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Reaction of Nitrilimines with 2-Aminopicoline, 3-Amino-1,2,4-Triazole, 5-Aminotetrazole and 2-Aminopyrimidine

Rami Y. Morjan[1], Basam S. Qeshta[1], Hussein T. Al-Shayyah[1], John M. Gardiner[2], Basam A. Abu-Thaher[1], Adel M. Awadallah[1*]

[1]Department of Chemistry, Islamic University of Gaza, Gaza, Palestine
[2]Manchester Institute of Biotechnology, School of Chemistry and EPS, The University of Manchester, Manchester, UK
Email: *awada@iugaza.edu.ps

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Abstract

The reaction of picoline derivatives 3-6 with hydrazonoylhalide 1a produced imidazo[1,2-a]pyridines 7-10, while the reaction of the same picoline derivatives with hydrazonoylhalide 1b afforded imidazo[1,2-a]pyridine-2-ones 11-13. The reaction of 1b, c with 3-amino-1,2,4-triazole 14 produced the acyclic adducts 18 and 19, respectively. Reaction of 1b, 1c with 5-aminotetrazole 20 produced the acyclic products 23 and 24, respectively. Finally, the reaction of 1b with 4, 6-dimethyl-2-aminopyrimidine 27 afforded compound 29 rather than its isomeric structure 28. The structure of the products was confirmed by the different spectroscopic analytical methods including IR, MS, ^{1}HNMR and ^{13}CNMR.

Keywords

Nitrilimines, Picolines, Imidazo[1,2-a]Pyridine, Triazoles, Tetrazoles, Pyrimidines

1. Introduction

Nitrilimines—generally generated *in situ* from hydrazonoyl halides are 1,3-dipolar species that are widely used for the synthesis of different heterocyclic systems *via* 1,3-dipolar cycloaddition reactions [1] [2], or cyclocondensation reactions [2]. Nitrilimines with C-acetyl or ester moiety represent a dielectrophilic system that may react with dinucleophilic heterocyclic systems at the carbonyl carbon of the acetyl or ester group, and at the terminal carbon of the dipole leading to fused heterocyclic systems. Recent examples include; the synthesis of im-

*Corresponding author.

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idazo[1,2-a]pyridines [3] [4] and their 2-one derivatives [4] from the reaction of nitrilimines with 2-amino-pyridine and imidazo[1,2-a]pyrazine from the reaction with 2-aminopyrazine [5]. Their reaction with 2-cyanomethylben-zimidazole gave pyrrolo[1,2-a]benzimidazole [6] [7]. The reaction of ethyl pyridine-2-acetate with nitrilimines having a C-acetyl moiety afforded the corresponding pyrrolo[1,2-a]pyridine, while their reaction with nitrilimines having a C-ester moiety afforded a cyclic adducts [8]. Similarly, the reaction of nitrilimines with a C-ester moiety with 2-cyanomethylbenzimidazole, and 2-amniobenzimidazole afforded a cyclic adducts [8]. In this work, we will investigate the reaction of nitrilimines with different amino heterocycles hoping to prepare new fused heterocyclic compounds that may be further tested for their biological activity. The following amino heterocycles will be used; aminopicolines, aminotriazole, aminotetrazole, aminoisoxazole and aminopyrimidine.

2. Materials and Methods

2.1. Experimental

^{1}H-NMR spectra were recorded at 300 or 400 MHz and ^{13}C-NMR spectra at 75 or 100 MHz on a Bruker AC300 or AC400 spectrometer. Chemical shifts are denoted in (ppm) relative to the internal solvent standard, TMS. ES-MS and HRMS were recorded on a Micromass LCT orthogonal acceleration time-of-flight mass spectrometer (positive and negative ion mode) with flow injection via a Waters 2790 separation module autosampler. FTIR spectra were recorded using Shimadzu 8201 spectrophotometer with KBr technique in region 4000 - 400 cm^{-1} that was calibrated by polystyrene. Melting point determinations were made using a Stuart Scientific SMP1 apparatus and are uncorrected. All chemicals and solvents were used without further purification, as supplied by Sigma-Aldrich unless otherwise stated. Hydrazonoylhalides **1a-c** were prepared using Japp-Klingman method according to reported literature procedure [9].

