

REVIEW

Organotellurium Compounds: From Molecular to Supramolecular Chemistry

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Supramolecules wherein discrete molecules are held together by non-covalent interactions are well known. It is more frequent in compounds containing conformational flexible molecule and functional groups like -COOH, -OH, -NH₂ that can participate in weak secondary interaction, thereby dictating the self-assembly. Moreover, metal complexes with different coordinating ligands can participate in supramolecular aggregations. Organotellurium compounds due to their unique property of hypervalency, three centre bonding and secondary bonding interactions finds its way towards supramolecular chemistry. The journey of organotellurium compounds from molecular to supramolecular chemistry is depicted here with reference to some selected examples.

Keywords: Organotellurium, Self-Assembly, Supramolecular associations, non-covalent interactions, Hypervalent.

INTRODUCTION

Organotellurium compounds classify the compounds containing a carbon (organic group) tellurium (Te-C) bond [1]. These compounds came into existence with the synthesis of dimethyl telluride, the first organotellurium compound, prepared by Wohler [2] in year 1840. Later on in 1910, Lederer [3] picked the work on the synthesis of organotellurium compounds. Further, studies on organotellurium compounds in detail, are available in the literature [4-17]. Reviews on the various important aspects of organotellurium compounds viz. theoretical, structural, NMR, mass spectral, radiation chemistry, ligand properties and their reactivity are also available [18,19]. Haiduc and Edelman [20] reported the chemistry of organotellurium compounds in 1999.

The synthetic routes of organotellurium compounds takes place by using (i) elemental tellurium (**Scheme-I**) (ii) tellurium tetrachloride (**Scheme-II**) and (iii) organotellurium compounds prepared by the above methods as intermediates (**Scheme-III**).

Types of organotellurium compounds

Tellurium is rich in various oxidation states with a wide range from -2 (H₂Te) to +6 (TeO₄²⁻). Naturally occurring tellurium compounds are found in various forms based on their oxidation states, viz. TeO₂ (+4) and TeO₃ (+6). The +4 and +6 oxidation states exhibit more stability and are more meaningful when compared with the other oxidation states. This feature of tellurium gives the formation of innumerable organotellurium compounds

viz. organotellurium(II) compounds (Table-1), organotellurium (IV) compounds (Table-2) and organotellurium(VI) compounds (Table-3). These compounds have been synthesized and characterized through various spectral studies. X-ray diffraction studies of selected compounds gave their geometries.

Supramolecular organotellurium compounds

Numerous examples of supramolecular organizations, formed by congregating molecular units, are seen with organometallic compounds. In organotellurium chemistry, Te---X (X = halogens, O, S) secondary interactions gives the basis of supramolecular self-assemblies. In organotellurium(IV) compounds viz. organotellurium dithiocarbamates (formed with ligands containing 1,1-dithio group including N,N-dialkyl dithiocarbamates) dimeric or polymeric structures were observed due to supramolecular associations through Te---S secondary bonds as discussed by King *et al.* [21]. A review by Haiduc & Zukerman [22] gives an account of various supramolecular assemblies (formed through secondary bonds) in organotellurium chemistry by explaining the active participation of Te---Te, Te---N, Te---O, Te---S and Te---X (X= Cl, Br, I) secondary non-covalent interactions. The great propensity of tellurium to form secondary interactions, which serve as organotellurium molecular Tectons (assembly of molecular units of supramolecular structures through non-covalent interactions) into supramolecular architectures is revealed by the data given by them.

The synthesis of organotellurium(IV) derivatives of O-alkyl-dithiocarbonates, dithiophosphates and monothiocarbamates

TABLE-1
ORGANOTELLURIUM(II) COMPOUNDS [Ref. 58]

