



## Studies on the Pyrazine Complexes of Some Diaryltellurium Dihalides

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### ABSTRACT

Nine new Pyrazine Complexes of diaryltelluriumdihalides,  $R_2TeX_2 \cdot Pyz$  ( $R = p$ -methoxyphenyl,  $p$ -hydroxyphenyl and 3-methyl- 4-hydroxy phenyl;  $X = Cl, Br, I$ ;  $Pyz =$  pyrazine) have been prepared by reactions of diaryltelluriumdihalides with pyrazine in 1:1 molar ratio. These complexes have been characterized by elemental analysis, conductance and cryoscopic measurements, infrared and proton magnetic resonance spectral studies. Conductance studies in nitrobenzene, acetone and acetonitrile predict their non-electrolyte type behaviour in these solvents, which is well supported by cryoscopic data in nitrobenzene. IR and  $^1H$  NMR studies suggest the unidentate nature of pyrazine in these complexes, involving only one nitrogen atom in coordination. Thus, tellurium in  $R_2TeX_2 \cdot Pyz$  complexes attains a coordination number of five probably in a square pyramidal.

**Key words:** Diaryltellurium dihalides, Pyrazine complexes.

### INTRODUCTION

Aryltellurium trihalides are known to behave as Lewis acids and form molecular complexes with several nitrogen, oxygen and sulphur donor bases. It has been reported<sup>1</sup> that Lewis acidity of tellurium atom falls significantly on going from  $R_2TeX_3$  to  $R_2TeX_2$ . Kulkarni and coworkers screened the adducts of  $a, a'$ -bis(3-nitro/amino benzoyl) tellurium dichloride with N donor bases for biological activity and were found to be effective Ach E inhibiting agents<sup>2</sup>.

ShekharSrivastava and coworkers<sup>3</sup> have prepared  $R_2TeX_2 \cdot L$  type compounds where  $R =$  benzyl and  $L = 2,2'$ -bipyridyl; 1,10-phenanthroline and neocuproine. In these adducts, donor ligands are coordinated to tellurium atom. In the view of this and in continuation of our earlier work<sup>4,5</sup> on reactions of various diorganyl telluriumdihalides  $R_2TeX_2$  with some N-donor bases we hereby, report the synthesis and characterization of pyrazine complexes of bis( $p$ -hydroxyphenyl), bis(3-methyl-4-hydroxyphenyl), bis( $p$ -methoxy phenyl) tellurium(IV) dihalides.

## EXPERIMENTAL

### Materials and Methods

All the chemicals used were of Analytical Reagents grade. Solvents were purified and dried by conventional methods.

### Preparations

All the preparations have been carried out in a dry conditions under inert atmosphere of dry nitrogen, as the compounds are sensitive to moisture and air.

### Preparation of Diaryltellurium(IV) Dihalides

Bis(*p*-hydroxyphenyl)tellurium dichloride, bis(3-methyl-4-hydroxyphenyl) tellurium dichloride and bis(*p*-methoxyphenyl)tellurium dichloride were obtained by reactions of tellurium tetrachloride with phenol, *o*-cresol and anisole respectively. The diaryltelluriumdichlorides were converted to corresponding dibromides and diiodides by halogen exchange processes by using KBr or KI in dry methanol.

### Preparation of Pyrazine Complexes

#### Pyrazine Complexes of Bis(*p*-hydroxyphenyl) tellurium(IV) Dihalides

A saturated solution of pyrazine (0.40 gm, 5.0 mmol) in benzene (~10 ml) was added to a saturated solution of 5.0 mmol of  $R_2TeX_2$  (1.92 g, 2.37 g, 2.84 g for chloride, bromide and iodide respectively) in about 20 ml of methanol. The contents were stirred for about 3 h which resulted in the separation of a dark coloured solid product. This was filtered, extracted with diethyl ether and dried over  $P_4O_{10}$  in vacuum desiccator.

#### Pyrazine Complexes of Bis(3-methyl-4-hydroxyphenyl) tellurium(IV)Dihalides

A saturated solution of pyrazine (0.40 gm, 5.0 mmol) in benzene (~10 ml) was added to a saturated solution of 5.0 mmol of  $R_2TeX_2$  (2.06 g, 2.51 g, 2.98 g for chloride, bromide and iodide, respectively) in about 20 ml of dry methanol. The contents were stirred for 4 h. Brown coloured solid product thus obtained was filtered, washed with diethyl ether and dried over  $P_4O_{10}$  in vacuum desiccator.