2.1.1. General Procedure for Syntheses of Compounds 7-10 and 11-13

Triethylamine (0.6 g, 0.006 mol) in dry THF (10 ml) was dropwise added to a stirred solution of hydrazonoyl chlorides **1a** or **1b** (0.005 mol) and substituted picolines **3 - 6** (0.65 g, 0.006 mol) in 25 ml THF at room temperature producing imidazopyridines **(7 - 10)** and **(11 - 13)**, respectively. Stirring was continued overnight, and the solvent was then evaporated in *vacuo*. The residual solid was washed with water to remove the triethylamonium salt, and the crude products were then recrystallized from the appropriate solvents to give the title compounds.

1) 3-[(4-Chlorophenyl)diazenyl]-2,8-dimethylimidazo[1,2-a]pyridine 7

Yield 93%, Yellow solid, mp 152˚C - 155˚C; IR (KBr): υ_{max} cm^{-1} = 1490, 1416, 1298, 1282, 1221, 1200, 1096, 811 and 774 cm^{-1}. MS: m/z $C_{15}H_{13}ClN_4$ (284/286 M^+, chlorine isotope effect), HRMS (Calculated 284.0902, Found 284.0898). ^{1}H NMR (300 MHz, DMSO-d_6): δ 9.62 (d, J = 7.0 Hz, 1H), 7.89 (d, J = 8.7 Hz, 2H), 7.60 (d, J = 8.7 Hz, 2H), 7.51 (d, J = 7.0 Hz, 1H), 7.20 (t, J = 7.0 Hz, 1H), 2.74 (s, 3H, CH_3), 2.58 (s, 3H, CH_3). ^{13}C NMR (100 MHz, DMSO-d_6): δ 166.2, 151.4, 138.2, 136.1, 135.4, 131.2, 129.0, 127.3, 124.0, 123.0, 119.2, 15.2, 10.3

2) 3-[(4-Chlorophenyl)diazenyl]-2,7-dimethylimidazo[1,2-a]pyridine 8

Yield 78%, Yellow solid, mp 174˚C - 177˚C; IR (KBr): υ_{max} cm^{-1} = 2993, 1520, 1473, 1462 and 796 cm^{-1}; MS: m/z $C_{15}H_{13}ClN_4$ (284/286 M^+., chlorine isotopes effect); HRMS (284.0902, Found 284.0901); ^{1}H NMR (300 MHz, DMSO-d6): δ 9.66 (d, J = 6.9 Hz, 1H), 7.88 (d, J = 8.7 Hz, 2H), 7.59 (d, J = 8.8 Hz, 2H + 1H pyr.), 7.16 (d, J = 7.0 Hz, 1H), 2.71 (s, 3H, CH_3), 2.49 (s, 3H, CH_3); ^{13}C NMR (100 MHz, DMSO-d6): δ 151, 149, 139, 136, 131, 129, 129, 124, 121, 119, 111, 21, 10.

3) 3-[(4-Chlorophenyl)diazenyl]-2,6-dimethylimidazo[1,2-a]pyridine 9

Yield 94%, green solid, mp. 145˚C - 147˚C; MS: *m/z* $C_{15}H_{13}ClN_4$ (284/286 $M^{+\cdot}$, chlorine isotopes effect); HRMS (Calculated 284.0902, Found 284.0897); ^{1}H NMR (300 MHz, DMSO-d_6): δ 9.55 (s, 1H), 7.91 (d, J = 8.7 Hz, 2H), 7.67 (d, J = 9.0 Hz, 1H), 7.60 (d, J = 8.7 Hz, 2H), 7.54 (d, J = 9.0 Hz, 1H), 2.70 (s, 3H, CH_3), 2.43 (s, 3H, CH_3); ^{13}C NMR (100 MHz, DMSO-d_6): (δ 161.0, 151.0, 138.0, 136.0, 131.0, 130.0, 129.0, 126.0, 124.0, 119.0, 111.0, 17.0, 10.0).

