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RESEARCH ARTICLE

SYNTHESIS OF A NOVEL SERIES OF 2-SUBSTITUTEDIMINO-4- SUBSTITUTEDIMINO- 6-(4-PYRIDINEIMINO) AMINO-1,3,5- DITHIAZINES

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ABSTRACT

Recently a novel series of 2-substitutedimino-4-substitutedimino-6-(4- pyridineimino)amino-1,3,5-dithiazines (VIIIa1-a10) have been synthesized by refluxing 1-(4-pyridine)imino-5-substituted dithiobiuretes (Va-h) with various isocyanodichlorides (VIIa-c) in acetone-ethanol medium in 1:1 molar proportion for 2 hours. The structures of all the synthesized compounds were justified on the basis of chemical characteristics, elemental analysis and IR and NMR spectral analysis.

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INTRODUCTION

Chemistry of heterocyclic compounds is much more interesting as their utility and to synthesize a novel series of compounds as previous one used as an intermediate of newer one. From the literature survey it can be concluded that when the heterocyclic compounds containing 1,3,5-dithiazino or 1,3,5-thiadiazino molecule as a parent nucleus then that molecule will enhance medicinal, pharmaceutical, agricultural and industrial tricks of that drug (Li and Chan, 1999; Cave *et al*., 2001; Imrie *et al*., 2007; Anastas and Warner, 1988; Nassar Ekhalass, 2010; Abdel-Aziz *et al*., 2010; Toyata *et al*., 1990; Wang *et al*., 2005 and Baldwin *et al*., 1980). Hence, nowadays the drug containing 1,3,5- dithiazino or 1,3,5-thiadiazino nucleus are widely used in pharmaceutical, medicinal, biochemical and biotechnological fields (Jakhar and Makrand, 2010; Braghiroli *et al*., 2002; . Ei Bialy *et al*., 2005; Witvrouw *et al*., 1998; Vandamme, 1998; Liu *et al*., 2006; Blum and Carter, 1974; Wan *et al*., 2001 and Zhang *et al*., 2003). It has been reported that dithiazine nucleus and its analogous possess antiviral, antifungal, antibacterial, anti-tuberculostatic and anti-helminthic properties (Zhang *et al*., 2002; Ertan *et al*., 1992).

Several dithiazines are widely used in the treatment of cancer (Lin *et al*., 1991) and anti-HIV (Huang *et al*., 1993; Bayoumi and Hafez, 2006) drugs. They are also used in agriculture (Muelas *et al*., 2006) as like fungicidal (Hu *et al*., 2005), insecticidal (Scendo *et al*., 2003). These 1,3,5- dithiazines are also effective against copper corrosion (Dafali *et al*., 2002) and used in lubricating oil (Bhattacharyra *et al*., 1995). The important reactions of substituted isocyanodichlorides have been briefly investigated by some researchers (Berad, 1985; Pathe and Paranjpe, 1981; Pathe *et al*., 1982; Pathe, 1982; Berad, 1982; Aparajit, 1993; Tayade, 1996; Deohate, 2004; Panpaliya, 2006 and Shelke, 2005).

In the view of utility and significances of these compounds in various fields or sciences and as a part of wider programme of this laboratory in the synthesis of nitrogen, nitrogen and sulphur containing heteroacycles and heterocycles to developed an alternative route for the synthesis of six member heterocycles, it is quite interesting to investigate one step cyclisation of 1-(4-pyridine)imino-5-substituted dithiobiurets (Va-h) with N-substitute disocyanodichlorides (VIIa-c) in acetone- ethanol medium to isolate 2-substitutedimino-4-substitutedimino-6-(4-pyridineimino)- amino-1,3,5-dithiazines (VIIIa1-a10).