#### Pyrazine Complexes of Bis(*p*-methoxyphenyl) tellurium(IV) Dihalides

A saturated solution of pyrazine (0.40 gm, 5.0 mmol) in benzene (~10 ml) was added to a saturated solution of 5.0 mmol of  $R_2TeX_2$  (2.06 g, 2.51 g, 2.98 g for chloride, bromide and iodide respectively) in methanol + chloroform (20:80). The contents were stirred of 4 h. Orange brown solid product obtained was filtered, extracted with diethyl ether and dried over  $P_4O_{10}$  in vacuum desiccators.

### Physical studies

Elemental analysis for Carbon, hydrogen and nitrogen contents were got estimated from Sophisticated Analytical Instrumentation Facility, Punjab University, Chandigarh on a ELEMENTAR VARIO EL III or CARLO ERBA Model 1108. The conductance studies were carried out in acetone, nitrobenzene and acetonitrile on a highly sensitive Systronics Conductivity Bridge Type 305. A dip type cell with smooth platinum electrodes was used. The molecular weights of the complexes of diaryltellurium(IV) dihalides have been determined cryoscopically using purified nitrobenzene as the solvent. A Beckmann's freezing point apparatus is used. Infrared spectra of the complexes of diaryltelluriumdihalides prepared during this course of study have been recorded in the region 4000-400  $cm^{-1}$  on a SHIMAZDU FTIR-8300 or PERKIN ELMER Model 2000 FTIR Spectrometer using KBr pellet technique. The spectra were got recorded from Dr. B.R. Ambedkar Centre for Biomedical Research, University of Delhi, Kurukshetra University, Kurukshetra and Sophisticated Analytical Instrumentation Facility, Panjab University, Chandigarh.  $^1H$  NMR spectra were recorded on a BRUKER AC-300F operating at 299.9486 MHz and BRUKER AVANCE II 400 NMR spectrometer operating at 400.13 MHz. The data were obtained from Sophisticated Analytical instrumentation facility, Punjab University, Chandigarh. The solutions were prepared in deuterated DMSO- $d_6$ . Spectra were recorded at room temperature immediately after preparing the solution using tetramethylsilane as an internal reference.

## RESULTS AND DISCUSSION

These diaryltelluriumdihalides when reacted with pyrazine give 1 : 1 type complexes,  $R_2TeX_2 \cdot L$  as per scheme-1

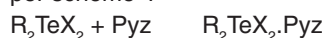


Table 1: Analytical Data, Physical Properties and Yields for Pyrazine Complexes of Diaryltellurium(IV) Dihalides.

$R_2TeX_2$ (R)	Complex (Empirical Formula)	Colour (Formula Wt.)	Analysis Found (Calculated) %					Yield %
			Te	X	C	H	N	
$R_2TeCl_2$ ( <i>p</i> -hydroxyphenyl)	$R_2TeCl_2 \cdot Pyz$	Purplish Pink (466.0)	27.87 (27.45)	16.12 (15.26)	40.37 (41.34)	3.40 (3.04)	6.51 (6.03)	135-137 78
$R_2TeBr_2$ ( <i>p</i> -hydroxy phenyl)	$R_2TeBr_2 \cdot Pyz$	Orange Brown (553.7)	23.42 (23.04)	28.57 (28.86)	34.41 (34.71)	2.60 (2.55)	5.12 (5.06)	146-148 75
$R_2TeI_2$ ( <i>p</i> -hydroxyphenyl)	$R_2TeI_2 \cdot Pyz$	Green Brown (647.7)	19.57 (19.70)	38.90 (39.19)	29.53 (29.67)	2.21 (2.18)	4.28 (4.33)	162-164 68
$R_2TeCl_2$ (3-methyl-4-hydroxyphenyl)	$R_2TeCl_2 \cdot Pyz$	Orange Brown (492.9)	25.74 (25.89)	14.41 (14.39)	43.79 (43.87)	3.57 (3.68)	5.34 (5.68)	139-141 80
$R_2TeBr_2$ (3-methyl-4-hydroxyphenyl)	$R_2TeBr_2 \cdot Pyz$	Dark Brown (581.8)	21.89 (21.93)	27.45 (27.47)	37.01 (37.16)	3.20 (3.12)	4.77 (4.82)	161-163 70
$R_2TeI_2$ (3-methyl-4-hydroxyphenyl)	$R_2TeI_2 \cdot Pyz$	Dark Brown (675.8)	18.83 (18.88)	37.43 (37.56)	31.84 (31.99)	2.71 (2.68)	4.21 (4.15)	168-170 72