4) 3-[(4-Chlorophenyl)diazenyl]-2,5-dimethylimidazo[1,2-a]pyridine 10

Yield 93%, orange solid, mp 157˚C - 159˚C; MS: *m/z* $C_{15}H_{13}ClN_4$ (284/286 $M^{+\cdot}$, chlorine isotopes effect); HRMS (Calculated 284.0902, Found 284.0908); ^{1}H NMR (300 MHz, DMSO-d_6): δ 7.74 (d, J = 9.1 Hz, 2H), 7.60 (d, J = 9.1 Hz, 2H), 7.45 - 7.60 (m, 2H, overlapped), 7.12 (d, J = 6.8 Hz, 1H), 3.00 (s, 3H, CH_3), 2.71 (s, 3H, CH_3).

5) 8-Methylimidzao[1,2-a]pyridine-2,3-dione-3-[4-chlorophenyl)hydrazone] 11

Yield, 30%, Yellow solid, mp > 300˚C; MS m/z $C_{14}H_{11}ClN_4O$ (286/288 M^+, chlorine isotopes effect). HRMS (Calculated 286.0695, Found 286.0685). ^{1}H NMR (400 MHz, DMSO-d_6): δ 12.46 (s, 1H, NH), 8.39 (d, J = 6.9 Hz, 1H), 7.64 (d, J = 6.9 Hz, 1H), 7.60 (d, J = 6.9 Hz, 2H), 7.42 (d, J = 6.9 Hz, 2H), 6.88 (t, J = 6.9 Hz, 1H), 2.25 (s, 3H, CH_3).

6) 6-Methylimidzao[1,2-a]pyridine-2,3-dione-3-[4-chlorophenyl)hydrazone] 12

Yield 78%, red solid, mp 96˚C - 98˚C; MS *m/z* $C_{14}H_{11}ClN_4O$ (286/288 M^+, chlorine isotopes effect); HRMS (Calculated. 286.0695, Found 286.0700) ^{1}H NMR (400 MHz, DMSO-d_6): δ 15.55 (s, 1H, NH), 8.23 (s, 1H), 7.66 (d, J = 8. Hz, 1H), 7.60 (d, J = 8.4 Hz, 2H), 7.54 (d, J = 8.4 Hz, 2H), 6.87 (d, J = 8.4 Hz, 1H), 2.27 (s, 3H, CH_3). ^{13}C NMR (100 MHz, DMSO-d_6): δ 202.0, 150.0, 149.0, 140.0, 136.0, 133.0, 131.0, 130.0, 127.0, 112.0, 111.0, 18.0.

7) 5-Methylimidzao[1,2-a]pyridine-2,3-dione-3-[4-chlorophenyl)hydrazone] 13

Yield 30%, Yellow solid, mp 232˚C - 235˚C; MS *m/z* $C_{14}H_{11}ClN_4O$ (286/288 M^+, chlorine isotopes effect); HRMS (Calculated 286.0695, Found 286.0698). ^{1}H NMR (300 MHz, DMSO-d_6): δ 13.17 (s, 1H, NH), 7.76 (t, J = 8.0 Hz, 1H), 7.46 (br.s, 4H, not splitted), 7.08 (d, J = 8.0 Hz, 1H, Py), 6.88 (d, J = 8.0 Hz, 1H, Py), 2.82 (s, 3H, CH_3).