Type	Examples
R_2Te and $RR'Te$	- Diphenyltellurides, dialkyl tellurides - Diaryltellurides $RTeR'$ ($R=R'=Ph$, $p-CH_3OC_6H_4$; $R=Ph$, $R'=p-CH_3OC_6H_4$) - Diaryl tellurides <i>viz.</i> $p-MeOC_6H_4TePh$ - Methyl vinyl telluride <i>Bis</i>(trifluoromethyl)- and <i>bis</i>(pentafluorophenyl)tellurides - N-[2-(4-methoxyphenyl telluroethyl)]benzamide and N-[2-(4-methoxyphenyl telluro-propyl)]phthalimide [(Te, N) type] <i>Bis</i>(triorganotellurium iodide); $O-C_6H_4(CH_2TeMe_2I)_2$ and $PhTeMe_2I$ 3,4- benzotelluracyclopentane [$C_8H_8Te(II)$] - Telluracyclopentane [$C_4H_8Te(II)$] <i>Bis</i>(<i>o</i>-isopropyl xanthate)tellurium - Tellurium azamacrocycles [<i>bis</i>(2-formylphenyl)telluride + diamines]
R_2Te_2 and $RR'Te_2$	- Diphenylditellurides [<i>Bis</i>(2,4,6-triisopropyl phenyl)ditelluride] R_2Te_2 ($R=Me$, Et, Me_2CH) $Te_2(CF_3)_2$ and $Te_2(C_6F_5)_2$ $[(CH_3)_2Te\{Te(CH_3)_2\}]^+$, $[(CH_3)_2Te\{Te(CH_3)_2\}]^+$, $[(CH_3)_2Te\{Se(CH_3)_2\}]^+$, $[(CH_3)_2Te\{S(CH_3)_2\}]^+$ and $[(CH_3)_2Te\{I(CH_3)_2\}]^+$
$RTeX$	2-Formyl phenyl tellurium derivatives, $RTeX$ ($X=Cl$, Br, I, CN, SCN, $SeCN$) $PhTeI$, (<i>p</i>-tolyl)TeI, (2-naphthyl)TeI $TeLBr$, $TeLCl$ [$L=(C_2H_5)_2NCS_2$]

TABLE-2
ORGANOTELLURIUM(IV) COMPOUNDS [Ref. 58]

