Environmental and Convenient Construction Strategy to Obtained New Supreme Series of Fused 1,2,4-Triazalo System Incorporating N-Acetyl Quanazole

Muna G. Thahee[1], Shaymaa K. Younis[2]

[1]*University of Mosul, College of Science, Department of Chemistry, E-mail: mona.23scp84@student.uomosul.edu.iq*
[2]*University of Mosul, College of Science, Department of Chemistry,*

KEYWORDS

N-acetyl guanozole , 1,2,4-triazolopyrimidine-1,2,4-triazolotriazine , MWI , grinding , Schiff base

ABSTRACT

The presentation goal is to develop new procedures for regioselective synthesis of unusal series of heterocyclic compounds (2-8) derived from N-acetyl guanazole (1) which received a great deal of attention due to it's unique properties. N-acetyl guaunazole used as active precursor to obtained two type of fused 1,2,4-triazole system. The first one including grinding for (1 min.) of a mixture consist of N-acetyl guanazole , malononitrile and substituted benzaldehlyde through direct one-pot multicomponent reaction to achieve a new series of 1,2,4-trazolo[1,5-a,4,3-a]bispyrimidine (2-7). Whereas, the second one involved firstly the conversion of N-acetyl gluanazole to the corresponding Schiff base (8-13) accelerated by microwave irradiation (MWI) for few minutes, then they will underwent intercyclization reaction with phenyl isothiocynate and also acceclerated by grinding then by microwave irradiation to afford 1,2,4-triazolo[1,5-a,4,3-a] bistriazine (14-18).

1. Introduction

N-acetyl guanzole is anitrogen rich five membered ring that become an important fine chemical intermediate consisting wide applications in medical and pharmaceutical and biological fields in addition to the organic synthesis intermediate . It was also recommended as anti-cancer(1,3) ,anti-convulsant(4,5) , anti-viral(6,7) , anti-cytokine(8,9) , anti-hypertensive and anti-inflammatory(10,11) . This type of compounds shown to be promising building block in the design of high performance energetic materials represented by fused 1,2,4-triazolo systems, which are well known class of aza bridgehead fused heterocyclic compound that have miscellaneous pharmaceutical , medical, biological and organic , synthetic application which have previously been reported(12,16) . These type of heterocyclic compound occupy a central position in modern heterocyclic chemistry , because they form an important recognition element in biological active molecule(17,20) . In this work and according to the advantages mentioned above , we prepared two types of bridgehead fused 1,2,4-triazolo system in two different pathways. The first one using grinding one-pot three component reaction for (one minute) among N-acetyl guanazole , malononitrile and substituted benzaldehlyde in ratio (1:2:2) to achieve fused 1,2,4-triazolo[1,5-a,4,3-a] bis pyrimidine derivatives (2-7). Wherease , the second pathway eas included two steps , firstly the N-acetyl guanazole converted to it՚s corresponding Schiff base derivatives (8-12) accelerated via microwave irradiation for few minutes. Then these bases were underwent intercyclization reaction using grinding and microwave irradiation with phenyl isothiocynate in ratio (1:2) to afford new fused 1,2.4-triazolo[1,5-a,4,3-a]bis 1,3,5-triazine derivatives (14-17).

Experimental

<u>Synthesis of N-acetyl guanazole(1):</u>(25)

This compound was prepared according to the literature in ratio (1:1) between hydrazine hydrate and cynoguanidine in acidic media from dil. Hydrochloric acid and it has conformity to the literature in all it's properties constant. Violet powder , M.p =303°C , Yield =91%.

Table (1) : Spectral data for compound (1)

Comp. No.	**FT-IR (KBr), υ (cm^{-1})**				**^{1}H-NMR, δ (ppm)**
	NH_2	**CH_3**	**C=O**	**C=N**	**CH_3=2.46(s,3H) ; NH_2=3.78(s,2H) ; NH=7.42(s ,2H).**

1	**339 0**	**2934**	**171 0**	**1640**	

Synthesis of fused 1,2,4-triazolo[1,5-a,4,3-a] bis pyrimidine (2-7)(24)

In small mortar a mixture of (0.001 mole \ 0.141 gm) of compound (1) , malononitrile (0.002 mole \ 0.132gm) and sub. Benzaladehyde (0.002 mole) , was grinding very well in basic media from triethylamine for (1minute). The completeness of reaction was traced by using T.L.C technique. Then , the formed colored solid mass was washed thoroughly with cold water followed by drying and recrystallized from DMSO to offered compounds (2-7) with physical properties and spectral data listed in TableS (2) and (3) respectively.

Table (2) : Physical properties for compound (2-7)

Comp. No.	**X**	**Molecular Formula**	**M.Wt.**	**I.P. °C**	**ield%**	**Colour**	**R_f Ethanol**
2	H	$C_{29}H_{19}N_9O$	379	75-77	84	Yellow	0.45
3	m-OCH_3	$C_{26}H_{23}N_9O_3$	439	99-101	81	white	0.68
4	o-Cl	$C_{24}H_{17}ClN_9O$	517	73-74	76	Green	0.71
5	p-Me_2N	$C_{28}H_{29}N_{11}O$	535	72-173	90	Orange	0.52
6	m-OH	$C_{24}H_{19}N_9O_3$	481	34-136	79	Brown	0.31
7	Piperonal	$C_{26}H_{19}N_9O_5$	537	82-183	95	Yellow	0.66

Table (3): Spectral data for compound (2-7)

Comp. No.	**FT-IR (KBr), *υ (cm^{-1})***							**^{1}H-NMR, δ (ppm)**
	NH_3	**CH_3**	**CN**	**C=O**	**C=N**	**C=C**	**Other**	
2	3198	2925	2223	1676	1589	1566	C-O-C asym.=1277 sym.=1038	—
3	3142	2937	2227	1681	1594	1569	—	—
4	3164	2941	2224	1698	1584	1558	C-Cl=756	CH_3-C=O :2.24 (s,3H) ; $2NH_2$:3.24 (s,4H) ; Ar-H :7.58-8.05 (m-8H) ; Pyrimidine-H :8.70 (s,2H) .
5	145	2918	2208	1642	1610	1563	—	CH_3-C=O :2.61 (s,3H); $2N(CH_3)$:3.12 (s,12H) ; $2NH_2$: 3.33 (s,4H) ; A-H(AB-System) :6.97 and 7.95 (d-d , 8H) ; pyrimidine-H :8.05(s,2H).
6	301	2934	2239	1711	1643	1615	OH=3389	—
7	373	2929	2225	1645	1614	1569	C-O-C asym.=1291 sym.=1033	—

Synthesis of 3,5-diarylidine-N-acetyl-1,2,4-triazole(8-12)(22)

In beaker (25 ml) , a mixture of compound (1) (0.001 mole \ 0.141gm) and sub. Benzaldehyde (0.002 mole) in acidic media from glacial acetic acid mixed with (2 ml) DMSO, was irradiated in microwave oven for (15 minutes) at (500 watt) . The completeness of the reaction was traced via T.L.C technique .The formed solid mass was washed with cold water (4×5 ml) , followed by drying then recrystallized from DMSO to give the compounds (8-13) with physical properties and spectral data were listed in

Tables (4) and (5) respectively .

Table (4) : Physical properties for compound (8-13)

Comp. No.	**X**	**Molecular Formula**	**M.Wt.**	**M.P. °C**	**Yield%**	**Colour**	**R_f Benzene :MeOH**
8	m-NO_2	$C_{18}H_{15}N_7O_5$	409	185−186	85	Green	0.29
9	p-OCH_3	$C_{20}H_{21}N_5O_3$	439	97−98	81	brown	0.31
10	p-Cl	$C_{18}H_{15}Cl_2N_5O$	387	153−155	89	Yellow	0.40
11	Piperonal	$C_{20}H_{15}N_5O_5$	405	133-134	89	black	0.35
12	m-OH	$C_{18}H_{17}N_5O_3$	351	101-102	73	Greenish brown	0.45
13	p-Me_2N	$C_{20}H_{27}N_7O$	381	185-186	91	brown	0.62

Table (5): Spectral data for compound (8-13)

Comp. No.	FT-IR (KBr), *υ (cm^{-1})*					^{1}H-NMR, δ (ppm)
	CH_3	C=O	C=N Cycl.	C=N Acycl.	Other	
8	2927 2872	1700	1612	1574	NO_2 asym.=1528 sym.=1350	—
9	2932 2837	1682	1597	1568	C-O-C asym. = 1251 sym. =1024	CH_3-C=O :2.23 (s,3H) ; 2OCH_3 :2.55(s,6H) ; Ar-H :7.81-8.70 (m,8H) ; N=CH :8.71 (s,1H) , N=CH :10.16 (s,1H).
10	2920 2810	1691	1613	1593	C-Cl=823	CH_3-C=O :2.55 (s,3H) ; Ar-H :7.13-7.50 (m,8H) ; N=CH :7.63(s,1H) , N=CH :7.89 (s,1H).
11	2920 2873	1693	1611	1525	—	—
12	2921 2851	1682	1581	1556	OH=3323	—
13	2901 2787	1682	1614	1586	C-O-C asym.=1252 ssym.=1032	—

Synthesis of fused 1,2,4-triazolo[1,5-a,4,3-a] bis1,3,5-triazine derivaties(14-18) :(21)(23)

A mixture in ratio (1:2) from compound (8-12) and phenyl iso thiocynate was grinding thoroughly for (5 minutes) in solid phase , then it was put in beaker (25 ml) followed by irradiation by microwave oven for (6 minutes) at (450 watt) .Cooling then test the completeness of the reaction via T.L.C technique followed by washing with cold-water (4×5) , dried and recrystallized from ethanol to afford the compounds (14-18) with physical properties and spectral data listed in Tables (6) and (7) respectively.