2.1.2. General Procedures for Syntheses of Compounds 18, 19, 23, 24 and 29

Triethylamine (0.6 g, 0.006 mol) in THF (30 ml) and methanol (10 ml) was drop-wise added to a stirred solution of hydrazonoyl chloride **1b** or **1c** (0.006 mol) and the appropriate azole (0.005 mol) at room temperature. Stirring was continued for 24 hours. The solvent was then removed and the solid precipitate was washed with water 50 ml and then washed with cold ethanol. The crude solid product was collected, dried and crystallized from hot ethanol.

1) Methyl 2-(3-amino-4H-1,2,4-triazol-4-yl)-2-(2-(4-chlorophenyl)hydrazono)acetate 18

Yield 50%, yellow solid, mp 255˚C - 257˚C; MS: m/z $C_{11}H_{11}ClN_6O_2$ (294/295 M^+, chlorine isotopes effect). ^{1}H NMR (300 MHz, DMSO-d_6): δ 10.88 (s, 1H, NH), 7.95 (s, 1H, N=CHN, triazole ring), 7.35 (s, 4H, aromatic ring), 5.95 (s, 2H, NH_2), 3.74 (s, 3H, OCH_3). ^{13}C NMR (100 MHz, DMSO) δ 161.5 (C=O), 154.0 (N=CN, triazol ring), 142.28 (C, aromatic ring), 138.76 (CH, NN=CHN, triazol ring), 129.32 (C, aromatic ring), 126.42 (s), 117.45 (CH, aromatic ring), 116.57 (CH, aromatic ring), 52.7 (OCH_3).

2) 1-(3-amino-4H-1,2,4-triazol-4-yl)-1-(2-(2,5-dimethylphenyl)hydrazono)propan-2-one19

Yield 40%, brown solid, mp194˚C - 196˚C; MS: m/z $C_{13}H_{16}N_6O$ (272 M^+) HRMS (Calculated 272.1386, found 272.1275). ^{1}H NMR (300 MHz, DMSO): δ 9.76 (s, 1H, NH), 7.89 (d, J = 1.3 Hz, 1H, CCHC, aromatic ring), 7.27 (s, 1H, N=CHN, trizole ring), 7.06 (d, J = 7.5 Hz, 1H, CH, aromatic ring), 6.84 (d, J = 7.5 Hz, 1H, CH, aromatic ring), 5.92 (s, 2H, NH_2), 2.46 (s, 3H, CH_3), 2.30 (s, 3H, CH_3), 2.25 (s, 3H, CH_3). ^{13}C NMR (100 MHz, DMSO) δ 190.5 (C=O), 153.3 (N=CN, triazol ring), 140.3 (C, aromatic ring), 138.8 (CH, NN=CHN, triazol ring), 135.8 (C, aromatic ring), 130.9 (CH, aromatic ring), 125.9 (aromatic ring), 124.5 (CH, aromatic ring), 123.1 (C, aromatic ring), 117.8 (CH, aromatic ring), 24.7 (CH_3), 20.8 (CH_3), 17.1 (CH_3).

3) Methyl 2-(5-amino-1H-tetrazol-1-yl)-2-(2-(4-chlorophenyl)hydrazono)acetate 23

Yield 30%, Yellow solid, mp 180˚C - 182˚C; IR (KBr) υ_{max} cm^{-1}: 3214, 3168, 3111 and 1748. MS: m/z $C_{10}H_{10}ClN_7O_2$ (296) $[M+H]^+$. ^{1}H NMR (300 MHz, DMSO), δ ppm: 11.12 (s, 1H, NH), 7.43 (d, J = 9.0 Hz, 2H, aromatic ring), 7.37 (d, J = 9.0 Hz, 2H, aromatic ring), 7.12 (s, 2H, NH_2), 3.79 (s, 3H, OCH_3). ^{13}C NMR (100 MHz, DMSO) δ 160.88 (C=O), 156.5 (C=N), 141.9, 129.7, 127.3, 116.9, 116.5 (aromatic ring), 53.1 (OCH_3).