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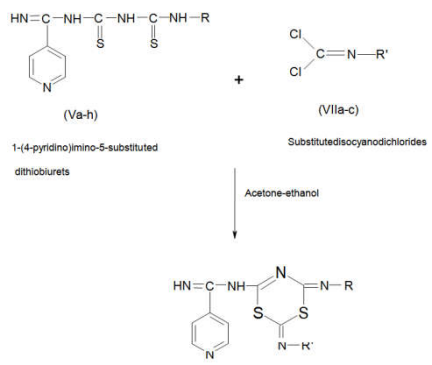
The tentative reaction for the formation of product is depicted below,

Scheme I



(VIIIa1-a10) 2-Substitutedimino-4-substitutedimino-6- (4-pyridineimino) amino-1,3,5-dithiazines

Where R= -methyl, -ethyl, -t-butyl, -phenyl, p-chlorophenyl, -o-tolyl, -m-tolyl, -p-tolyl.

R'= -Methyl, -ethyl, -phenyl.

MATERIALS AND METHODS

The melting points of all the synthesized compounds were recorded using hot paraffin bath and are uncorrected. The carbon and hydrogen analysis was carried out on Carlo-Ebra-1106 analyser, nitrogen estimation was carried out on Colman-N- analyser-29. IR spectra were recorded on Perkin-Elmer spectrometer in the range. 4000-400 cm^{-1} in KBr pellets. PMR spectra were recorded on Bruker AC-300F spectrometer with TMS as internal standard using $CDCl_3$ and DMSO-d_6 as solvent. The purity of the compounds was checked on Silica Gel-G plates by TLC with layer thickness of 0.3 mm. All chemicals used were of AR grade (Indian make) except allylthiourea Lancaster (Germany make). Alkyl/Aryl isothiocynates have been prepared by known literature methods (Tayade, 1996).

Experiment No. 1

Synthesis 2-methylimino-4-methylimino-6-(4-yridineimino) amino-1,3,5- dithiazine (VIIIa1)

A reaction mixture of 1-(4-pyridine) imino-5-methyldithiobiuret (Va) with methylisocyanodichloride (VIIa) in 1:1 molar ratio was refluxed on water bath in acetone-ethanol medium for 4 hours. During heating evolution of hydrochloride gas was clearly noticed. After distillation of excess of acetone-ethanol blood red colour product was isolated this on basification with dilute ammonium hydroxide dark brown crystals were afforded, yield 85%, m.p. 1230C.

Experiment No. 2

Synthesis 2-methylimino-4-ethylimino-6-(4-pyridineimino) amino-1,3,5-dithiazine (VIIIa2)

A reaction mixture of 1-(4-pyridine)imino-5-ethyldithiobiuret (Vb) with methylisocyanodichloride (VIIa) in 1:1 molar ratio was refluxed on water bath in acetone-ethanol medium for 4 hours. During heating evolution of hydrochloride gas was clearly noticed. After distillation of excess of acetone-ethanol blood red colour product was isolated this on basification with dilute ammonium hydroxide dark brown crystals were afforded, yield 83%, m.p. 1050C.

Experiment No. 3

Synthesis 2-methylimino-4-t-butylimino-6-(4-yridineimino) amino-1,3,5- dithiazine (VIIIa3)

A reaction mixture of 1-(4-pyridine)imino-5-t-butyldithiobiuret (Vc) with methylisocyanodichloride (VIIa) in 1:1 molar ratio was refluxed on water bath in acetone-ethanol medium for 4 hours. During heating evolution of hydrochloride gas was clearly noticed. After distillation of excess of acetone-ethanol blood red colour product was isolated this on basification with dilute ammonium hydroxide dark brown crystals were afforded, yield 78%, m.p. 1660C.

Experiment No. 4

Synthesis 2-methylimino-4-phenylimino-6-(4-pyridineimino)amino-1,3,5- dithiazine (VIIIa4)

A reaction mixture of 1-(4-pyridine)imino-5-phenyldithiobiuret (Vd) with methylisocyanodichloride (VIIa) in 1:1 molar ratio was refluxed on water bath in acetone-ethanol medium for 4 hours. During heating evolution of hydrochloride gas was clearly noticed. After distillation of excess of acetone-ethanol blood red colour product was isolated this on basification with dilute ammonium hydroxide dark brown crystals were afforded, yield 76%, m.p. 1800C.

