

with the calculation. Since the K-X ray escape fraction is negligibly small the intensity of the single scatter photons was found by numerically integrating the Gaussian fitted to the above peak and dividing it by the appropriate peak-to-total ratio. The remaining portion in the low energy region is treated as that due to multiple scatter events. The intensity of multiple scatter photons as a function of scatterer thickness in Al and Cu is shown in figure 2. The results show that multiple scatter events in the backward direction increase with the sample thickness and saturate after a particular depth is reached, depending on the material. This is generally in agreement with the analytical results<sup>9</sup>. The saturation depths correspond to 8.3 cm in Al and 2.2 in Cu. Table I gives the saturation depth,  $\mu_d$ , the optical thickness and  $\lambda$ , the sample thickness<sup>5</sup>.

TABLE I  
Saturation depths at 662 KeV

Material	Saturation depth in cm	$\lambda$ in cm	$\mu_d$
Al	8.3	4.035	1.669
Cu	2.2	1.172	1.426

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## FLUORINE CONTAINING BIO-ACTIVE HETEROCYCLES: SYNTHESIS AND BIOLOGICAL ACTIVITY OF SOME FLUORINE CONTAINING TRIAZINO [5,6-b] INDOLES

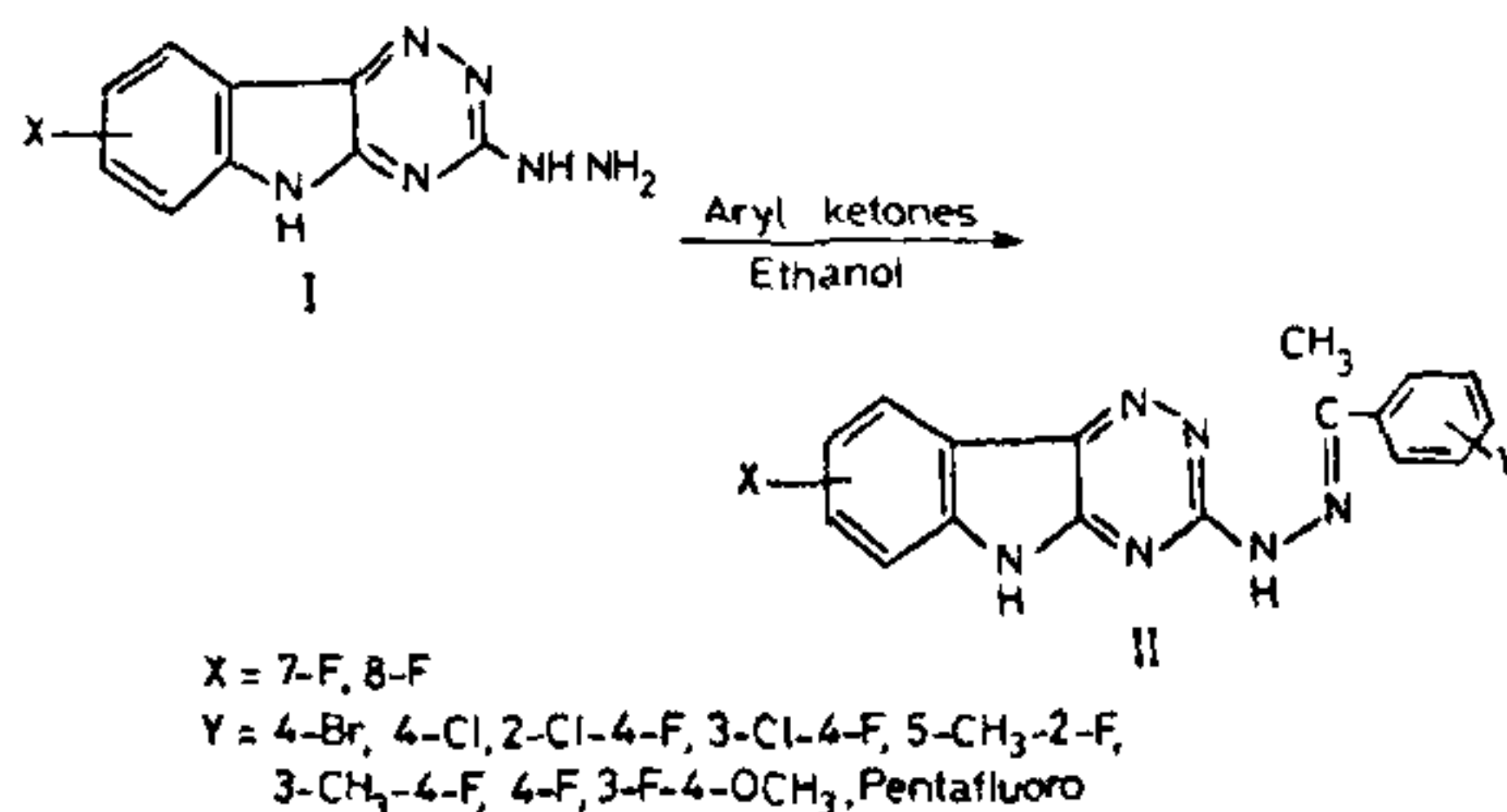
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CONDENSED derivatives of indole-2, 3-dione have been found to possess antibacterial, antifungal, antiviral and carcinogenic activity<sup>1</sup>. 5-Bromoisatin has shown analgesic activity stronger than that of aspirin<sup>2</sup>. 3-Substituted triazino [5, 6-b] indoles have exhibited antiviral activity against rhinovirus respiratory infection in Chimpanzees<sup>3</sup>. The hydrazino derivatives of these indole derivatives have been found to possess antiviral, anticonvulsant activity<sup>4</sup>.