4) 1-(5-amino-1H-tetrazol-1-yl)-1-(2-(2,5-dimethylphenyl)hydrazono)propan-2-one 24

Yield 48%, pale Yellow solid, mp 161˚C - 162˚C; IR (KBr):υ_{max} cm^{-1} = 3327.56, 2980.65, 2360, 1660 cm^{-1}. MS: m/z $C_{12}H_{17}N_7O$ (273). ^{1}H NMR (400 MHz, DMSO) δ 10.24 (s, 1H, NH), 7.26 (s, 1H, CH, aromatic ring), 7.08 (d, J = 7.6 Hz, 1H, CH, aromatic ring), 6.92 (s, 2H, NH_2), 6.88 (d, J = 7.7 Hz, 1H, CH, aromatic ring), 2.47 (s, 3H, CH_3), 2.30 (s, 3H, CH_3), 2.25 (s, 3H, CH_3). ^{13}C NMR (100 MHz, DMSO) δ 189.3 (C=O), 155.96 (NC=N, tetrazol ring), 140.04 (C, aromatic ring), 135.91(N=CN), 131.09 (CH, aromatic ring), 125.12 (CH, aromatic ring), 124.75 (C, aromatic ring), 123.82 (C, aromatic ring), 118.66 (CH, aromatic ring), 24.65 (CH_3), 20.70 (CH_3), 17.25 (CH_3).

5) Methyl-2-[(4-chlorophenyl) hydrazono]-2-(2'-imino-4',6'-dimethylpyrimidin-1'-(2'H)-yl)acetate 29

Yield 52%, brown solid, mp 130˚C - 132˚C MS: m/z $C_{15}H_{16}ClN_5O_2$(333/335 M^+, chlorine isotopes effect).^{1}H NMR (400 MHz, DMSO) δ 9.80 (s, 1H), 8.04 (s, 1H), 7.73 (s, 1H), 7.70 (d, 2H), 7.59 (d, 2H), 3.86 (s, 3H), 2.27

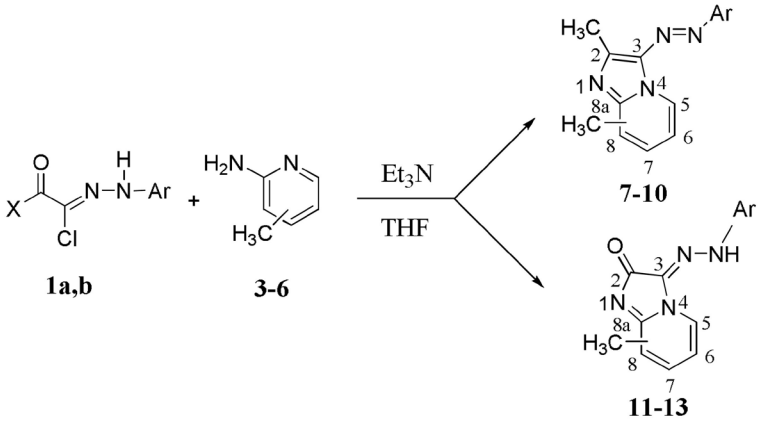
(s, 3H), 1.91 (s, 3H).