Experiment No. 5

Synthesis 2-methylimino-4-p-Cl-phenylimino-6-(4-pyridineimino)amino-1,3,5- dithiazine (VIIIa5)

A reaction mixture of 1-(4-pyridine)imino-5-p-Cl-phenyldithiobiuret (Ve) with methylisocyanodichloride (VIIa) in 1:1 molar ratio was refluxed on water bath in acetone-ethanol medium for 4 hours. During heating evolution of hydrochloride gas was clearly noticed. After distillation of excess of acetone-ethanol blood red colour product was isolated this on basification with dilute ammonium hydroxide dark brown crystals were afforded, yield 75%, m.p. 1900C.

Experiment No. 6

Synthesis 2-methylimino-4-o-tolylimino-6-(4-pyridineimino) amino-1,3,5- dithiazine (VIIIa6)

A reaction mixture of 1-(4-pyridine) imino-5-o-tolyldithiobiuret (Vf) with methylisocyanodichloride (VIIa) in 1:1 molar ratio was refluxed on water bath in acetone-ethanol medium for 4 hours. During heating evolution of hydrochloride gas was clearly noticed. After distillation of excess of acetone-ethanol blood red colour product was

isolated this on basification with dilute ammonium hydroxide dark brown crystals were afforded, yield 76%, m.p. 1540C.

Experiment No. 7

Synthesis 2-methylimino-4-m-tolylimino-6-(4-pyridineimino) amino-1,3,5- dithiazine (VIIIa7)

A reaction mixture of 1-(4-pyridine) imino-5-m-tolyldithiobiuret (Vg) with methylisocyanodichloride (VIIa) in 1:1 molar ratio was refluxed on water bath in acetone-ethanol medium for 4 hours. During heating evolution of hydrochloride gas was clearly noticed. After distillation of excess of acetone-ethanol blood red colour product was isolated this on basification with dilute ammonium hydroxide dark brown crystals were afforded, yield 78%, m.p. 1980C.

Experiment No. 8

Synthesis 2-methylimino-4-p-tolylimino-6-(4-pyridineimino) amino-1,3,5- dithiazine (VIIIa8)

A reaction mixture of 1-(4-pyridine) imino-5-p-tolyldithiobiuret (Vh) with methylisocyanodichloride (VIIa) in 1:1 molar ratio was refluxed on water bath in acetone-ethanol medium for 4 hours. During heating evolution of hydrochloride gas was clearly noticed. After distillation of excess of acetone-ethanol blood red colour product was isolated this on basification with dilute ammonium hydroxide dark brown crystals were afforded, yield 76%, m.p. 2070C.

Experiment No. 9

Synthesis 2-ethylimino-4-phenylimino-6-(4-pyridineimino) amino-1,3,5-dithiazine (VIIIa9):

A reaction mixture of 1-(4-pyridine) imino-5-phenyldithiobiuret (Vd) with ethyl isocyanodichloride (VIIb) in 1:1 molar ratio was refluxed on water bath in acetone-ethanol medium for 4 hours. During heating evolution of hydrochloride gas was clearly noticed. After distillation of excess of acetone-ethanol blood red coloured product was isolated this on basification with dilute ammonium hydroxide brown crystals were afforded, yield 82%, m.p. 2100C.

Experiment No. 10

Synthesis of 2-phenylimino-4-phenylimino-6-(4-pyridineimino) amino-1,3,5- dithiazine (VIIIa10)

A reaction mixture of 1-(4-pyridine)imino-5-phenyldithiobiuret (Vd) with phenyl isocyanodichloride (VIIc) in 1:1 molar ratio was refluxed on water bath in acetone-ethanol medium for 4 hours. During heating evolution of hydrochloride gas was clearly noticed. After distillation of excess of acetone-ethanol blood red colour product was isolated this on basification with dilute ammonium hydroxide dark brown crystals were afforded, yield 76%, m.p. 2450C.