Table (6) : Physical properties for compound (14-18)

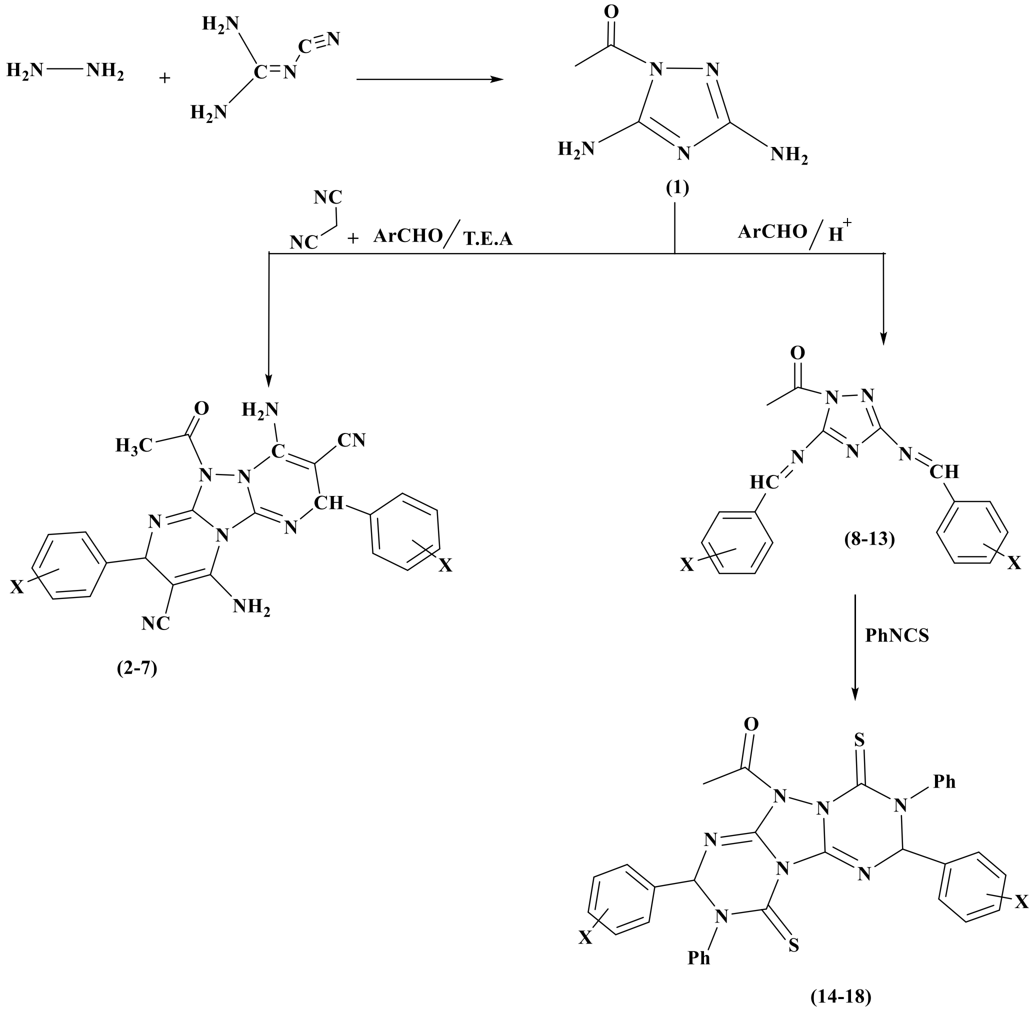
Comp. No.	**X**	**Molecular Formula**	**M.Wt**	**M.P. °C**	**Yield %**	**Colour**	**Rf Benzene :MeOH**
14	m-NO_2	$C_{28}H_{17}N_8O_5S$	541	76−77	92	Green	0.39
15	p-OCH_3	$C_{27}H_{20}N_6O_3S$	508	55−56	79	Green	0.45
16	p-Cl	$C_{25}H_{17}Cl_2N_6OS$	519	87−88	74	White	0.56
17	Piperonal	$C_{27}H_{20}N_6O_5$	508	85-86	96	black	0.35
18	m-OH	$C_{25}H_{18}N_6O_3$	450	79-80	87	Violet	0.41

Table (7): Spectral data for compound (14-18)

Comp. No.	FT-IR (KBr), υ (cm^{-1})				^{1}H-NMR, δ (ppm)
	C=O	C=N	C=S	Others	
14	1688	1611	1013	NO_2 asym.1523 sym.1345	–
15	1678	1598	1015	C-O-C asym.1242 sym.1162	CH_3-C=O:2.42(s,3H),Fused triozino-H:3.27(s,1H);Ar-H(AB System):7.56 &7.95(d-d,8H),Pheny:7.27-7.95(m,10H).
16	1682	1584	1032	C-Cl=694	CH_3-C=O:2.60(s,3H); 2OCH_3:2.77(s,6H),Fused triazino-H:3.87(s,1H),Ar-H(AB System:7.56 &793(d-d,8H);Phenyl:7.04-7.51(m,10H).
17	1690	1583	1020	C−O−C asym.1247 sym.1088	–
18	1689	1614	1012	OH=3352	–

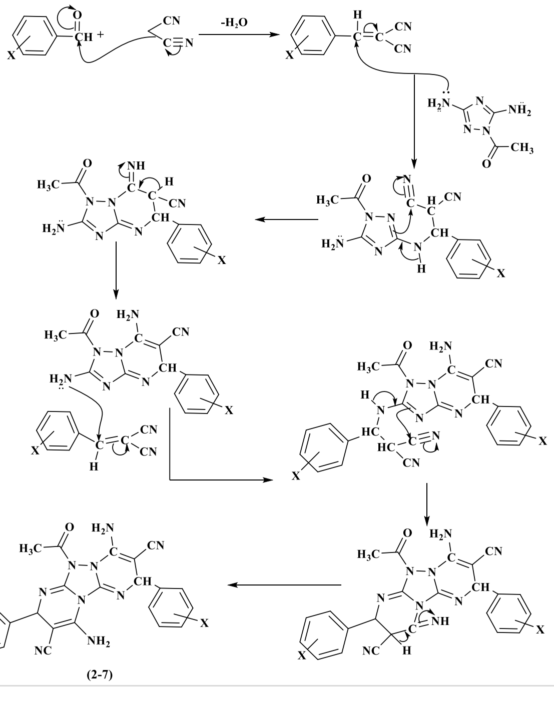
2. Result and Discussion

Environmental and convenient strategy that used in this work to affored new fused 1,2,4-triazalo system (2-18) were proceeded through the general synthetic pathway below :



Scheme (1): Synthetic pathway for compounds (1-18)

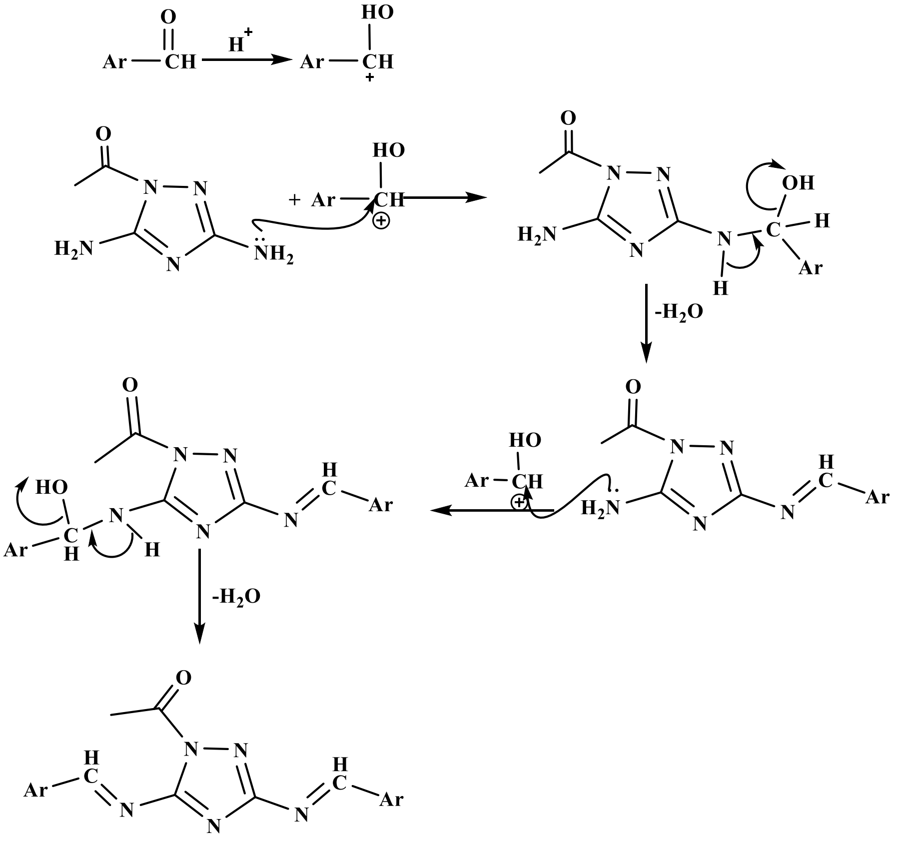
One-pot multicomponent reaction was proceeded through direct sequence addition reaction among N-acetyl guanazole , malonbnitrile and substituted Benzaldehyde in basic media and also we believed that the reaction proceeded through the following suggested mechanism , Scheme (2)



Scheme (2): Synthetic mechanism for compounds (2-7)

The formed compounds (2-7) were identified via spectrophotometric methods, so they shown in FT-IR strong stretching vibrating bond at υ cm^{-1} (3142-3373) refer to primary amino group and at (2208 -2239) refer to cyno group which gave strong evidence that support the suggested structure in addition to the other absorption bonds listed in Table (3). On the other hand, in ^{1}H-NMR spectroscopy, compounds (4 and 5) were given as example for this series , shown absorption peak listed in Table (3) . It is worth noting that compound (5) gave clear AB-system as (d-d) peak at δ ppm (6.97 and 7.95) refer to (8H) in symmetric molecule for phenyl ring with para substituent. Actually, all these peaks came in agreement with suggested structure.