Table 2: Molar Conductance Data for Pyrazine Complexes of DiaryltelluriumDihalides

$R_2TeX_2$ (R)	Complex	Nitrobenzene		Acetone		Acetonitrile		
		Molar Conc. $C \times 10^3$ mol L <sup>-1</sup>	Molar Cond. $\Lambda_m$ ohm <sup>-1</sup> cm <sup>2</sup> mol <sup>-1</sup>	Molar Conc. $C \times 10^3$ mol L <sup>-1</sup>	Molar Cond., $\Lambda_m$ Ohm <sup>-1</sup> cm <sup>2</sup> mol <sup>-1</sup>	Molar Conc. $C \times 10^3$ mol L <sup>-1</sup>	Molar Cond., $\Lambda_m$ Ohm <sup>-1</sup> cm <sup>2</sup> mol <sup>-1</sup>	
$R_2TeCl_2$ ( <i>p</i> -hydroxy-phenyl)	$R_2TeCl_2 \cdot Pyz$	1.00	1.38	1.00	5.36	1.00	17.60	
		1.67	1.30	1.67	4.76	1.67	13.44	
		2.14	1.21	2.14	4.02	2.14	11.20	
	$R_2TeBr_2$ ( <i>p</i> -hydroxy-phenyl)	$R_2TeBr_2 \cdot Pyz$	2.50	0.95	2.50	3.54	2.50	10.05
			2.71	0.80	2.71	1.10	2.71	8.62
			1.00	2.13	1.00	15.67	1.00	24.20
$R_2TeI_2$ ( <i>p</i> -hydroxy-phenyl)	$R_2TeI_2 \cdot Pyz$	1.67	2.06	1.67	11.36	1.67	19.51	
		2.14	1.98	2.14	10.47	2.14	17.23	
		2.50	1.36	2.50	8.54	2.50	16.54	
	$R_2TeCl_2$ (3-methyl-4-hydroxy phenyl)	$R_2TeCl_2 \cdot Pyz$	2.71	1.02	2.71	2.45	2.71	13.36
			1.00	8.93	1.00	18.76	1.00	35.23
			1.67	6.67	1.67	12.59	1.67	29.90
$R_2TeBr_2$ (3-methyl-4-hydroxyphenyl)	$R_2TeBr_2 \cdot Pyz$	2.14	4.33	2.14	9.97	2.14	28.56	
		2.50	4.01	2.50	7.32	2.50	27.50	
		2.71	2.76	2.71	3.98	2.71	14.28	
	$R_2TeI_2$ (3-methyl-4-hydroxyphenyl)	$R_2TeI_2 \cdot Pyz$	1.00	0.28	1.00	7.40	1.00	24.67
			1.67	0.17	1.67	5.06	1.67	20.13
			2.14	0.13	2.14	4.76	2.14	18.70
$R_2TeCl_2$ (3-methyl-4-hydroxyphenyl)	$R_2TeCl_2 \cdot Pyz$	2.50	0.12	2.50	4.52	2.50	14.36	
		2.71	0.99	2.71	4.36	2.71	10.57	
		1.00	1.76	1.66	15.70	1.00	27.60	
$R_2TeBr_2$ (3-methyl-4-hydroxyphenyl)	$R_2TeBr_2 \cdot Pyz$	1.67	1.47	1.67	10.63	1.67	24.55	
		2.14	1.40	2.14	9.54	2.14	18.76	