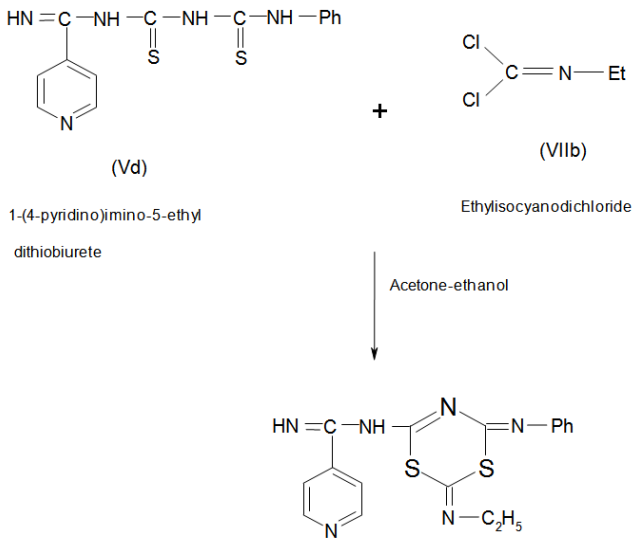
RESULTS AND DISCUSSION

Synthesis 2-ethylimino-4-phenylimino-6-(4-pyridineimino) amino-1,3,5- dithiazine (VIIIa9)

A reaction mixture of 1-(4-pyridine)imino-5-phenyldithiobiuret (Vd) with ethyl isocyanodichloride (VIIb) in 1:1 molar ratio was refluxed on water bath in acetone- ethanol medium for 4 hours. During heating evolution of hydrochloride gas was clearly noticed. After distillation of excess of acetone-ethanol blood red colour product was isolated this on basification with dilute ammonium hydroxide brown crystals were afforded, yield 82%, m.p. 2100C.

The probable reaction for the formation of (VIIIa9) is depicted below,

Reaction



(VIIIa9) 2-Ethylimino-4-phenylimino-6-(4-pyridineimino)amino-1,3,5-dithiazine

Properties of (VIIIa9)

- It was brown crystalline solid having m.p.210^{0}C.
- It gave positive test for nitrogen and sulphur and negative test for chlorine.
- It does not desulphurized when boiled with alkaline plumbite solution which clearly indicates that sulphur is not free and gets cyclised[38-39].
- It was soluble in benzene, acetic acid, DMF and DMSO.
- **Elemental analysis**: The result of elemental analysis is given in Table No. V-1). From the analytical data the molecular formula was found to be $C_{17}H_{17}N_6S_2$.

Table No. V 1

Elements	Found (%)	Calculated (%)
Carbon	54.64	55.43
Hydrogen	3.50	4.34
Nitrogen	22.82	22.82
Sulphur	17.05	17.39

- IR Spectrum: IR spectrum of compound was carried out in KBr pellets, an important absorption are correlated as follows in Table No. V-2.

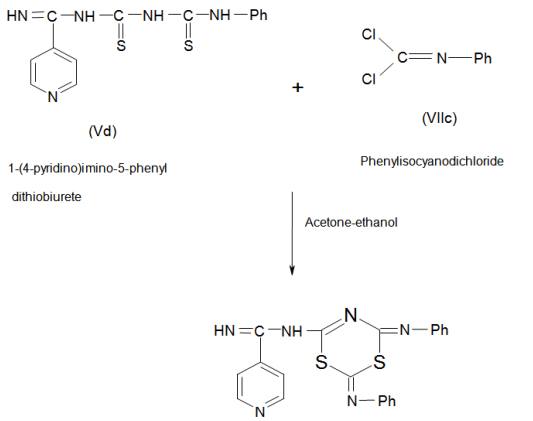
Table No. V-2

Absorption Observed cm^{-1}	Assignment	Absorption Expected cm^{-1}
3376	-NH Stretching	3500-3000
3166	-C-H stretching	3150-3000[40]
2163.1	-S-C=N stretching	2270-1940
1588	-C=N in pyridine ring	1660-1500[30,41]
1504.1	C=N stretching	1750-1450[42]
1090.5	C-N stretching	1360-1000[42]
668.12	C-S stretching	800-600[42]