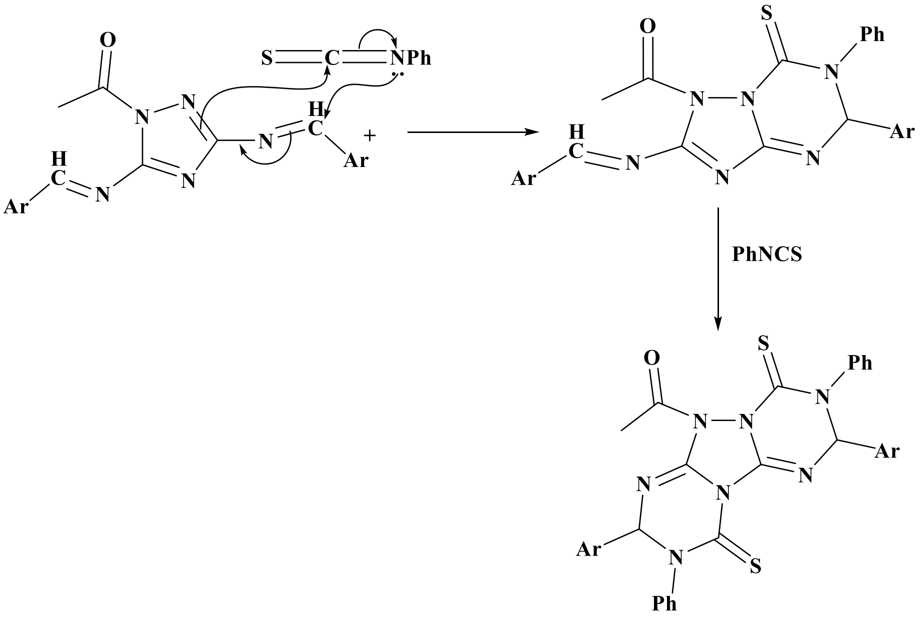
The second part in this presentation was convert the compound (1) to the corresponding Schiff bases (8-13) through it's reaction with substituted benzaldehyde in acidic media accelerated via microwave irradiation for only few minutes, Scheme (3). These bases were characterized spectrally, so they shown in FT-IR spectroscopy strong stretching vibrational bands at υ cm^{-1} (1581-1613) refer to the C=N functional group with the absence of the NH_2 stretching vibration bands. This gave an intial inducation of the validity of the proposed structure. wherease, in ^{1}H-NMR spectroscopy the compounds (9 ,10) as examples for this series gave clear evidence of the validity of the proposed composition, It gave two identical peaks at δ (ppm) :8.71 ,7.63 (s ,1H) and 10.16 , 7.89 (s , 1H) refer to the two CH=N in different postions in the molecule , and also this spectrum came in agreement with suggested composition .



(8-13)

Scheme (3): Synthetic mechanism for compounds (8-13)

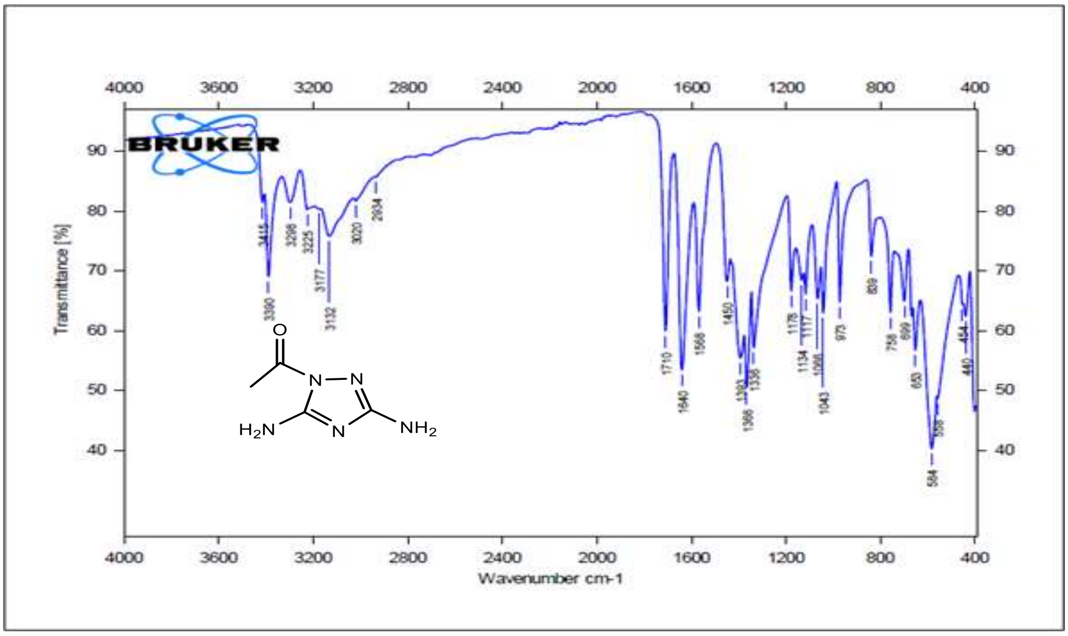
Finally , compounds (8-13) were underwent intercyclization reaction with phnyl isothiocynate in ratio (1:2). The reaction was proceeded throung grinding for few minutes then irradiated by microwave oven at power (500 watt) for (15 minutes), the following Scheme (4) describes the steps for the preparation of such compounds.

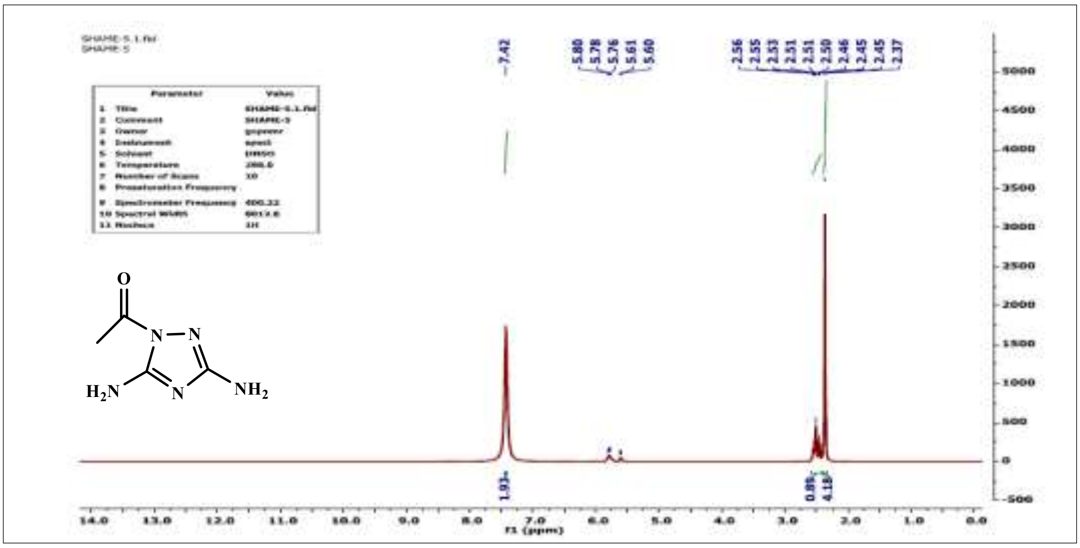


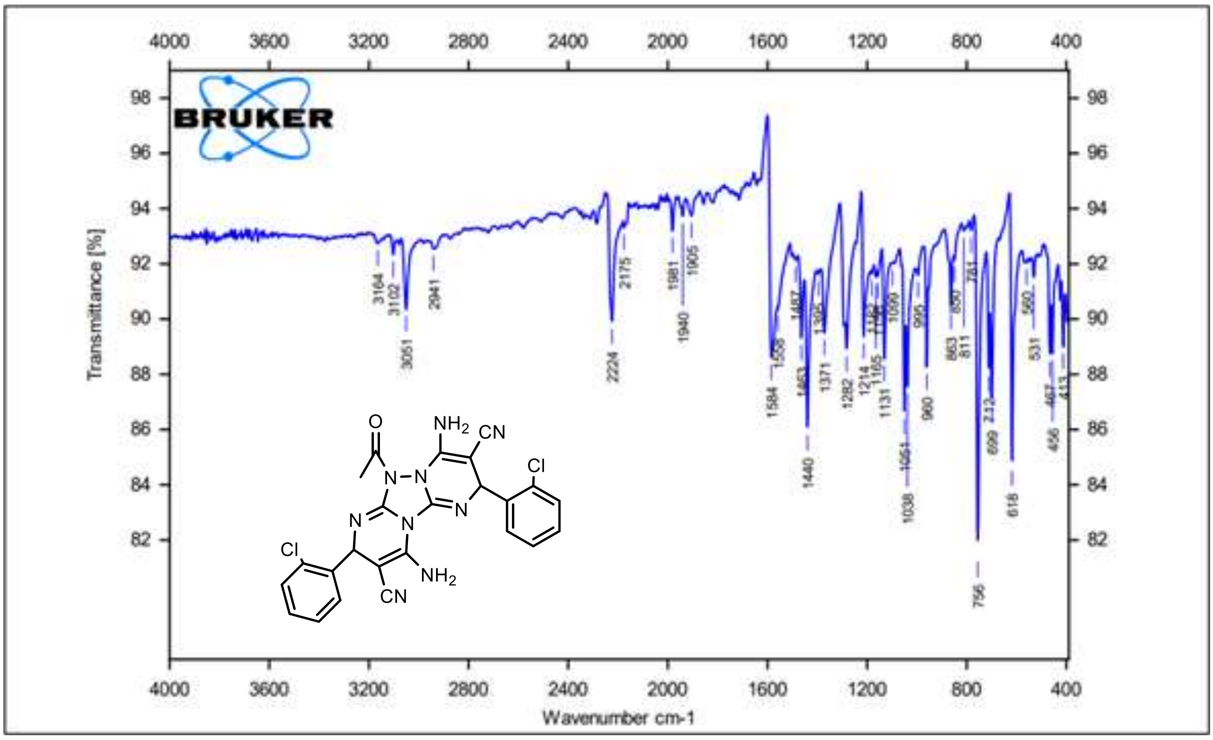
(14-18)