Type	Examples
$RTeX_3$	2-biphenyl tellurium tribromides $RTe(teto)X_3$ ($X=Cl$, Br, I; <i>teto</i> = tetraethyl dithioamide) - Phenyl tellurium triiodide, Phenyl tellurium trichloride and Phenyltellurium tribromide (α-naphthyl)TeI₃
R_2TeX_2	(α-Me₂TeI₂), α-Me₂TeBr₂, (α-Me₂TeCl₂) - Tetraphenyl dichalcogenoimido diphosphinates <i>viz.</i> [$C_4H_8TeI\{Ph_2(Se)PNP(Se)Ph_2\}$], [$C_4H_8TeI\{Ph_2(Se)PNP(Se)Ph_2\}$], [$C_4H_8TeI\{Ph_2(S)PNP(S)Ph_2\}$], [$C_4H_8TeI\{Ph_2(S)PNP(S)Ph_2\}$], [$C_4H_8Te\{Ph_2(S)PNP(S)Ph_2\}_2$] $R_2Te(N_3)_2$ ($R=Me$, Ph) - Biphenylene tellurium(IV) dichloride - Diaryl tellurium dihalides ($p-RC_6H_4$)₂TeCl₂ ($R=H$, CH_3O) - Telluracyclohexane-1,1-diiodide ($C_6H_{10}TeI_2$); $C_5H_{10}TeX_2$ ($X=F$, Cl, Br, CN, NCO, NCS, NCS_2, N_3) - Diorganotellurium (IV) <i>bis</i>(salicylaldehyde) [$R_2Te(p-OC_6H_4CHO)_2$] ($R=Ph$, Me, $p-MeOC_6H_4$) $Me_2Te[C_5(CO_2Me)_3]_2$ $C_6H_6OSi_2Te$ and its derivatives. 2-Methyl-1-organo-1-halo-telluracyclopentane ($R=Me$, Et, allyl, $PhCH_2$, Ph, Cl, Br, iodo; $R'=Cl$, Br, iodo, BPh_4) - Diphenyl <i>bis</i> (N,N-dialkyl dithiocarbamates) tellurium(IV) and chlorodiphenyl (N,N-dialkyl dithiocarbamato)tellurium(IV) <i>viz.</i> $Ph_2Te(S_2CNR_2)_2$ ($R=Me$, Et), $Ph_2TeCl(S_2CNR_2)$ - Diphenyl tellurium(IV) monothiocarbamates <i>viz.</i> $Ph_2Te[SCONet]_2$, $Ph_2TeCl[SCONet]_2$. $C_5H_{10}TeCl(S_2CNHC_6H_5)$ Me_2TeBr_2, $Me_2Te(OCOC_6H_5)_2$ <i>m-oxobis</i>(1,1,2,3,4,5-hexahydro-1-nitrato tellurophe) [$C_4H_8TeI(S_2PMe_2)_2$], [$C_4H_8TeI(S_2PET_2)_2$], [$C_4H_8TeI(S_2CNC_6H_5)_2$], [$C_4H_8TeI(SOCNC_5H_{10})_2$], [$C_4H_8TeI(S_2CNEt_2)_2$] $R(OCOR')_2$ [$R=CH_3$, C_6H_5, C_6H_4, C_6H_3; $R'=OCO$, C_6H_5, 4-$NO_2C_6H_4$, 3,5-(NO_2)₂C_6H_3, 4-$OCH_3C_6H_4$] ($p-XC_6H_4$)₂TeCl₂ ($X=H$, Me, MeO), ($p-MeOC_6H_4COC-H_2$)₂TeBr₂ [4-MeC₆H₄COCH₂)Ph₃P]₂[TeBr₆] [$PhTe(CH_3)_2$]₂[TeX₆] ($X=Cl$, Br), [Ph_3Te][$PhTeX_4$] ($X=Cl$, Br, I) - CT complexes of Te(IV): $R_2TeI_2 \cdot IX$ [$R_2=(CH_3)_2$, C_4H_8, C_5H_{10}; $X=Cl$, I] $C_4H_8Te[S_2P(OCH_2)_2CMe-nPr]_2$, $C_4H_8OTe[S_2P(OCH_2)_2CEt]_2$ R_2TeX_2, [$R_2=(C_2H_5)_2$, $C_4H_7(CH_3)$; $X=I$, C_6H_5OCO, 4-$NO_2C_6H_4OCO$, 3,5-(NO_2)₂C_6H_3OCO, $C_6H_5CH=CHOCO$, 4-$OCH_3C_6H_4OCO$] $[(CH_3)_2TeNO_3]_2O$, $[(CH_3)_2TeOC_6H_2(NO_2)_3]_2O$, [$C_4H_8TeOC_6H_2(NO_2)_3]_2O$ $[(R_2Te)_2O]_2$ ($R=p-MeOC_6H_4$, Me) <i>bis</i>(Ferrocenyl carboxylato)telluranes [$TePR_2NPR_2Te$] ($R=Ph$, <i>i</i>-Pr, <i>t</i>-Bu) [K(18-crown-6)]₂[Te₂As₂] - TeI₂[(C₁₃H₁₀N₂S)₂].4C₄H₈TeI₂
R_3TeX	- Triphenyl telluronium thiocyanate - Triphenyl telluronium diorganyl phosphinodithiocarbamates (C_6F_5)₃TeCl, ($CF_3C_6F_4$)₃TeCl and ($CF_3C_6F_4$)₃TeBr - Trichloro-(4-methoxy phenyl) tellurium(IV)
R_4Te	- Tetraphenyl tellurium - Tetraorganotellurium(IV) [(R_4Te) ($R=Me$, Bu, CH_2SiMe_3 and vinyl)] - Tetramethyl chalcogens [Me_4S, Me_4Se and Me_4Te]