3. Results and Discussion

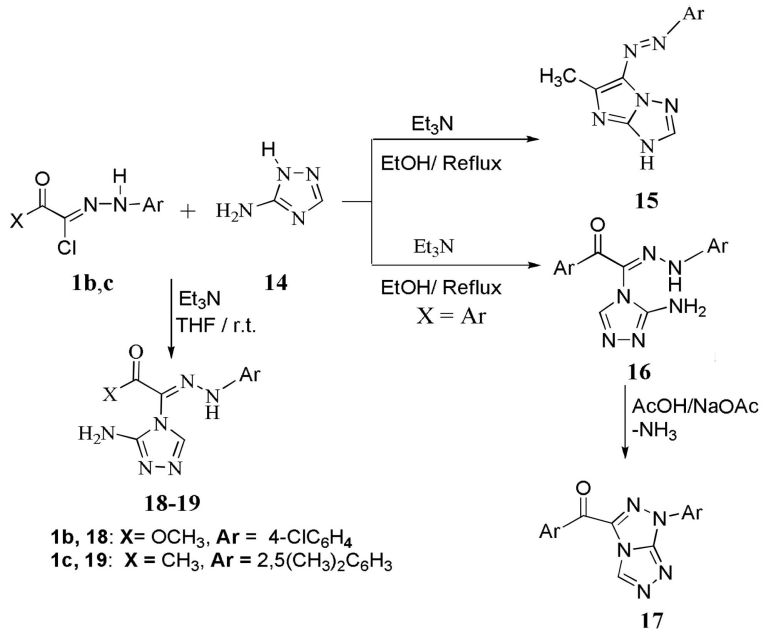
The reaction of picoline derivatives **3-6** with hydrazonoyl chloride **1a** in THF in the presence of Et_3N as a base at room temperature produced imidazo[1,2-a]pyridine **7-10** in 85% - 90% yield, while the reaction of hydrazonyl-chloride **1b** with substituted picolines **3-6** under the same reaction conditions afforded the expected imidazo[1,2-a]pyridine-2-ones **11-13** in 30% - 70% yield (**Scheme 1**). The structure of the products was confirmed by the different spectroscopic analytical methods including IR, MS, ^{1}HNMR and ^{13}CNMR.

The reaction of **1** with 3-amino-1,2,4-triazole **14** in refluxing ethanol was reported by Shawali *et al.* to give imidazo[1,2-b] triazole **15** in very low yield [10]. On the other hand, and under the same reaction conditions, Graf reported that the reaction of **14** with hydrazonylchloride **1** (X = Ar) afforded the acyclic adducts **16** [11]. Treatment of **16** with refluxing AcOH/AcONa led to formation of the cyclic fused product **18** *via* loss of a molecule of NH_3. The structure of **16** and **17** were determined by X-ray crystallography.

In this work, the reaction of hydrozonylchlorides **1b**, **c** with 3-amino-1,2,4-triazole **14** in the presence of Et_3N and THF at room temperature (**Scheme 2**) afforded the acyclic adducts **18** and **19**, respectively. This as-



Scheme 1. Reaction of hydrazonylchloride 1a, b with substituted picoline 3-6.

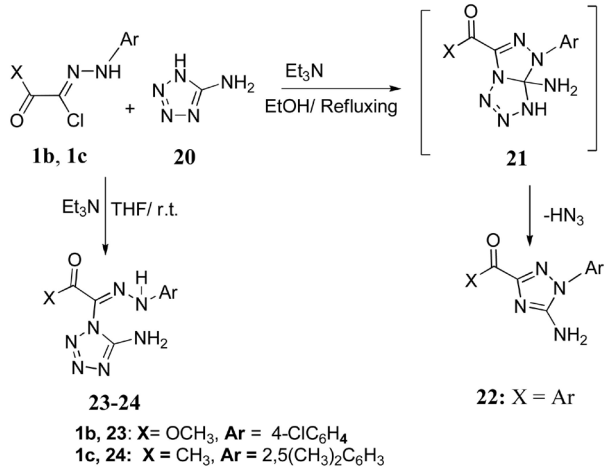


Scheme 2. Reaction of hydrazonylhalides 1b, c with 3-aminotriazole 14.