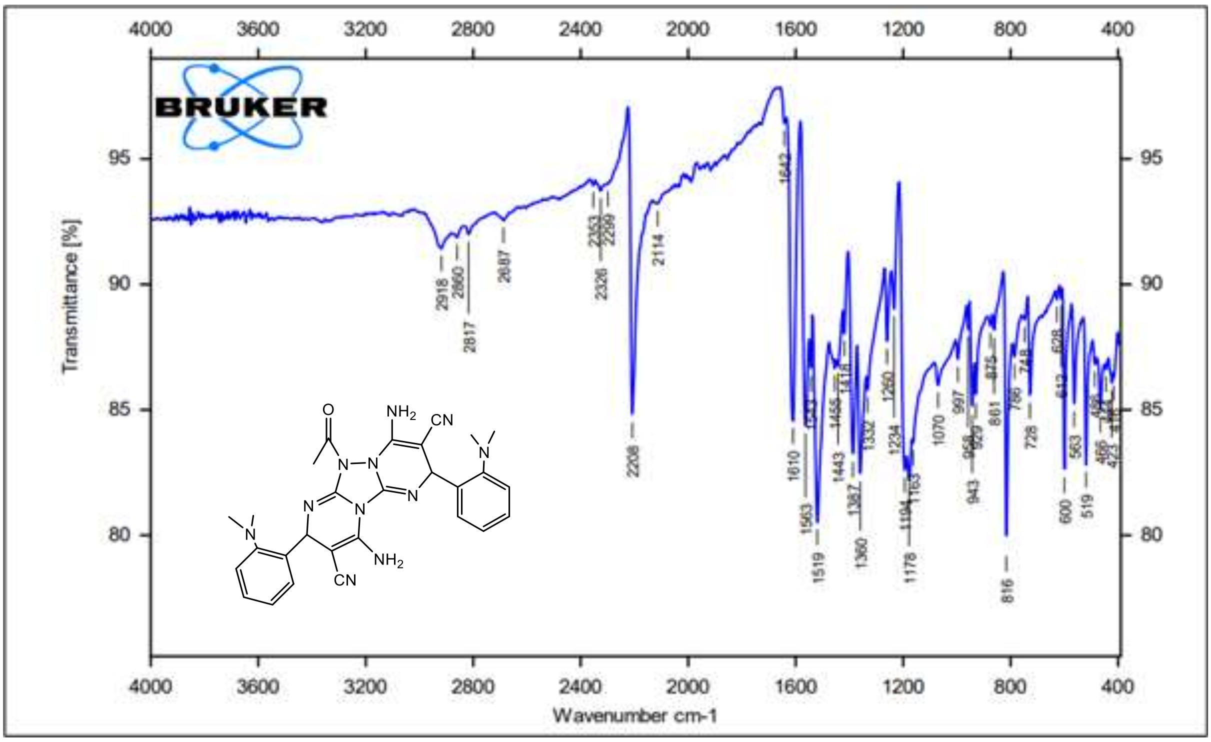
Scheme (4): Synthetic mechanism for compounds (14-18)

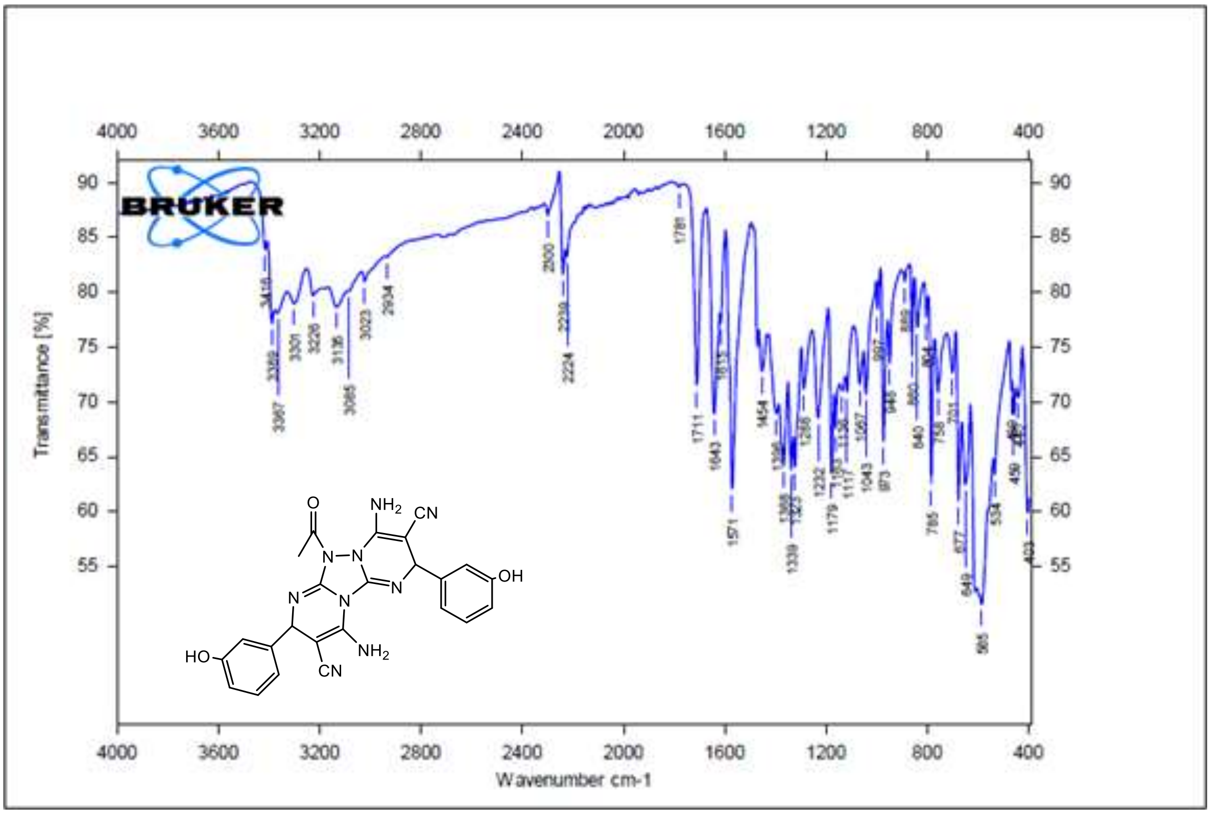
Basically , these compound shown in FT-IR spectroscopy stretching vibrational bands at υ cm^{-1} (1012-1032) refer to the (C=S) functionl groups in addition to the other absorption bands listed in Table (7) . While in ^{1}H-NMR spectroscopy , the compounds (14,15) as example for this series , gave clear AB-system at δ ppm. 7.56 and 7.93(d-d , 8H) ,7.56 and 7.97 (d-d,8H) . repectively with the absonce of CH=N absorption peak . Actually , these spectroscopic images proved the validity of the poposed structure and thus the aim of this work was successfully achieved .

