, Gural AG. Antibacterial effect of furacriline on Brucella. Antibiotiki Moscow. 1973; 18: 71.