PMR-Spectrum: The PMR spectrum (Tayade, 1995) of compound was carried out in $CDCl_3$ and DMSO-d_6. This spectrum distinctly displayed the signals due to Ar-protons at δ 6.6618-8.2035ppm, NH protons at δ 3.9266-4.2573 ppm, =NH protons at δ 3.1793-3.1976 ppm, -CH proton at δ 2.6138 ppm and $-CH_3$ protons at δ 1.2922-1.4456 ppm.

Synthesis of 2-phenylimino-4-phenylimino-6-(4-pyridineimino) amino-1,3,5-dithiazine (VIIIa10)

A reaction mixture of 1-(4-pyridine) imino-5-phenyldithiobiuret (Vd) with phenyl isocyanodichloride (VIIc) in 1:1 molar ratio was refluxed on water bath in acetone-ethanol medium for 4 hours. During heating evolution of hydrochloride gas was clearly noticed. After distillation of excess of acetone-ethanol blood red coloured product was isolated this on basification with dilute ammonium hydroxide dark brown crystals were afforded, yield 76%, m.p. 2450C. The probable formation of (VIIIa10) is depicted below,



(VIIIa10) 2-Phenylimino-4-phenylimino-6-(4-pyridineimino)amino-1,3,5-dithiazine

Properties of (VIIIa10)

- It was dark brown crystalline solid having m.p.245^{0}C.
- It gave positive test for nitrogen and sulphur and negative test for chlorine.

It does not desulphurized when boiled with alkaline plumbite solution which clearly indicates that sulphur is not free and gets cyclised (Hector, 1992 and Hector, 1992).

- It was soluble in benzene, acetic acid, DMF and DMSO.
- **Elemental analysis**: The result of elemental analysis is given in Table No. V-3

Table No. V-3

Elements	Found (%)	Calculated (%)
Carbon	59.70	60.57
Hydrogen	2.98	3.84
Nitrogen	20.19	20.19
Sulphur	14.51	15.38

- From the analytical data the molecular formula was found to be $C_{21}H_{17}N_6S_2$.

. **IR Spectrum:** IR spectrum of compound was carried out in KBr-pellets, an important absorption are correlated as follows in Table No. V-4

.

Table No. V-4

Absorption observed cm^{-1}	Assignment	Absorption Expected cm^{-1}
3376.8	NH Stretching	3500-3000
3176.0	(Ar) C-H stretching	3150-3000[30]
1635.0	C=N stretching	1750-1450[42]
1504.1	(Ar) C=C stretching	1600-1450
1254.3	C-N stretching	1360-1000[42]
723.14	C-S stretching	800-600[42]

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PMR-Spectrum: The PMR spectrum (Tayade, 1995) of compound was carried out in $CDCl_3$ and DMSO-d_6. This spectrum distinctly displayed the signals due to Ar-protons at δ 6.647-8.1570 ppm, NH protons at δ 3.5515 ppm and =NH protons at δ 2.5627-2.5850 ppm.

XRD Analysis: The XRD Analysis of the compound No. **(VIIIa10)** was carried out, during the analysis the start position is 02 Th which shows reading from 5.0084 and the end position 02 This 79.9784 It take 25.1973 sec. For complete analysis the analysis of this compound was carried out at 250C. Copper is used as anode material. The peak list obtained during analysis is shown in **Table No.V-5.** The measurements conditions are as depicted below, Measurement Conditions: (Bookmark 1)

Dataset Name MSL-18
File name C:\X'Pert Data\DEC2014\MSL-18.xrdml
Comment Configuration=Flat Sample Stage,