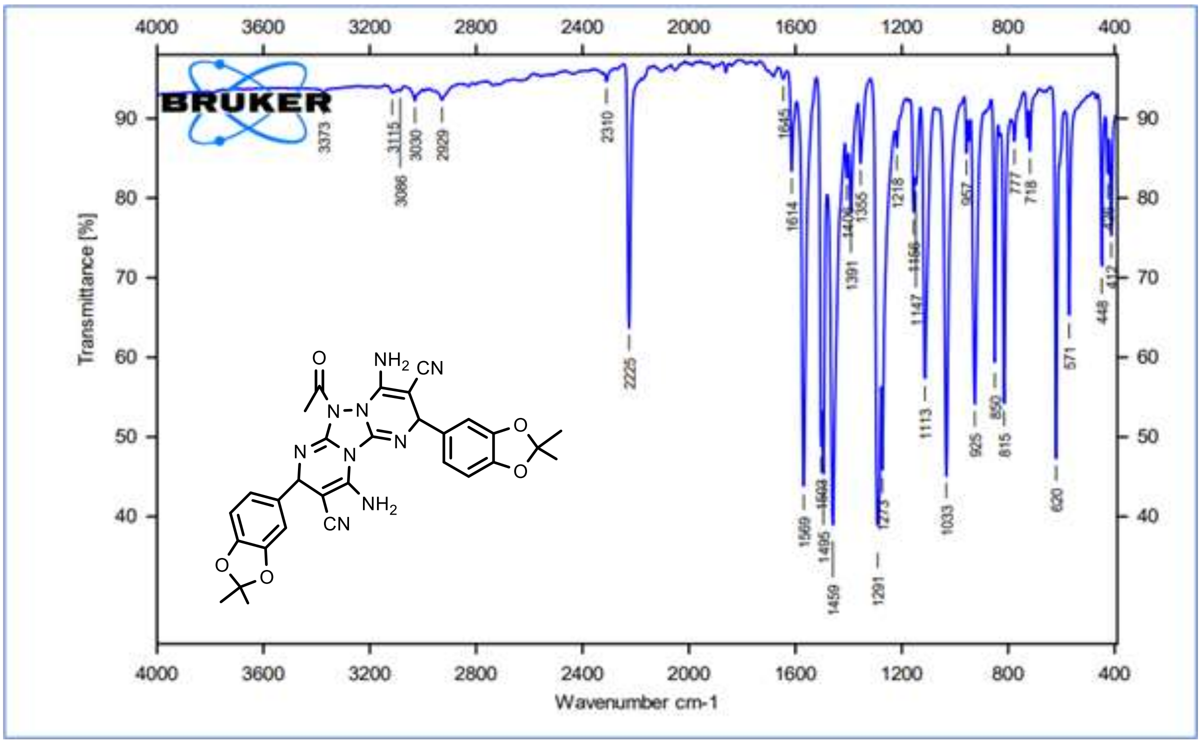


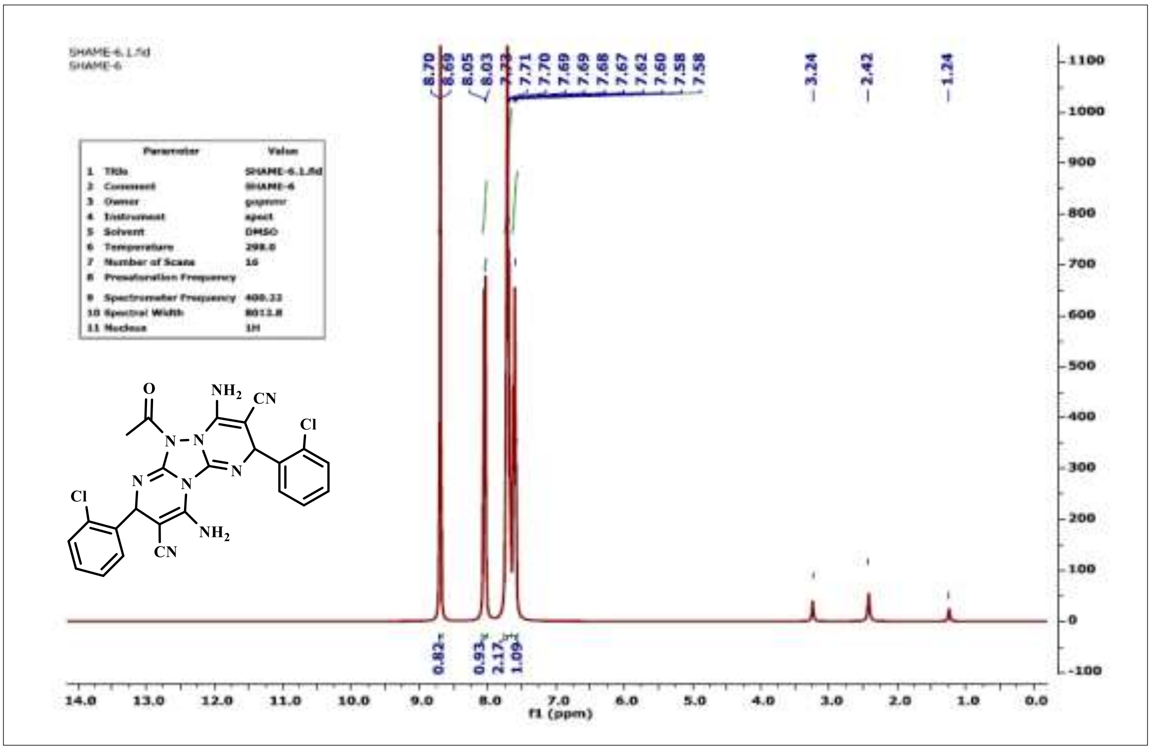

SHAME-5.1.fid
SHAME-5
Parameter
Value
1 Title
SHAME-5.1.fid
2 Comment
SHAME-5
3 Owner
gopanr
4 Instrument
spect
5 Solvent
DMSO
6 Temperature
298.0
7 Number of Scans
10
8 Presaturation Frequency
9 Spectrometer Frequency
400.22
10 Spectral Width
6013.6
11 Nucleus
1H
7.42
5.80
5.78
5.76
5.61
5.60
2.56
2.55
2.53
2.51
2.51
2.50
2.46
2.45
2.45
2.37
1.93
0.89
4.18
O
N
N
N
H2N
NH2
5000
4500
4000
3500
3000
2500
2000
1500
1000
500
0
-500
14.0
13.0
12.0
11.0
10.0
9.0
8.0
7.0
6.0
5.0
4.0
3.0
2.0
1.0
0.0
f1 (ppm)


BRUKER
Transmittance [%]
Wavenumber cm-1
4000
3600
3200
2800
2400
2000
1600
1200
800
400
98
96
94
92
90
88
86
84
82
3164
3102
3051
2941
2224
2175
1981
1940
1905
1584
1558
1487
1463
1440
1396
1371
1282
1214
1182
1165
1131
1109
1061
1038
995
960
863
850
811
781
756
712
699
618
560
531
467
456
413
O
NH2
CN
N
N
N
N
N
N
Cl
Cl
NH2
CN

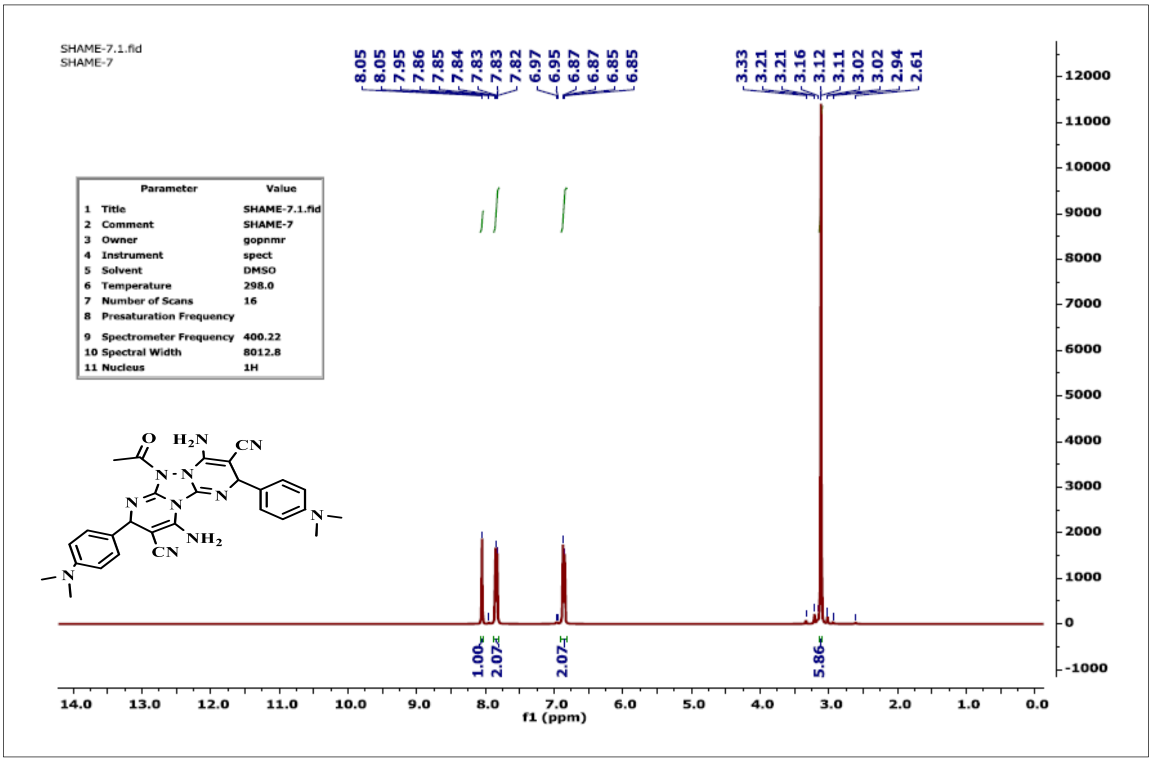

BRUKER
Transmittance [%]
Wavenumber cm-1
4000
3600
3200
2800
2400
2000
1600
1200
800
400
95
90
85
80
2918
2860
2817
2687
2353
2326
2299
2208
2114
1642
1610
1563
1543
1519
1455
1443
1418
1387
1360
1332
1260
1234
1194
1178
1163
1070
997
958
943
929
875
861
816
786
748
728
628
613
600
563
519
486
466
423
O
NH2
CN
N
N
N
N
N
N
N
NH2
CN


BRUKER
Transmittance [%]
Wavenumber cm-1
4000
3600
3200
2800
2400
2000
1600
1200
800
400
90
85
80
75
70
65
60
55
3419
3389
3367
3301
3226
3136
3085
3023
2934
2300
2239
2224
1781
1711
1643
1613
1571
1454
1386
1368
1339
1323
1288
1232
1179
1153
1136
1117
1067
1043
997
973
948
889
860
840
804
785
758
701
677
649
585
534
459
403
O
NH2
CN
OH
HO
N
N
N
N
N
N
NH2
CN