Keeping the above observations in view, a large number of fluorine containing triazino [5,6-b] indole derivatives are reported along with their spectral studies viz. IR, PMR, <sup>19</sup>FNMR and antibacterial activities [Scheme-I.]



Scheme I

### 7-/8-Fluoro-3-hydrazino-1,2,4-triazino [5, 6-b] indoles [I<sub>1</sub>].

An appropriate fluorine containing indole-2,3-dione (0.051 M), thiosemicarbazide (0.055 M) and potassium carbonate (0.075 M) in water (200 ml) were refluxed to yield 1,2,4-triazino [5,6-b] indole-3-thiones<sup>5</sup>, which were recrystallized from a suitable solvent. Subsequently, the 1,2,4-triazinoindole-3-thione (0.02 mole) and hydrazine hydrate (20 ml) were refluxed for 4-5 hr. The reaction mixture was filtered and the product washed with water to give corresponding 3-hydrazino-1,2,4-triazino [5,6-b] indoles, which were purified by recrystallization from DMF. The products are homogeneous to TLC.

7-Fluoro-3-hydrazino-1,2,4-triazino [5,6-b] indole, yield 85%, m.p. 282-3. (Found N 38.10 C<sub>9</sub>H<sub>7</sub>FN<sub>6</sub>

TABLE I

## 7-/8-Fluoro-3-[(1'-methyl-substituted benzylidene) hydrazino]-1,2,4-triazino [5,6-b] indoles II

Sl. No.	X	Y	Molecular formula	M.P. (°C)	Yield (%)
1.	8-F	3-Cl-4-F	C <sub>17</sub> H <sub>11</sub> ClF <sub>2</sub> N <sub>6</sub>	305-7	62
2.	8-F	5-CH <sub>3</sub> -2-F	C <sub>18</sub> H <sub>14</sub> F <sub>2</sub> N <sub>6</sub>	287	61
3.	8-F	3-CH <sub>3</sub> -4-F	C <sub>18</sub> H <sub>14</sub> F <sub>2</sub> N <sub>6</sub>	305	61
4.	8-F	3-F-4-OCH <sub>3</sub>	C <sub>18</sub> H <sub>14</sub> F <sub>2</sub> N <sub>6</sub> O	318	62
5.	8-F	4-F	C <sub>17</sub> H <sub>12</sub> F <sub>2</sub> N <sub>6</sub>	312-4	67
6.	8-F	4-Cl	C <sub>17</sub> H <sub>12</sub> ClFN <sub>6</sub>	323	66
7.	8-F	4-Br	C <sub>17</sub> H <sub>12</sub> BrFN <sub>6</sub>	325-6	68
8.	8-F	Pentafluoro	C <sub>17</sub> H <sub>8</sub> F <sub>6</sub> N <sub>6</sub>	288-9	64
9.	7-F	2-Cl-4-F	C <sub>17</sub> H <sub>11</sub> ClF <sub>2</sub> N <sub>6</sub>	291	60
10.	7-F	3-Cl-4-F	C <sub>17</sub> H <sub>11</sub> ClF <sub>2</sub> N <sub>6</sub>	296-7	62
11.	7-F	5-CH <sub>3</sub> -2-F	C <sub>18</sub> H <sub>14</sub> F <sub>2</sub> N <sub>6</sub>	235-6	61
12.	8-F	2-Cl-4-F	C <sub>17</sub> H <sub>11</sub> ClF <sub>2</sub> N <sub>6</sub>	180	65
13.	7-F	4-Cl	C <sub>17</sub> H <sub>12</sub> ClFN <sub>6</sub>	278-9	66
14.	7-F	Pentafluoro	C <sub>17</sub> H <sub>8</sub> F <sub>6</sub> N <sub>6</sub>	193-5	62