Owner=jagtar, Creation date=6/11/2007 3:57:00 PM

Goniometer=PW3050/60 (Theta/Theta); Minimum step size 2Theta:0.001; Minimum step size Omega:0.001
Sample stage=PW3071/xx Bracket
Diffractometer system=XPERT-PRO
Measurement program=PU, Owner=jagtar, Creation

date=4/15/2008 1:52:59 PM

Measurement Date / Time	12/22/2014 10:23:21 AM
Operator	Panjab University
Raw Data Origin	XRD measurement (*.XRDML)
Measurement Temperature [°C]	25.00
Anode Material	Cu
K-Alpha1 [Å]	1.54060
K-Alpha2 [Å]	1.54443
K-Beta [Å]	1.39225
K-A2 / K-A1 Ratio	0.50000
Generator Settings	40 mA, 45 kV
Diffractometer Type	0000000011023505
Diffractometer Number	0
Goniometer Radius [mm]	240.00
Dist. Focus-Diverg. Slit [mm]	100.00
Incident Beam Monochromator	No
Spinning	No

Table No.V-5 Main Graphics, Analyze View: (Bookmark 2)

Peak List: (Bookmark 3)

Pos. [°2Th.]	FWHM [°2Th.]	d-spacing [Å]	Rel. Int. [%]	Area [cts*°2Th.]
6.4994	0.1673	13.59976	2.92	9.14
11.7005	0.2007	7.56351	2.49	9.34
14.4967	0.1171	6.11027	16.76	36.68
17.0249	0.1338	5.20818	8.00	20.01
17.4373	0.2007	5.08593	4.73	17.76
19.0513	0.2007	4.65853	9.34	35.04
19.8273	0.1171	4.47792	6.80	14.88
20.3690	0.1338	4.36004	8.92	22.31
22.4546	0.1338	3.95958	21.72	54.32
23.1006	0.1673	3.85029	31.55	98.65
23.5226	0.1673	3.78217	13.65	42.67
24.7387	0.2007	3.59894	9.35	35.08
25.0677	0.1506	3.55244	24.40	68.66
25.6387	0.1673	3.47461	10.97	34.29
25.9308	0.1338	3.43613	8.93	22.34
26.3228	0.2342	3.38584	5.82	25.48
26.9600	0.1673	3.30724	92.02	287.69
27.6054	0.2007	3.23137	26.26	98.53
28.1047	0.1338	3.17509	10.57	26.43
28.6717	0.2007	3.11358	10.62	39.86
30.4961	0.2007	2.93134	4.53	16.98
32.3093	0.1338	2.77085	12.61	31.54
32.8235	0.2007	2.72861	100.00	375.19
33.4476	0.1506	2.67911	9.33	26.26
34.1173	0.1840	2.62804	12.51	43.02
35.1864	0.2342	2.55061	1.59	6.94
35.8038	0.4015	2.50802	3.14	23.58
37.9522	0.2676	2.37085	8.26	41.34
39.1795	0.2676	2.29936	4.63	23.19
40.1171	0.1673	2.24776	5.36	16.75
41.1751	0.2676	2.19242	4.04	20.20
44.2931	0.2007	2.04505	3.74	14.04
45.6411	0.4015	1.98774	2.50	18.73
46.9639	0.1338	1.93479	12.31	30.79
47.9079	0.2007	1.89885	2.21	8.28
50.5417	0.2007	1.80590	2.20	8.25
52.1953	0.2676	1.75252	2.54	12.72
52.8830	0.1004	1.73134	6.26	11.75
58.3899	0.1171	1.58049	13.12	28.72
61.7645	0.2007	1.50200	2.63	9.88
67.6458	0.1004	1.38501	2.12	3.98
68.5650	0.1224	1.36754	4.55	14.07
68.8088	0.2007	1.36442	3.80	14.27
73.3292	0.8029	1.29107	1.10	16.58
78.0407	0.1632	1.22348	5.56	22.92