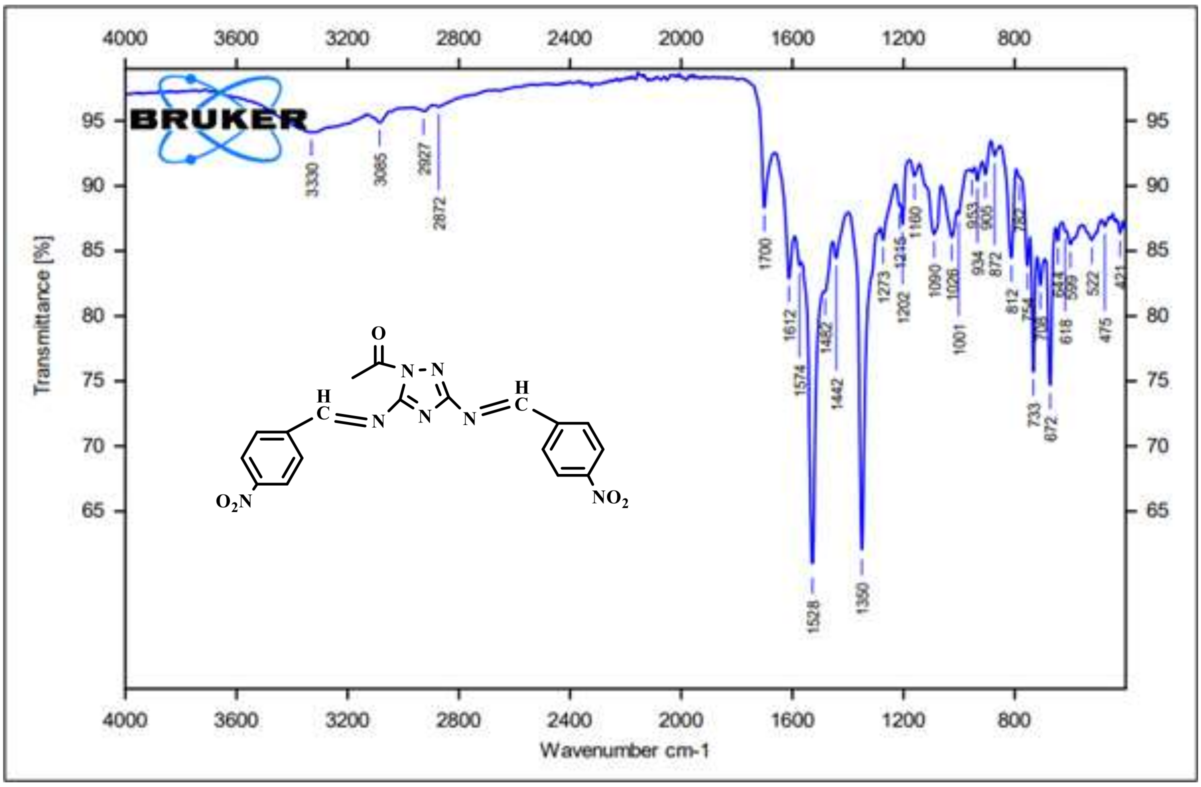


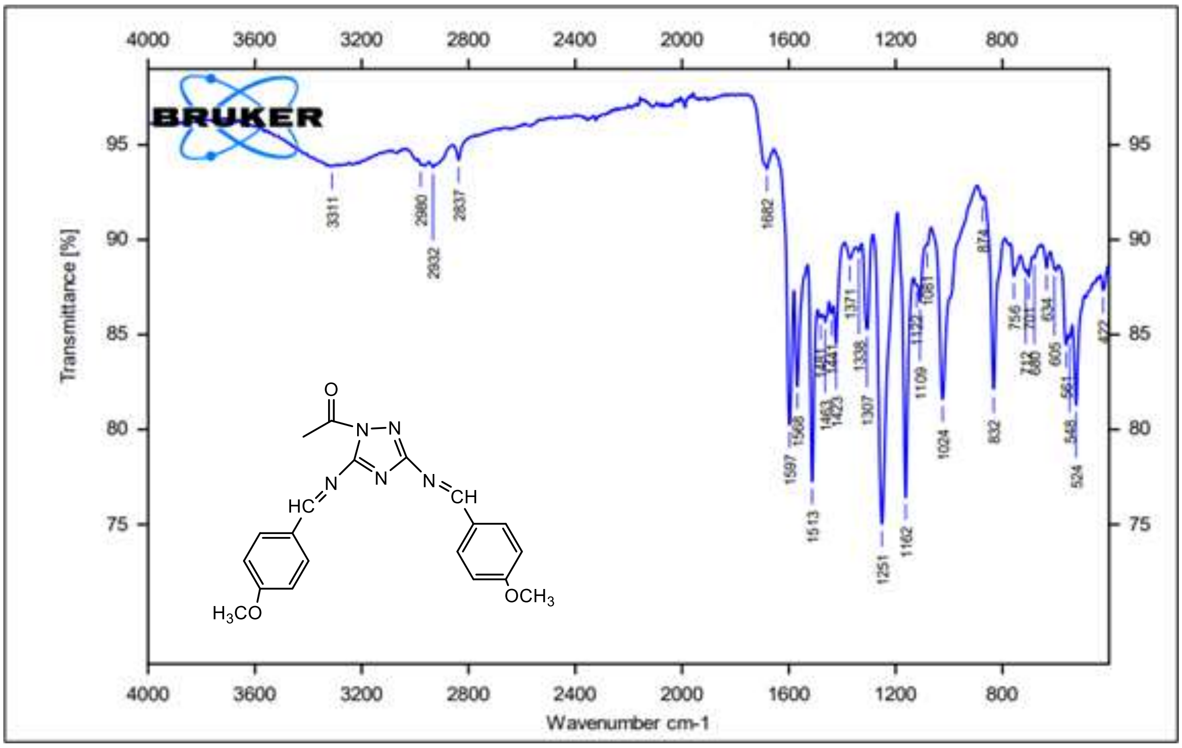


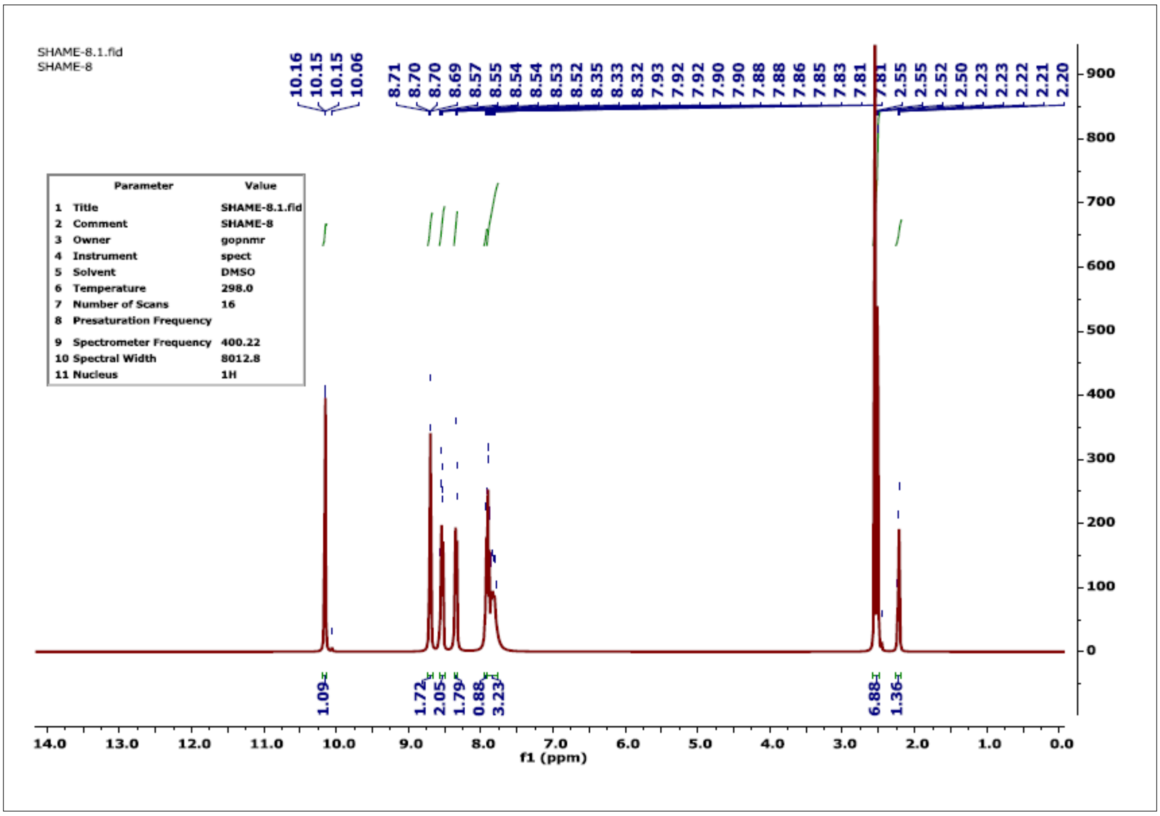
	Parameter	Value
1	Title	SHAME-6.1.fid
2	Comment	SHAME-6
3	Owner	gsgnmr
4	Instrument	spect
5	Solvent	DMSO
6	Temperature	298.0
7	Number of Scans	16
8	Presaturation Frequency	
9	Spectrometer Frequency	400.22
10	Spectral Width	8012.8
11	Nucleus	1H