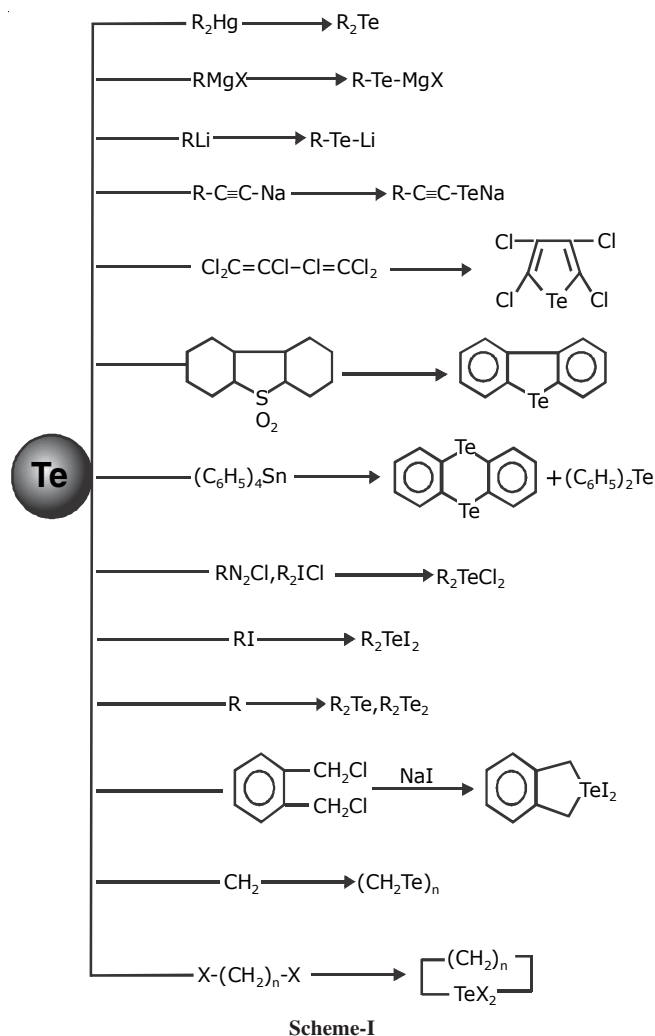


TABLE-3
ORGANOTELLURIUM(VI) COMPOUNDS [Ref. 58]

Type	Examples
RTeX_5	$(\text{C}_2\text{F}_5)_2\text{TeF}_4\text{Cl}$, CH_3TeF_5 , $(\text{CH}_3\text{O})\text{TeF}_4\text{Cl}$
R_2TeX_4	$(\text{C}_2\text{F}_5)_2\text{TeF}_4$
R_6Te	Ar_6Te ($\text{Ar} = 4\text{CF}_3\text{C}_6\text{H}_4$, C_6H_5)

were reported by Drake *et al.* [23-27]. The structural diversity at tellurium in the complexes is observed due to (a) strong tendency to form supramolecular associations through inter- and intramolecular Te---donor atom interactions and (b) the presence of stereochemically active lone pair at the tellurium centre. Further, Dakternieks *et al.* [28-30] and others [31,32] reported supramolecular associations in organotellurium dithiocarbamates, dithiocarbonates and dithiophosphates formed through Te---S secondary interactions.