Table No. V-6

Expt No.	Comd No.	1-(4-Pyridine) imino-5-substituted dithiobiuret	2- Substituedimino-4-substitutedimino-6-(4-pyridineimino) amino-1,3,5-dithiazine	Yield %	m.p. ^{0}C
1.	(VIIIa1)	1-(4-Pyridine) imino-5-methyldithio biuret (Va)	2-Methylimino-4-methyl-imino-6-(4-pyridineimino) amino-1,3,5-dithiazine	85	123
2.	(VIIIa2)	1-(4-Pyridine) imino-5-ethyldithiobiuret (Vb)	2-Methylimino-4-ethyl-imino-6-(4-pyridineimino) amino-1,3,5-dithiazine	83	105
3.	(VIIIa3)	1-(4-Pyridine) imino-5-t-butyldithiobiuret (Vc)	2-Methylimino-4-t-butyl-imino-6-(4-pyridineimino) amino-1,3,5-dithiazine	78	166
4.	(VIIIa4)	1-(4-Pyridine) imino-5-phenyldithiobiuret (Vd)	2-Methylimino-4-phenyl-imino-6-(4-pyridineimino) amino-1,3,5-dithiazine	76	180
5.	(VIIIa5)	1-(4-Pyridine) imino-5-p-chlorophenyl dithiobiuret (Ve)	2-Methylimino-4-p-chlorophenylimino-6-(4-pyridineimino)amino-1,3,5-dithiazine	75	190
6.	(VIIIa6)	1-(4-Pyridine) imino-5-o-tolyldithiobiuret (Vf)	2-Methylimino-4-o-tolyl-imino-6-(4-pyridineimino) amino-1,3,5-dithiazine	76	154
7.	(VIIIa7)	1-(4-Pyridine) imino-5-m-tolyldithiobiuret (Vg)	2-Methylimino-4-m-tolyl-imino-6-(4-pyridineimino) amino-1,3,5-dithiazine	78	198
8.	(VIIIa8)	1-(4-Pyridine) imino-5-p-tolyldithiobiuret (Vh)	2-Methylimino-4-p-tolyl-imino-6-(4-pyridineimino) amino-1,3,5-dithiazine	76	207

Similarly, 1-(4-pyridine)imino-5-methyldithiobiuret (Va), 1-(4- pyridine)imino-5-ethyldithiobiuret (Vb), 1-(4-pyridine)imino-5-t-butyldithio- biuret (Vc), 1-(4-pyridine)imino-5-phenyldithiobiuret (Vd), 1-(4-pyridine) imino-5-p- chlorophenyldithiobiuret (Ve), 1-(4-pyridine)imino-5-o-tolyldithio- biuret (Vf), 1-(4- pyridine)imino-5-m-tolyldithiobiuret (Vg) and 1-(4-pyridine)imino-5-m-tolyldithiobiuret (Vh) were interacted with methyl isocyanodichloride (VIIa) by above mentioned method to isolate 2 -methylimino-4-methylimino-6-(4-pyridineimino)amino-1,3,5-dithiazine (VIIIa1), 2-methylimino-4-ethylimino-6- (4-pyridineimino)amino-1,3,5-dithiazine (VIIIa2), 2-methyl-4-t-butylimino-6-(4-pyridineimino)amino-1,3,5-dithiazine (VIIIa3), 2-methylimino-4-phenylimino-6-(4- pyridineimino) amino-1,3,5-dithiazine (VIIIa4), 2-methylimino-4-p-

chlorophenylimino-6-(4-pyridineimino)amino-1,3,5-dithiazine (VIIIa5), 2- methylimino-4-o-tolylimino-6-(4-pyridineimino) amino-1,3,5-dithiazine (VIIIa6), 2-methylimino-4-m-tolyl- imino-6-(4-pyridineimino)amino-1,3,5-dithiazine (VIIIa7) and 2-methylimino-4-p-tolylimino-6-(4-pyridineimino)amino -1,3,5-dithiazine (VIIIa8) respectively are depicted in Table No. V-6.

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