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SYNTHESIS OF IMIDAZOLATO AND 2-METHYL-IMIDAZOLATO DERIVATIVES OF TELLURIUM(IV)

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INTRODUCTION

The chemistry of S-N compounds has been well established^[1] in the past few years and the stability was accounted in terms of extensive π bonding^[2,3]. The diversity of these compounds tempted chemsits to explore analogous Se-N and Te-N compounds. However, the poor or even absence of π bonding with heavier Se and Te posed problem to synthesize such compounds in the past. The chemistry of Se-N and Te-N compounds has been developed extensively by introducing new synthetic precursors^[4] in the last decade or so. We have observed that N-trimethyl silylimidazole and 2-methyl silylimidazole are potential reagents for peperation of monomeric transition metal imidazolates and Tin(IV) imidazolate^[5,6]. We, therefore, thought worthwhile to synthesize amido derivatives of Te(IV) incorporating these π delocalised heterocycles which acts as potential 4e (both σ and π) donors.

Results and Discussion

The reagents Me₃SiIz and Me₃SiMeIz were reacted with R₂TeCl₂ and ReTCl₃ (R=CH₃, C₆H₅) at room temperature leading to the solid products as shown below:

$$R_{2} \operatorname{TeCl}_{2} + 2\operatorname{Me}_{3} \operatorname{Si} - \operatorname{N} \longrightarrow 2\operatorname{Me}_{3} \operatorname{SiCl} + R_{2} \operatorname{Te} - \left(\operatorname{N} \right)_{2}$$

$$1; R = \operatorname{CH}_{3}, R' = \operatorname{H},$$

$$2; R = \operatorname{CH}_{3}, R' = \operatorname{CH}_{3}$$

$$4; R = \operatorname{C}_{6} \operatorname{H}_{5}, R' = \operatorname{CH}_{3}$$

$$R \operatorname{TeCl}_{2} + 3\operatorname{Me}_{3} \operatorname{Si} - \operatorname{N} \longrightarrow 3\operatorname{Me}_{3} \operatorname{SiCl} + \operatorname{RTe} - \left(\operatorname{N} \right)_{2}$$

$$5; R = \operatorname{C}_{6} \operatorname{H}_{5}, R' = \operatorname{H}_{6}$$

$$6; R = \operatorname{C}_{6} \operatorname{H}_{5}, R' = \operatorname{CH}_{3}$$

The observed reactivity of these reagents could be understood in terms of the affinity of the Me₃ Si group for chlorine effecting the release of Me₃ SiCl.

The compounds were fairly stable at room temperature and soluble in usual organic solvents. The results of elemental analysis and molecular ion peaks (Table 1) support the proposed stoichiometry. The IR spectra (Table - 2) of these compounds exhibit bands characteristic of imidazolate moiety and the bands arising from organic substituents. The imidazolate ring vibrations assigned as R_1 , R_2 , R_3 and R_4 were found in the 1600 - 1300 cm⁻¹ region comparable with that of free imidazole as these are reported^[7] to be metal-independent. The strong band at ca 480 cm⁻¹ was assigned to ν asy (Te - N) stretching vibration.

The ¹H-NMR spectra of (Table-2) compounds (1-6) exhibited signals arising from the imidazolate protons^[8] including those arising from organic substituents at appropriate positions. The lone pair at imine nitrogen of the imidazole moiety may coordinate to the tin atom. However, intramolecular coordination seems to be sterically improbable and intermolecular coordination, leading to a octahedral configuration for these compounds appears more favourable.

UV spectra of these compounds dissolved in methyl cyanide exhibit two well resolved maxima in the regions 340-360 nm and 270-280 nm which may reasonably be assigned to the expected $\eta \rightarrow \sigma^*$ (Te 5px \rightarrow 4a₁, 3b₂) transitions and are consistent with that reported^[9] for Te(IV) derivatives..

TABLE 1: Yield, elemental analysis and molecular ion peaks of the compounds

Compunds	Yield (%)	%	M/z			
		C	Н	N	Te	
1.	68	33.2	4.4	19.2	44.0	289.6
		(32.9)	(4.1)	(19.2)	(43.7)	
2.	65	38.2	5.2	17.9	40.2	317.6
		(37.5)	(5.0)	(17.5)	(39.9)	
3.	65	51.9	4.0	13.1	30.4	414.6
		(51.9)	(3.8)	(13.4)	(30.7)	
4.	75	53.8	4.8	12.9	28.9	441.6
		(54.1)	(4.5)	(12.6)	(28.7)	
5.	75	44.3	3.4	20.9	30.9	404.6
		(44.3)	(3.4)	(20.7)	(31.4)	
6.	70	47.7	4.0	18.7	38.4	445.6
•		(48.2)	(4.4)	(18.7)	(28.5)	

TABLE 2: Spectroscopic data of compunds 1-6 in CDCl₃

Compo		IR virbrations cm ⁻¹ Imidazolate ring vibrations				Chemical Shift (δ ppm) (Imidezolate protons)	
	R_1	R ₂	R_3	R_4			-
1.	1580s	1480m	1405m	1320w	480 s	7.24(H)	7.45(2H)
2.	1590s	1495m	1410m	1310w	475s	8.64(H)	2.29(3H),7.1(2H)
3.	1610s	1470m	1415s	1310s	480s	7.30(H),	7.48(2H)
4.	1620s	1475m	1430s	1325s	470s	8.60(H),	2.30(3H),7.2(2H)
5.	1590s	1485m	1420s	1330s	475s	7.32(H),	7.50(2H)
6.	1600s	1490m	1425s	1310m	480s	8.66(H),	2.28(3H),6.6(2H)

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