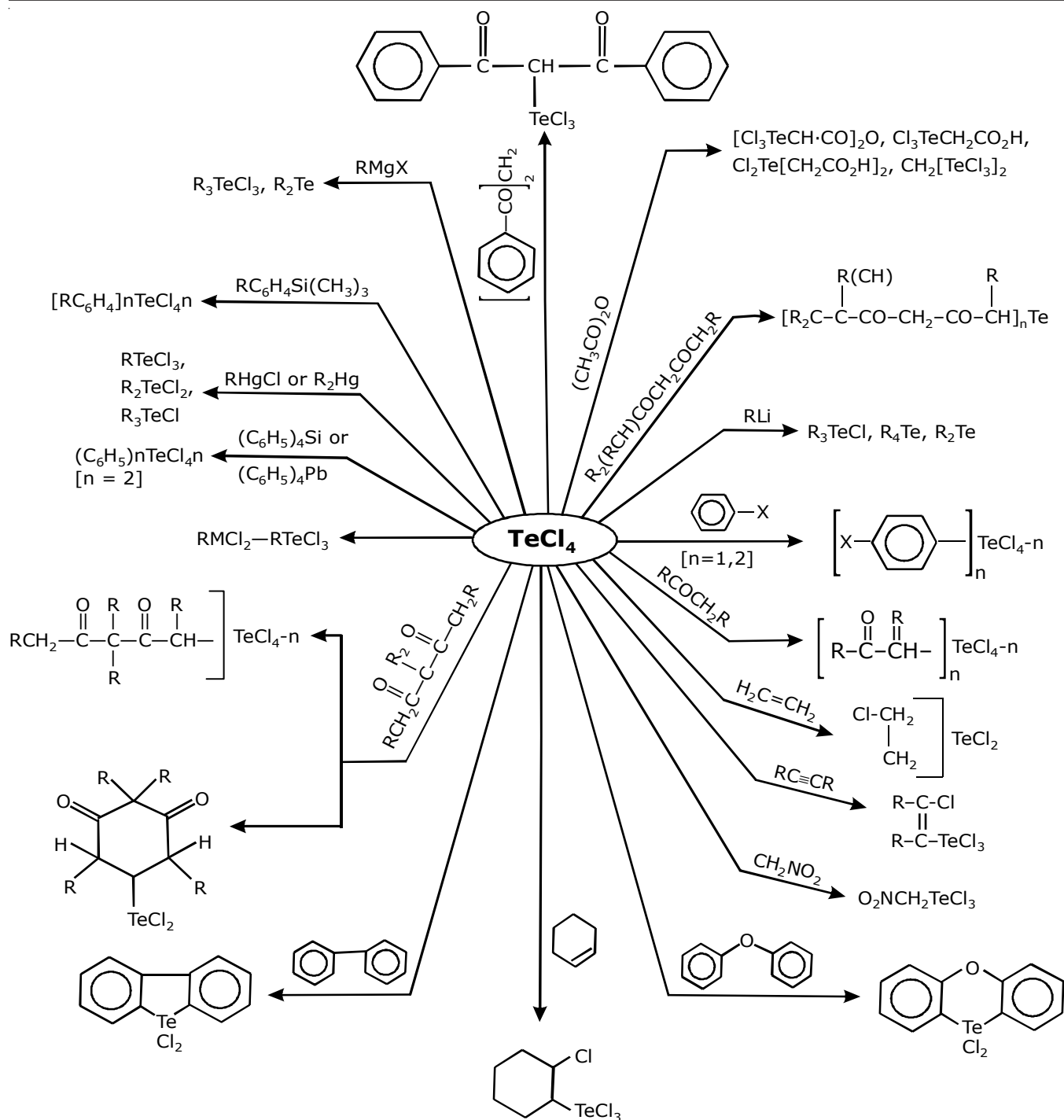
These complexes have been characterized by elemental analyses, IR, (^1H , ^{13}C , ^{31}P , ^{125}Te) NMR, mass spectroscopy and single crystal X-ray diffraction studies. Dimeric species in $\text{C}_4\text{H}_8\text{TeI}(\text{S}_2\text{CNC}_4\text{H}_6)$, $[\text{C}_4\text{H}_8\text{TeI}_2]_2$ and $[\text{C}_4\text{H}_8\text{TeI}(\text{SOCNC}_5\text{H}_{10})]$ are observed through intermolecular Te---S and intramolecular Te---I secondary bonds, respectively. The intermolecular Te---S secondary interactions in $[\text{C}_4\text{H}_8\text{TeI}(\text{S}_2\text{PMe}_2)]$, $[\text{C}_4\text{H}_8\text{TeI}(\text{S}_2\text{PEt}_2)]$ and $[\text{C}_4\text{H}_8\text{TeI}(\text{S}_2\text{CNEt}_2)]$ results in the formation of unidimensional polymer. The geometry around Te(IV) is distorted trigonal

bipyramidal. All the structures of organotellurium compounds possess different types of supramolecular associations. Supramolecular associations of mono and bis (dialkyl-, diaryl dithiocarbamates) of type R_2TeX_2 (where $\text{R} = \text{C}_4\text{H}_8$, C_5H_{10} and C_8H_8 ; $\text{X} = \text{O}$, Cl and I) built up through Te---S secondary interactions and/or C-H---X ($\text{X} = \text{O}$, Cl , I) hydrogen bonds are also reported [33].