requires N 38.53%).

8-Fluoro-3-hydrazino-1,2,4-triazino [5,6-b] indole. Yield 87%, m.p. 295-7° (Found N 38.21 C<sub>9</sub>H<sub>7</sub>FN<sub>6</sub> requires N 38.53%).

7-/8-Fluoro-3-[(1-methyl-substituted benzylidene) hydrazino]-1,2,4-triazino [5,6-b] indoles II.

An appropriate 3-hydrazino-1,2,4-triazino [5,6-b] indole (0.01 mole) and substituted acetophenone (0.01 mole) were refluxed in 20-25 ml absolute alcohol for 3-4 hr<sup>6</sup>. One drop of hydrochloric acid was added to initiate the reaction. On cooling the reaction mixture, crystals separated, filtered and recrystallized from ethanol. Their analytical data and melting points are given in table 1.

Some of the type II compounds were screened against gram negative bacteria *Escherichia coli* by sterile disc method<sup>7</sup> in different concentration ranging from 4 to 20 µg/ml. Three replicates of each test compound on various concentrations were used. The results obtained are recorded in table 2.

The PMR spectra of compounds of type I show new resonance signals between δ 7.95-8.2 ppm, which may be attributed to the protons of >NH and NH<sub>3</sub><sup>+</sup> (protonated). The aromatic protons appear between δ 7.0-7.7 ppm. The IR spectra of these compounds have shown the absorption band between 3325-2960 cm<sup>-1</sup> (>NH), 1560 cm<sup>-1</sup> (>C=N) and 1530 cm<sup>-1</sup> (N=N). All the compounds of type II, have been characterized by IR and <sup>19</sup>FNMR spectra. The >NH absorption band appears between 3500-3380 cm<sup>-1</sup> and C=N absorption between 1590-1580 cm<sup>-1</sup>. The <sup>19</sup>FNMR spectra of 7-/8-fluoro-3-[(1'-methyl-

TABLE 2

Sensitivity of gram negative bacteria *Escherichia coli* against 7-/8-fluoro-3-[(1'-methyl-substituted benzylidene) hydrazino]-1,2,4-triazino [5,6-b] indoles

Sl. No.*	Concentrations of different compounds used (mg/ml)		
	12	16	20
1.	±	±	±
2.	±	+	+
3.	±	±	+
4.	±	±	+
5.	+	++	+++
6.	++	++	+++
7.	++	+++	+++
8.	+	+	+
9.	+	+	+
10.	+	+	++

\* The serial nos. of compounds correspond to those given in Table 1.

— Insensitive

Area of inhibition of bacterial zone;

± 0.1-0.3 cm; + 0.3-0.5 cm;

++ 0.5-0.7 cm; +++ 0.7-0.9 cm.

pentafluorobenzylidene) hydrazino]-1,2,4-triazino [5,6-b] indoles show the resonance signal due to 7-/8-fluorine between δ 39.4-40 ppm and three resonance



signal due to aromatic fluorine atom at  $\delta$  71.0 ppm (2F),  $\delta$  179.0 (1F) and  $\delta$  88.0 (2F).

In all the above compounds the absorption band due to Ar. C-F stretching vibration appear between 1260-1200  $\text{cm}^{-1}$ .