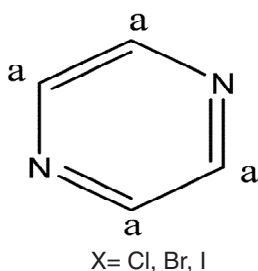
$R_2TeI_2$		2.50	1.32	2.50	9.21	2.50	16.34
$R_2TeI_2$ (3-methyl-4-hydroxyphenyl)	$R_2TeI_2 \cdot Pyz$	2.71	1.14	2.71	9.01	2.71	12.23
		1.00	2.73	1.00	18.00	1.00	32.66
		1.67	1.98	1.67	14.01	1.67	28.74
		2.14	1.40	2.14	12.09	2.14	20.22
		2.50	1.29	2.50	12.48	2.50	18.26
		2.71	1.21	2.71	12.40	2.71	10.73
$R_2TeCl_2$	$R_2TeCl_2 \cdot Pyz$	1.00	4.80	1.00	6.06	1.00	12.00
$R_2TeCl_2$ ( <i>p</i> -methoxy-phenyl)		1.67	4.31	1.67	5.78	1.67	9.34
		2.14	3.93	2.14	5.53	2.14	8.69
		2.50	3.34	2.50	4.84	2.50	8.63
		2.71	3.76	2.71	4.39	2.71	8.18
$R_2TeBr_2$	$R_2TeBr_2 \cdot Pyz$	1.00	5.40	1.00	72.45	1.00	18.76
$R_2TeBr_2$ ( <i>p</i> -methoxy-phenyl)		1.67	4.67	1.67	68.73	1.67	16.45
		2.14	4.44	2.14	54.36	2.14	15.23
		2.50	4.32	2.50	44.14	2.50	15.06
		2.71	4.10	2.71	38.12	2.71	14.33
$R_2TeI_2$	$R_2TeI_2 \cdot Pyz$	1.00	6.00	1.00	83.76	1.00	24.60
$R_2TeI_2$ ( <i>p</i> -methoxy-phenyl)		1.67	5.02	1.67	74.39	1.67	22.28
		2.14	4.81	2.14	63.38	2.14	20.75
		2.50	4.70	2.50	59.76	2.50	20.40
		2.71	4.67	2.71	40.44	2.71	20.30

Pyz = pyrazine

$\Delta_m$  reported<sup>2</sup> for 1 : 1 electrolyte : nitrobenzene = 20 - 30, acetone = 100 - 140 and acetonitrile = 120 - 160.

**Table 3: Molecular Weight Data for Pyrazine Complexes of Diaryltellurium(IV) Dihalides**

$R_2TeX_2$ (R)	Complex	Formula Weight	Concentration in mmoles/ litre of solvent	Molecular weight found
$R_2TeCl_2$ ( <i>p</i> -hydroxyphenyl)	$R_2TeCl_2.Pyz$	466.0	1.421	412.4
			2.137	439.9
			2.953	459.8
			3.131	470.4
$R_2TeBr_2$ ( <i>p</i> -hydroxyphenyl)	$R_2TeBr_2.Pyz$	553.7	1.377	521.7
			2.178	535.8
			2.784	546.3
			3.182	574.6
$R_2TeI_2$ ( <i>p</i> -hydroxyphenyl)	$R_2TeI_2.Pyz$	647.7	1.276	601.4
			2.047	620.3
			3.009	628.8
			3.921	654.6
$R_2TeCl_2$ (3-methyl-4-hydroxyphenyl)	$R_2TeCl_2.Pyz$	492.9	1.257	453.7
			2.193	468.9
			2.804	482.6
			3.314	501.3
$R_2TeBr_2$ (3-methyl-4-hydroxy phenyl)	$R_2TeBr_2.Pyz$	581.8	1.078	548.7
			1.931	560.4
			2.487	579.9
			3.208	588.3
$R_2TeI_2$ (3-methyl -4-hydroxyphenyl)	$R_2TeI_2.Pyz$	675.8	1.269	643.3
			2.084	657.8
			2.978	674.3
			3.483	689.1
$R_2TeCl_2$ ( <i>p</i> -methoxyphenyl)	$R_2TeCl_2.Pyz$	492.9	1.374	451.4
			2.038	477.6
			2.938	483.3

**Scheme 1. Formation of Pyrazine complexes**

Where

R = *p*-hydroxyphenyl, 3-methyl-4-hydroxyphenyl, *p*-methoxyphenyl

Pyz = pyrazine

All the compounds were analysed for their tellurium and halogen contents and some for carbon, hydrogen, nitrogen contents as well (results are given in table I). These complexes of diaryltellurium(IV) dihalides are generally coloured, crystalline solid which are fairly stable in dry air. These are generally soluble in polar organic solvents and insoluble in non polar solvents.