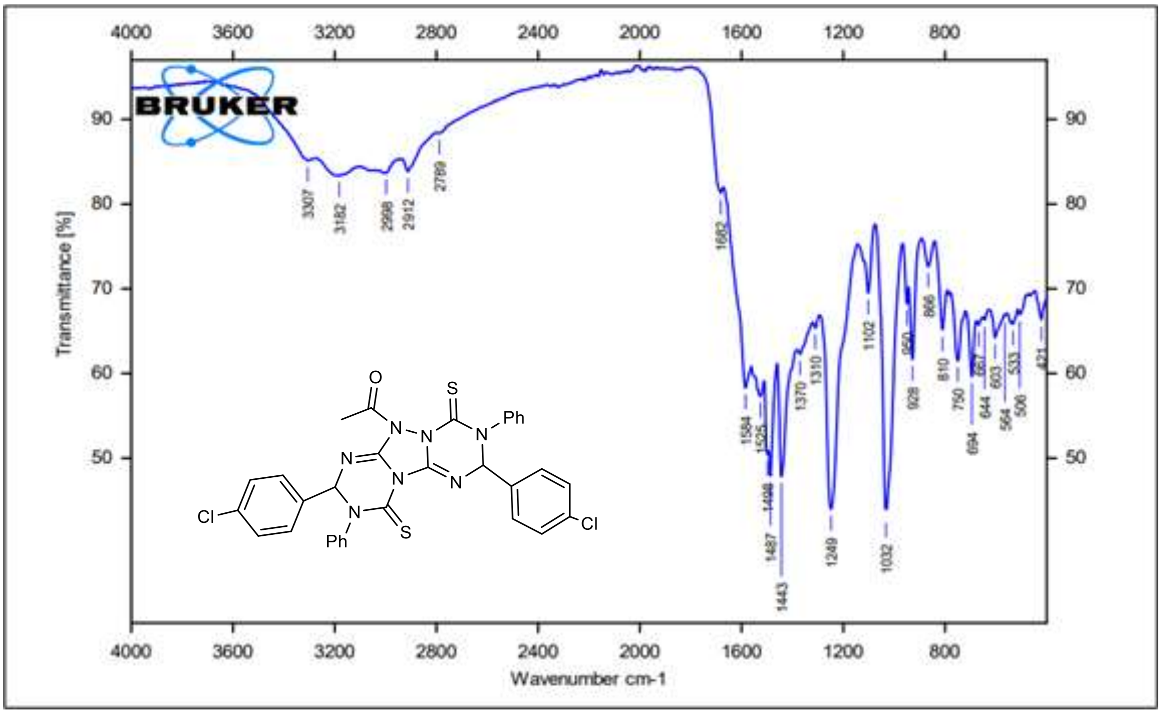
	Parameter	Value
1	Title	SHAME-7.1.fid
2	Comment	SHAME-7
3	Owner	gopnmr
4	Instrument	spect
5	Solvent	DMSO
6	Temperature	298.0
7	Number of Scans	16
8	Presaturation Frequency	
9	Spectrometer Frequency	400.22
10	Spectral Width	8012.8
11	Nucleus	1H

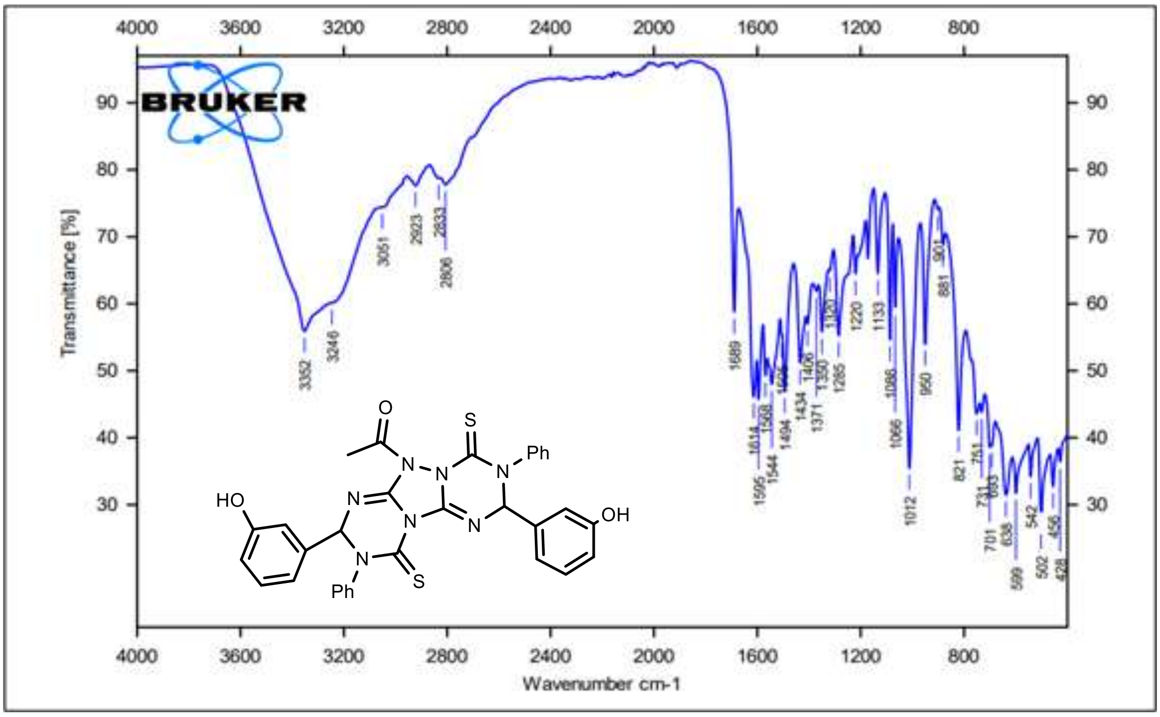


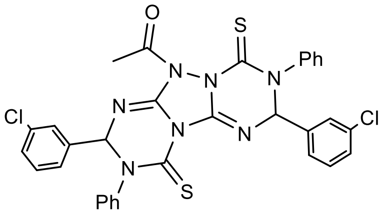


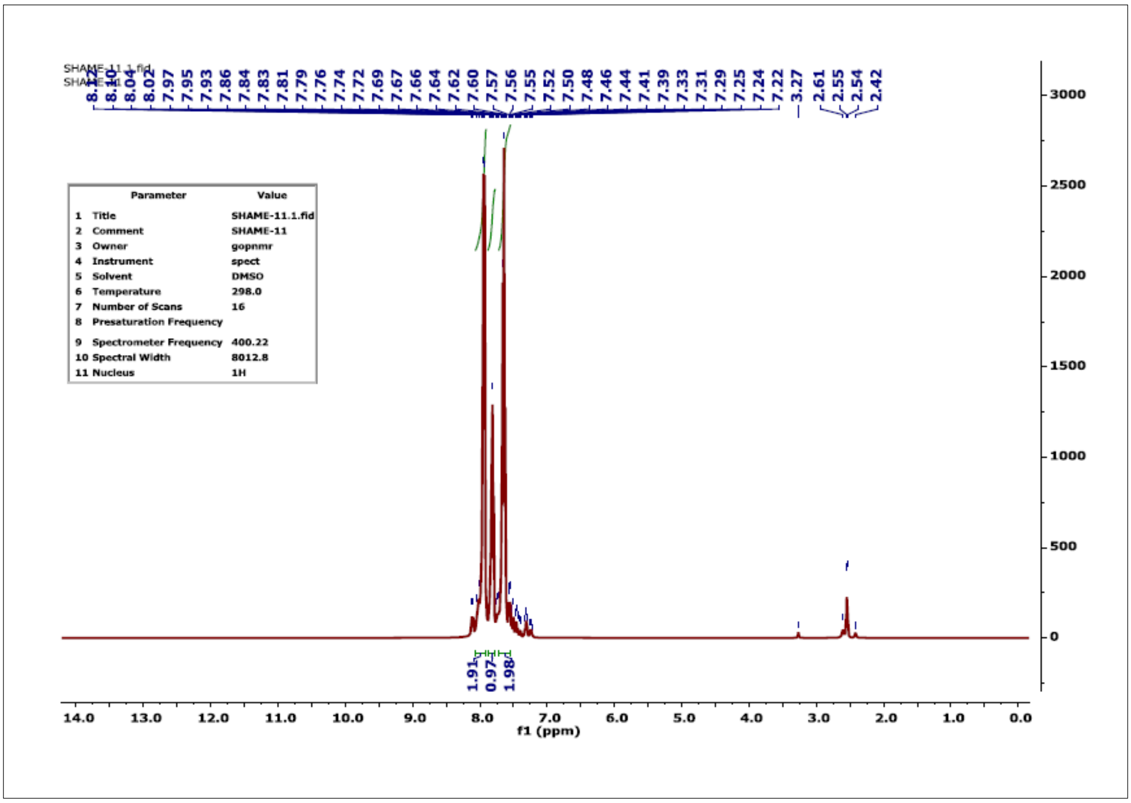


	Parameter	Value
1	Title	SHAME-8.1.fid
2	Comment	SHAME-8
3	Owner	gopnmr
4	Instrument	spect
5	Solvent	DMSO
6	Temperature	298.0
7	Number of Scans	16
8	Presaturation Frequency	
9	Spectrometer Frequency	400.22
10	Spectral Width	8012.8
11	Nucleus	1H