The antibacterial activity of all synthesized compounds given in table 1 was studied against gramnegative bacteria *Escherichia coli* (table 2). The results indicated that only a few compounds possess prominent activity. Compound Nos. 5, 6, 7 possess maximum activity.

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## DIPHENYL LEAD (IV) DIODIDE COMPLEXES WITH SUBSTITUTED 2-AMINOTHIAZOLES

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ORGANO lead(IV) compounds have generated considerable interest<sup>1,2</sup>. The synthesis and spectral studies on organo lead(IV) complexes of dithiocarbamates<sup>3</sup>, triphenylphosphin oxide<sup>4</sup>, 1-chloro-1,2,3-benzotriazoles<sup>5</sup>, acetylacetone<sup>6</sup> have been reported. The spectral studies on five coordinate diorganolead(IV) complexes of ONO and SNO donor ligands have been documented<sup>7</sup>. The organo lead(IV) complexes with ONNO tetradentate ligands have been studied by DiBianca *et al.*<sup>8</sup>. However, attempts have not been made on complexes of diorganolead(IV) with aminothiazoles. The present communication deals

with the preparation and spectral properties of diphenyl lead(IV) diiodide complexes of substituted 2-aminothiazoles.

All chemicals used were of BDH grade. The ligands 2-amino-4-phenylthiazole (apt)<sup>9</sup>, 2-amino-4-ethyl-5-methylthiazole (aemt)<sup>10</sup>, 2-amino-4,5,6,7-tetrahydrobenzothiazole (athbt)<sup>11</sup>, 2-amino-4-methyl-5-carbethoxythiazole (amct) and diphenyl lead(IV) diiodide<sup>12</sup> were synthesized according to published methods.

The complexes were prepared by mixing diphenyl lead(IV) diiodide and the appropriate ligand solutions in 1,2-dichloroethane in the ratio of 1:2 moles. The mixture was refluxed for 2 hrs and concentrated to a small volume. The yellowish orange coloured complex was isolated by the addition of petroleum ether (40-60°). The solid complex was filtered, washed with aliquots of 1,2-dichloroethane followed by dry petroleum ether. It was dried under vacuum.

The complexes were analysed for the metal, nitrogen, sulphur and iodine by standard procedures. The analytical data agreed with the calculated values of the complexes within the limits of experimental errors. The molar conductance measurements of the complexes in acetonitrile at a concentration  $\sim 1 \times 10^{-3}$  M were taken on ELICO conductivity bridge. Infrared spectra (4000-400  $\text{cm}^{-1}$ ) of ligands and their complexes were recorded on Carl-Zeiss UR-10 spectrophotometer. The NMR spectra were taken on a Varian T-60 spectrometer in  $\text{CDCl}_3$  using TMS as internal standard.

The complexes are crystalline and yellowish orange in colour and have  $\text{Ph}_2\text{PbL}_2\text{I}_2$  stoichiometry. The molar conductance in acetonitrile solution lies in the range 12-17 mhos  $\text{cm}^2 \text{mole}^{-1}$ . The low conductance accounts for the non-electrolytic nature of all the complexes. An attempt to determine the molecular weight of these complexes failed due to poor solubility of the complexes in common organic inert solvents.

2-Aminothiazoles can coordinate to central metal through sulphur, ring nitrogen or amino nitrogen. Recent studies on the complexes of Co(II), Ni(II), Cu(II) and Ti(IV) with the above ligands, have indicated coordination through amino nitrogen of ligand molecule.<sup>13-16</sup> However, metal sulphur bonding in Ag(I)<sup>17</sup> and metal ring nitrogen bonding in Co(II), Cu(II) and Hg(II) complexes of 2-aminobenzothiazoles have been reported.<sup>18</sup> This makes it interesting to ascertain the coordination of substituted 2-aminothiazoles to diorganolead(IV). The infrared band assignment for the major bands is presented in table 1.

The solution spectra of ligands exhibit bands at  $\sim 3500 \text{ cm}^{-1}$  and  $\sim 3400 \text{ cm}^{-1}$  whereas solid phase spectra show in the range 3450-3350  $\text{cm}^{-1}$  and 3300-3100  $\text{cm}^{-1}$ . These bands are assigned to  $\nu_{\text{NH}}$  vibrations of  $\text{NH}_2$  group. Thus the difference about