**Pyz = pyrazine**

The  $L_M$  values for the pyrazine complexes in nitrobenzene, acetone and acetonitrile are generally much lower than those reported for 1 : 1 electrolytes thus suggesting their non-electrolyte type behaviour in these solvents. Also the  $L_M$  values in a particular

Table 4: IR Data (cm<sup>-1</sup>) for Pyrazine Complexes of DiaryltelluriumDihalides

R <sub>2</sub> TeCl <sub>2</sub> .Pyz R = ( <i>p</i> -hydroxy-phenyl)	R <sub>2</sub> TeBr <sub>2</sub> .Pyz R = ( <i>p</i> -hydroxy-phenyl)	R <sub>2</sub> TeI <sub>2</sub> .Pyz R = ( <i>p</i> -hydroxy-phenyl)	R <sub>2</sub> TeCl <sub>2</sub> .Pyz R = (3-methyl-4-hydroxy phenyl)	R <sub>2</sub> TeBr <sub>2</sub> .Pyz R = (3-methyl-4-hydroxyphenyl)	R <sub>2</sub> TeI <sub>2</sub> .Pyz R = (3-methyl-4-hydroxyphenyl)	Assignment
3400-3200 s b 3100m 3059 s b 3010 sb 2978 s b 2887 s b 2760 s b	3414-3214 sb 3118m 3044 s 2963 s 2913 m	3466-3287 s b 2924 m 2880 s b	3413-3202 sb 3117s 3100 m 3054 s 3000 m 2943 m 2925 m 2910 m	3393-3195 sb 3112m 3080 m 3050 s b 2930 s b 2870 m	3454-3140 sb 3100m 2925 s 2920 s 2880 s 2810m	n <sub>O-H</sub> aromatic C-H stretching + N(CH)ofPyz+nR-CH <sub>3</sub>
2553 m b 2490 m b 2400 m b 2280 m b 2036 m b 1640 w 1569 s	2450 m b 2371 mb 2248 m b 2016 mb	2371 mb 2350 m b 2310 mb 2280 m b	2430 m b 2180mb 2100 m	2540 m b 2230 m b 2030 m b	2610 mb 2470 m b 2250 m b 2110mb	overtones and combination bands
1478 vs ring 1450 s CH3	1475 s 1418m 1420 sh	1480 vs 1403 m	1575 s 1495 vs 1477 vvs	1573 s 1492 s 1475 s	1620 m 1514s 1518m 1488 s 1448 s	V <sub>C=C</sub> sym. ring stretch inplaneantisym.
1370 sh	1367 sh	1362 sh 1340 w	1395 s 1347 m	1394 m 1345 m	1420 sh 1402 m 1342 m	stretch + antisym. bending + u ring of pyz. d <sub>(OH)</sub> <sup>+</sup> in the plane antisym. ring def. + sym. methyl C-H bending nc-o
1257 s	1268 s	1280 s	1273 vvs	1274 vs	1274 vs	

1245 sh	1240 m	1170 m	1239 m	1235 m	1244 m	aromatic in the plane
1168 s	1150 w	1104 m	1170 vs	1168 m	1156m	vib. + ring of Pyz.
1102 m			1160 m	1150 w	1114s	
			1125 vs	1123 s		
1051 m	1054 m	1060 m	1067 m	1065 m	1060 m	sym. in plane ring
1001 sh			1034 m	1032 w	1020 w	breathing
			1010w	1008 w		
958 m	967 m	960 m	985 m	981 m	957 m	Pyz. <i>unidentate</i>
925 m	886 s	741 m	919m	917m	898 s	C-H out of plane H-
831 m	826 m		912 m	879 s	872 s	bending vib + $\nu_{\text{CH}}$
758 m	750 m		882 m	850 m	800 m	Pyz.
	718 m		859 m	807 m	780 m	
			808 s	742 s		
			758 s	719 m		
			744 s			
			721 m			
685 wb	644 m	615 m	681 w		680 m	ring
bending vib.						
607 m	524 m	550 sh	650 m		640 mb	
580 sh		491 m	620 m		598 w	
515 m			599 w	*	550 w	
			553 m			
440 m			450 m	455 m		phenyl modes + g
			437 s			ring of Pyz.
424 w				435 w		

ws = very very strong, vs = very strong, s = strong, sh = shoulder, m = medium, vw = very weak, b - broad