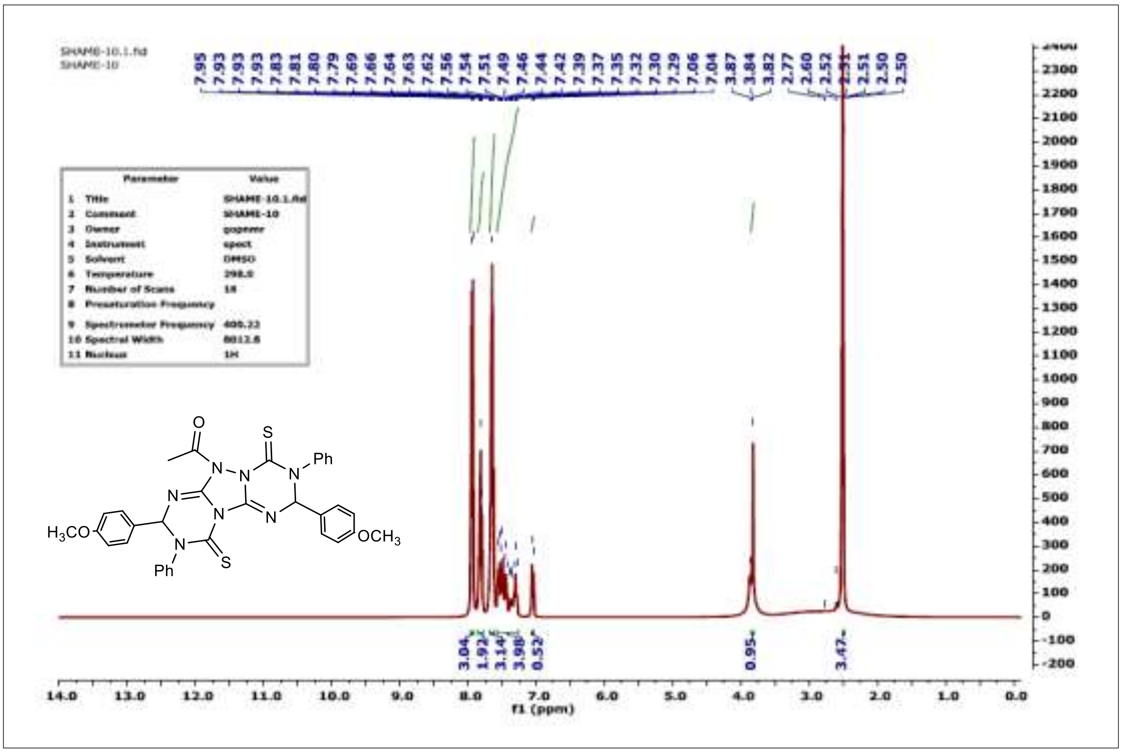

BRUKER
Transmittance [%]
Wavenumber cm-1
4000
3600
3200
2800
2400
2000
1600
1200
800
90
80
70
60
50
3307
3182
2998
2912
2789
1682
1584
1525
1498
1487
1443
1370
1310
1249
1102
1032
950
928
866
810
750
694
667
644
603
564
533
506
421
O
S
N
N
N
Ph
N
N
N
N
Cl
Cl
Ph
S


BRUKER
Transmittance [%]
Wavenumber cm-1
4000
3600
3200
2800
2400
2000
1600
1200
800
90
80
70
60
50
40
30
3352
3246
3051
2923
2833
2806
1689
1614
1595
1508
1544
1494
1466
1434
1406
1371
1350
1320
1265
1220
1133
1086
1066
1012
950
901
881
821
761
731
683
701
638
599
542
502
456
428
O
S
N
N
N
Ph
N
N
N
N
HO
OH
Ph
S


O
S
N
N
N
Ph
N
N
N
N
Cl
Cl
Ph
S



	Parameter	Value
1	Title	SHAME-11.1.fid
2	Comment	SHAME-11
3	Owner	gopnmr
4	Instrument	spect
5	Solvent	DMSO
6	Temperature	298.0
7	Number of Scans	16
8	Presaturation Frequency	
9	Spectrometer Frequency	400.22
10	Spectral Width	8012.8
11	Nucleus	1H



	Parameter	Value
1	Title	SHAME-10.1.fid
2	Comment	SHAME-10
3	Owner	gopnmr
4	Instrument	spect
5	Solvent	DMSO
6	Temperature	298.0
7	Number of Scans	16
8	Presaturation Frequency	
9	Spectrometer Frequency	400.22
10	Spectral Width	8012.8
11	Nucleus	1H

3. Conclusion and future scope

Selective and environmental strategy have been successfully applied to achieve new fused 1,2,4-triazole system using green chemistry techniques represental by the grinding and microwave irradiation which reducing the reaction time with yield enhancement, selective, very ecomonmical and environmentally friendly with proven the synthetic formula.

Reference

[1] Abdulmajeed, A. P. M. A. (2022). Crying and its repercussions in the group (the Fedayee and the Beast) by Amal Al-Zahawi: an objective approach. *The Arab Gulf*, *50*(2).

[2] Meshal Al-Jahdali, S Salem Bagabas, M., Mamdoh Al-Manabre, L., Zahi Al-Sofee, R., Yousef Rednah, R., & Ahmad Sindi, R.,. (2022). [Retracted] Evaluation of Knowledge, Attitude, and Practice of Health Practitioners towards Fertility Preservation in Cancer Patients in an Environmental Region of Saudi Arabia. *Journal of Environmental and Public Health*, *2022*(1), 6404837

[3] Marcelino, K. R., Ling, L., Wongkiew, S., Nhan, H. T., Surendra, K. C., Shitanaka, T., ... & Khanal, S. K. (2023). Nanobubble technology applications in environmental and agricultural systems: Opportunities and challenges. *Critical Reviews in Environmental Science and Technology*, *53*(14), 1378-1403.

[4] Grover, G., Nath, R., Bhatia, R., & Akhtar, M. J. (2020). Synthetic and therapeutic perspectives of nitrogen containing heterocycles as anti-convulsants. *Bioorganic & Medicinal Chemistry*, *28*(15), 115585.

[5] Pandya, V. G., & Mhaske, S. B. (2022). Construction of tetrahydrobenzo [f] quinoline scaffolds via polar [4+ 2]-Cycloaddition reaction with arynes as dienophiles. *Tetrahedron Letters*, *101*, 153901.

[6] Zarenezhad, E., Farjam, M., & Iraji, A. (2021). Synthesis and biological activity of pyrimidines-containing hybrids: Focusing on pharmacological application. *Journal of Molecular Structure*, *1230*, 129833.

[7] Finger, V., Kufa, M., Soukup, O., Castagnolo, D., Roh, J., & Korabecny, J. (2023). Pyrimidine derivatives with antitubercular activity. *European Journal of Medicinal Chemistry*, *246*, 114946

[8] Verma, T., Sinha, M., & Bansal, N. (2020). Heterocyclic compounds bearing triazine scaffold and their biological significance: A review. *Anti-Cancer Agents in Medicinal Chemistry (Formerly Current Medicinal Chemistry-Anti-Cancer Agents)*, *20*(1), 4-28.

[9] Joule, J. A., Mills, K., and Smith, G. F. (2020). **Heterocyclic Chemistry**. CRC

[10] Bosica, G., Cachia, F., De Nittis, R., & Mariotti, N. (2021). Efficient one-pot synthesis of 3, 4-dihydropyrimidin-2 (1 H)-ones via a three-component Biginelli reaction. *Molecules*, *26*(12), 3753.

[11] Oliveira, A. S., Rolo, J., Gaspar, C., Cavaleiro, C., Salgueiro, L., Palmeira-de-Oliveira, R., ... & Palmeira-de-Oliveira, A. (2022). Chemical characterization and bioactive potential of Thymus× citriodorus (Pers.) Schreb. preparations for anti-acne applications: Antimicrobial, anti-biofilm, anti-inflammatory and safety profiles. *Journal of ethnopharmacology*, *287*, 114935.

[12] Arora, N., Dhiman, P., Kumar, S., Singh, G., & Monga, V. (2020). Recent advances in synthesis and medicinal chemistry of benzodiazepines. *Bioorganic Chemistry*, *97*, 103668.

[13] Łowicki, D., & Przybylski, P. (2022). Cascade synthetic strategies opening access to medicinal-relevant aliphatic 3-and 4-membered N-heterocyclic scaffolds. *European Journal of Medicinal Chemistry*, *238*, 114438.

[14] Shoemaker, A. H., & Griffith, D. R. (2021). Synthetic Approaches to Non-Tropane, Bridged, Azapolycyclic Ring Systems Containing Seven-Membered Carbocycles. *Synthesis*, *53*(01), 65-78.

[15] Das, S. (2024). Annulations involving p-benzoquinones: Stereoselective synthesis of fused-, spiro-and bridged molecules. *New Journal of Chemistry*.

[16] Kerru, N., Gummidi, L., Maddila, S., Gangu, K. K., & Jonnalagadda, S. B. (2020). A review on recent advances in nitrogen-containing molecules and their biological applications. *Molecules*, *25*(8), 1909.

[17] Yang, Z., Wang, T., Chen, H., Suo, X., Halstenberg, P., Lyu, H., ... & Dai, S. (2020). Surpassing the organic cathode performance for lithium-ion batteries with robust fluorinated covalent quinazoline networks. *ACS Energy Letters*, *6*(1), 41-51.

[18] Dze, K. C., & Samad, F. (2020). Heterocycles, their synthesis and industrial applications: a review. *Int J Appl Sci Eng Technol*, *8*, 36-56.

[19] Djebaili, R. (2022). *Structure-activity approaches for prediction of chemical reactivity and pharmacological properties of some heterocyclic compounds* (Doctoral dissertation, Université de mohamed kheider biskra).

[20] Xia, S. B., Cai, Y. Q., Yao, L. F., Shi, J. Y., Cheng, F. X., Liu, J. J., ... & Zheng, J. C. (2022). Nitrogen-rich two-dimensional π-conjugated porous covalent quinazoline polymer for lithium storage. *Energy Storage Materials*, *50*, 225-233.

[21] Younis, S. K. (2010). Synthesis of some New 1, 2, 4, 5-Tetrazine Derivatives Via Diels-Alder Reaction. *Rafidain Journal of Science*, *21*(2), 65-76.

[22] Kadhim, K. J., & Munahi, M. G. (2017). Synthesis, characterization and biological evaluation of some novel Schiff's bases derived from vanillin. *Journal of Global Pharma Technology*, *10*(9), 383-386.

[23] Younis, S. K. (2022). Convenient methodology for Synthesis a New Series of Heterocyclic Compounds Including The Enamine Moiety. *NeuroQuantology*, *20*(9), 3676.

[24] Bhale, P.S., S.B. Dongare, and U.B. Chanshetti, *Simple grinding, catalyst-free, one-pot, three-component synthesis of polysubstituted amino pyrazole.* Research Journal of Chemical Sciences _ ISSN, 2014. **2231**: p. 606X.

[25] Chernyshev, V. M., Rakitov, V. A., Taranushich, V. A., & Blinov, V. V. (2005). Acyl and sulfonyl derivatives of 3, 5-diamino-1-R-1, 2, 4-triazoles. *Chemistry of Heterocyclic Compounds*, *41*, 1139-1146.