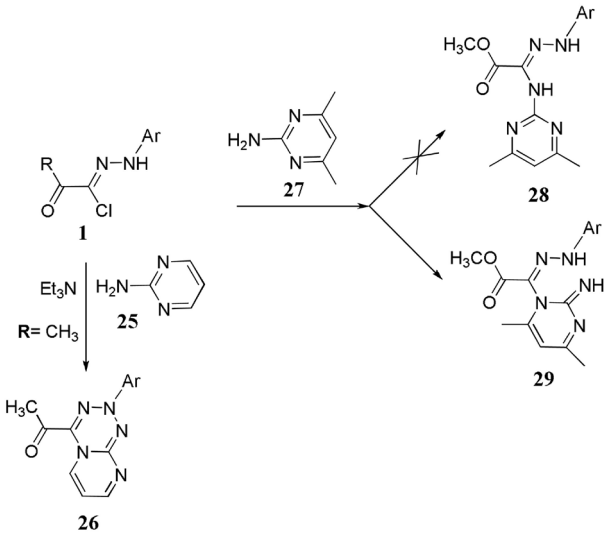
signment is based on the appearance of the NH_2 group and the C=O group in both IR and NMR spectra. Similar to Graf product, the reaction is proposed to occur at the N4 of the triazole **14**.The fused heterocyclic system reported by Shawali [10] was not obtained. Attempt to cyclize compounds **18** and **19** using AcOH/NaOAc (Graf method) was unsuccessful.

Graf reported [12] that the reaction of hydrazonylhalides **1** with 5-aminotetrazole **20** in refluxing ethanol produced compounds **22** *via* the rearrangement accompanied by loss of hydrazoic acid (HN_3) from the intermediate cycloaddition product **21**. In this work; the reaction of hydrazonyl halides **1b, 1c** with 5-aminotetrazole **20** in THF in the presence of Et_3N at room temperature afforded the acyclic adducts **23** and **24**, respectively (**Scheme 3**).

The reaction of hydrazonylhalide **1a** with 2-aminopyrimidine **25** was reported by Awadallah *et al.* [8] to produce the pyrimido [2,1-d] 1,2,3,5-tetrazine **26**. The reaction of **1b** with 4, 6-dimethyl-2-aminopyrimidine **27** in THE in presence of Et_3N as a base at room temperature produced however, the acyclic adduct, **29**. Two different possible isomeric structures are possible **28** and **29** (**Scheme 4**). The NMR data obtained provided an unambiguous confirmation that the actual obtained product is in fact compound **29** rather than compound **28**. The assignment is based on the appearance of two CH_3 groups at the pyrimidine ring. In compound **28** these two CH_3 are identical as they are in the same chemical and electronic environment, so they should appear as one peak



Scheme 3. Reaction of hydrazonylhalides 1b,1c with 5-aminotetrazole 20.



Scheme 4. Reaction of hydrazonylhalides 1b with 4, 6-dimethyl 2-aminopyrimidin 27.

with an integration value corresponding to 6H. On the other hand, the two CH_3 groups in compound **29** are different and two peaks with equal integration are expected. ^{1}HNMR spectra obtained supported structure **29** rather than **28**. The mass spectra form compound **29** in both positive mode 234 $[M+1]^+$ and negative mode 232 $[M-1]^+$ is shown in **Figure 1**.

4. Conclusion

A new series of heterocyclic compounds was synthesized *via* the reaction of Nitrilimines with 2-aminopicoline, 3-amino-1,2,4-triazole, 5-aminotetrazole and 2-aminopyrimidine. The structures of the products were characterized by IR, ^{1}H-NMR, ^{13}C-NMR and MS. The melting points of the synthesized compounds are listed in **Table 1**.