**Table 5: IR Data (cm<sup>-1</sup>) for Pyrazine Complexes of Bis(*p*-methoxyphenyl)tellurium Dihalides**

R <sub>2</sub> TeCl <sub>2</sub> .Pyz	R <sub>2</sub> TeBr <sub>2</sub> .Pyz	R <sub>2</sub> TeI <sub>2</sub> .Pyz	Assignment
3200 s	3178 s	3129 s	aromatic C-H stretch + n(CH) of Pyz.
3075 m	3096 m	3100 m	
3010 m	2994 m	3000 m	
2900 m b			
2175 m	2184 m	2200 m	overtones and combination bands
2100 mb	2093 m b	2100 mb	
2040 m	1988 m	2000 m	
1900 m	1856 m	1975 m	
1871 m	1743 m	1875 m	
		1800 m	
		1725 m	
1600 m	1586 s	1582 vs	$\nu_{C=C}$ skeletal in plane ring stretch
1580 vs			
1492 s	1498 s	1500 s	n ring of Pyz
1456 m	1460 m	1487 s	
	1410 m	1457 m	
		1420 m	
1399 m	1348 m	1300 s	asym. C-O-C stretch
1301 s	1283 s	1275 s	
1255 s	1258 s	1246 vs	
1175 s	1178 s	1179 vs	g ring of Pyz. + aromatic in the plane vib. + sym.
C-O-C	1110 m	1107 m	1110 m stretch.
1053 m	1024 s	1023 vs	
1021 vs		1000 w	
965 s	953 s	957 s	Pyz. unidentate
815 vs	835 s	880	C-H out of plane H-bending vib + $\nu_{CH}$ of Pyz.
780 s sh		800	
		776 m	
715 m	718 w	712 m	out of the plane ring bending vib.
690 w	640 w	610 w	
610 w		580 w	

vs = very strong, s = strong, sh = shoulder, m = medium, w = weak, vw = very weak, b = broad

solvent for pyrazine adducts varies in the order.  
 R<sub>2</sub>TeI<sub>2</sub>.Pyz > R<sub>2</sub>TeBr<sub>2</sub>.Pyz > R<sub>2</sub>TeCl<sub>2</sub>.Pyz

#### Cryoscopic measurements

The cryoscopic data for the pyrazine complexes (Table III) show that molecular weights of these complexes are quite close to the formula weights, thus suggesting their molecular monomeric nature in nitrobenzene, as predicted by conductance

studies as well.

#### IR Spectra

The important IR data (cm<sup>-1</sup>) for pyrazine complexes of diaryltelluriumdihalides are presented in Tables IV and V. The spectra are quite complex and the frequencies from which concrete conclusion can be drawn are listed in the Tables. This may be mentioned that assignments made in the table are

**Table 6: <sup>1</sup>H INMR Data (d ppm) for Pyrazine Complexes of DiaryltelluriumDichlorides in DMSO-d<sub>6</sub>.**

Compound	Chemical Shift		Assignment
R <sub>2</sub> TeCl <sub>2</sub> .Pyz (R = <i>p</i> -hydroxyphenyl)	*		
	2.21 <sup>s</sup>	(6H)	-CH <sub>3</sub>
	6.94 <sup>d</sup>	(2H)	H <sub>b</sub>
			Phenyl } }
R <sub>2</sub> TeCl <sub>2</sub> .Pyz (R = 3-methyl-4-hydroxyphenyl)	7.50 - 7.69 <sup>m</sup>	(4H)	H <sub>A</sub> & H <sub>c</sub>
	8.05 <sup>b</sup>	(2H)	-OH
	8.61 <sup>s</sup>	(4H)	Ha (pyrazine)
	3.86 <sup>s</sup>	(6H)	-OCH <sub>3</sub>
	7.02 <sup>d</sup>	(4H)	H <sub>b</sub>
			Phenyl } }
R <sub>2</sub> TeCl <sub>2</sub> .Pyz (R = <i>p</i> -methoxyphenyl)	7.94 <sup>d</sup>	(4H)	H <sub>A</sub>
	8.60 <sup>s</sup> , 8.22 <sup>s</sup>	(4H)	Ha(pyrazine)

s = singlet, b = broad, m = multiple, cm = complex multiple, d = doublet

\* Spectra not well resolved.

based on comparison of spectra of parent R<sub>2</sub>TeX<sub>2</sub> and pyrazine. The ring vibrations of these may be mixed with each other.

The pyrazine molecule (C<sub>4</sub>H<sub>4</sub>N<sub>2</sub>) has the centrosymmetric and planar structure with D<sub>2h</sub> symmetry in the vapour and solid phases. The assignments of the infrared spectra of pyrazine in solid<sup>6</sup>, liquid<sup>7-8</sup> and vapour<sup>9-10</sup> phases and in solution<sup>6,10,11</sup> have been reported to be in accord with the D<sub>2h</sub> symmetry. The important infrared frequencies which are observed<sup>10,11</sup> in free pyrazine are 3061, 3011 (n<sub>CH</sub>), 1483, 1412 (n<sub>ring</sub>), 1150 (n<sub>ring</sub>), 1130 (D<sub>CH</sub>)<sub>9</sub>, 1061 (D<sub>CH</sub>), 1019 (D<sub>ring</sub>), 785 (u<sub>cm</sub>) and 418 (n<sub>ring</sub>). In metal complexes in which both N-atoms of pyrazine are coordinated to two metal atoms, the D<sub>2h</sub> symmetry is preserved but is removed in unidentate coordination. Several modes of pyrazine in complexes have upward shift in wave number in comparison with those of free pyrazine. This blue shift in pyrazine bands in the complexes indicates the coordination through N-atom as in pyridine<sup>12,13</sup>. This blue shift in *pp* pyrazine stretching may be due to back bonding from tellurium to pyrazine ring through formation of extensive  $\pi$ -bonding.

Lever *et al*<sup>14-17</sup> and Kantara<sup>18</sup> have reported that in complexes where pyrazine is bonded through

only one N-atom, the low local symmetry experienced by the pyrazine moiety atoms allows a band to appear in the 950-1000 cm<sup>-1</sup> region, which may be taken as an evidence for unidentate nature of pyrazine. This band is reported to be absent in the bridged polymers where pyrazine functions as bidentate ligand. In pyrazine complexes of diaryltellurium(IV) dihalides prepared during this course of study exhibit a medium to strong intensity band in this region, thereby confirming the unidentate nature of pyrazine in these complexes. Thus, tellurium in R<sub>2</sub>TeX<sub>2</sub>.Pyz complexes attains a coordination number of five probably in a square pyramidal environment.

#### <sup>1</sup>H NMR Spectra

The <sup>1</sup>H NMR data for pyrazine complexes are given in Table VI. The four equivalent protons of free pyrazine which resonate at 8.59<sup>19</sup> d ppm appear at slightly downfield side in R<sub>2</sub>TeCl<sub>2</sub>.Pyz complexes due to shifting of electron density from N of pyrazine to Te of R<sub>2</sub>TeCl<sub>2</sub>. Also in some cases separation between the pyrazine protons has been observed which may be due to monodentate nature of pyrazine. Conversely, the aryl protons of R<sub>2</sub>TeCl<sub>2</sub> group resonate at upfield side as compared to the parent diaryltellurium dichloride, indicating thereby the shielding of phenyl protons especially those *ortho* TeCl<sub>2</sub> group. This may be due to an

increase in electron density at the tellurium atom after accepting the electrons from the pyrazine molecule. Similar shift has also been reported for pyridine complexes<sup>20,21</sup> of  $\text{RTeCl}_3$ . Thus tellurium in these pyrazine complexes acquires a coordination number of five.

### CONCLUSION

Bis (*p*-hydroxyphenyl), bis (3-methyl-4-hydroxyphenyl) and bis(*p*-methoxyphenyl) tellurium (IV) dichlorides are obtained by direct reaction of tellurium tetrachloride with phenol, *o*-cresol and anisole respectively.

These diaryltelluriumdihalides have been reacted with pyrazine. The solution behaviour of complexes of diaryl tellurium (IV) dihalides has

been investigated by conductance measurement in nitrobenzene, acetone and acetonitrile and cryoscopic measurement in nitrobenzene. The Structural aspects of the complexes of diaryltelluriumdihalides have been investigated conjointly by infrared and proton magnetic resonance spectroscopy. These complexes of diaryl tellurium (IV) dihalides are generally coloured, crystalline solid which are fairly stable in air. These are generally soluble in polar organic solvents and insoluble in non polar solvents.

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