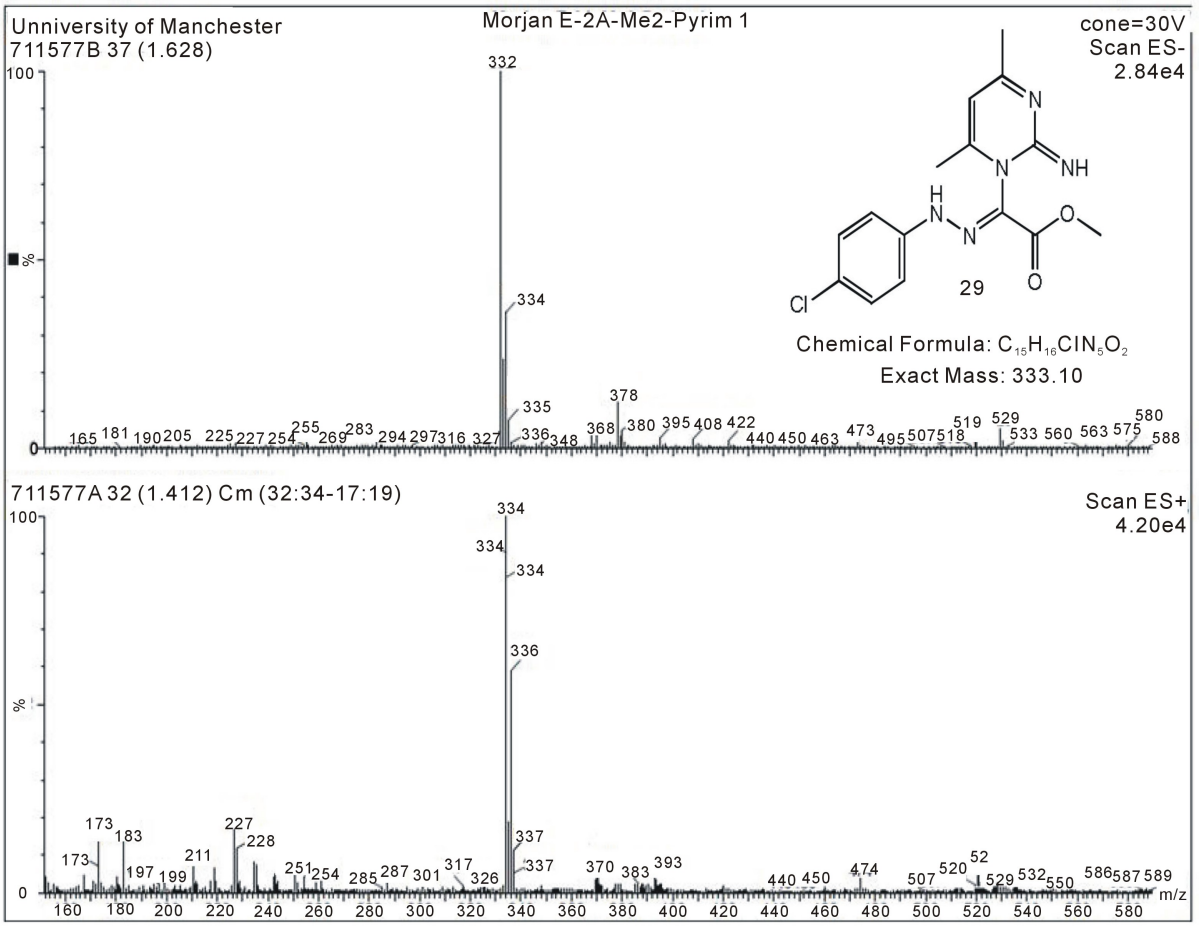


Figure 1. Mass spectra for compound 29 in both positive and negative mode.

Table 1. The melting points of the synthesized compounds.

Comp.	M.F.	M.W	M.P˚C	Comp.	M.F.	M.W	M.P˚C
7	$C_{15}H_{13}ClN_4$	284	152 - 155	**13**	$C_{14}H_{11}ClN_4O$	286	232 - 235
8	$C_{15}H_{13}ClN_4$	284	174 - 177	**18**	$C_{11}H_{11}ClN_6O_2$	294	255 - 257
9	$C_{15}H_{13}ClN_4$	284	145 - 147	**19**	$C_{13}H_{16}N_6O$	272	194 - 196
10	$C_{15}H_{13}ClN_4$	284	157 - 159	**23**	$C_{10}H_{10}ClN_7O_2$	295	180 - 182
11	$C_{14}H_{11}ClN_4O$	286	>300	**24**	$C_{12}H_{17}N_7O$	273	161 - 162
12	$C_{14}H_{11}ClN_4O$	286	96 - 98	**29**	$C_{15}H_{16}ClN_5O_2$	333	130 - 132

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