Singh *et al.* [34] reported the stabilization of novel low valent organotellurium compounds on incorporating [2-[1-(3,5-dimethyl phenyl)-2-naphthyl]-4,5-dihydro-4,4-dimethyl-hyloxazole, influenced by both (a) steric effects and (b) Te---N intramolecular non-bonded interactions. The synthesis was achieved by organolithium route. All the compounds were characterized by elemental analysis, IR, (^1H , ^{13}C , ^{125}Te) NMR and mass spectroscopic studies. The single crystal X-ray crystallography studies confirmed the presence of Te---N intramolecular non-bonding secondary interactions in all the compounds. Syntheses of mono-substituted dithiocarbamates compounds containing organotellurium(IV) heterocyclic group *viz.* $\text{C}_4\text{H}_8\text{OTeI}[\text{S}_2\text{CN}(\text{CH}_2\text{CH}_3)_2]$, $\text{C}_5\text{H}_{10}\text{TeI}[\text{S}_2\text{CN}(\text{CH}_2\text{CH}_3)_2]$, $\text{C}_4\text{H}_8\text{TeI}[\text{S}_2\text{CN}(\text{CH}_2\text{CH}_3)_2]$, $\text{C}_8\text{H}_8\text{TeI}[\text{S}_2\text{CN}(\text{CH}_2\text{CH}_3)_2]$, $\text{C}_4\text{H}_8\text{OTeI}[\text{S}_2\text{CN}(\text{CH}_2\text{CH}_3)_2]$ and $\text{C}_5\text{H}_{10}\text{TeI}[\text{S}_2\text{CN}(\text{CH}_2\text{CH}_3)_2]$ are reported [35]. These compounds were characterized by IR, multinuclear NMR (^1H , ^{13}C and ^{125}Te) and EI-MS spectroscopic studies. The structures of some of these compounds were unambiguously established by single crystal X-ray diffraction studies. The synthesis and characterization of mixed-ligand diorganotellurium (IV) compounds containing dithiocarbamates and dithiophosphate ligands are also reported [36].

Srivastava *et al.* [37] reported a self-assembly through Te---I and I---I intermolecular non-covalent interactions in R_2TeI_2 type complexes *viz.* $\text{C}_4\text{H}_8\text{TeI}_2$, $\text{C}_5\text{H}_{10}\text{TeI}_2$ and $\text{C}_8\text{H}_8\text{TeI}_2$ resulting in 2D zig-zag ribbons in $\text{C}_4\text{H}_8\text{TeI}_2$, trimeric molecular aggregates in $\text{C}_5\text{H}_{10}\text{TeI}_2$ and 3D polymer in $\text{C}_8\text{H}_8\text{TeI}_2$ is reported. The structures of all these heterocyclic organotellurium diiodides were confirmed by single crystal X-ray diffraction studies. In each case, existence of distorted octahedral (six coordinated) geometry around Te atom with Te---I and I---I secondary bonds is observed, leading to the formation of supramolecular assemblies. Srivastava *et al.* [38] also reported charge transfer (CT) complexes formed through $n \rightarrow \sigma^*$ orbital interactions, where hypervalent Te---I bonds of telluranes [$\text{C}_4\text{H}_8\text{TeI}_2$, $\text{C}_5\text{H}_{10}\text{TeI}_2$ and $\alpha\text{-Me}_2\text{TeI}_2$] are utilized to form the charge transfer (CT) complexes. A prodigious binuclear species $\text{C}_8\text{H}_{16}\text{Te}_2\text{I}_6$ [$\text{IC}_4\text{H}_8\text{TeI-I-I-IC}_4\text{H}_8\text{TeI}$] is formed whose bridging I_2 acts as a supramolecular glue. One of the iodine atom of this I_2 molecule is exo-bound to Te of another $\text{C}_4\text{H}_8\text{TeI}_2$ molecule through weak Te---I contact making (formally) a trinuclear species. In the unit cell, such two trinuclear species are connected together through weak Te---I interactions thereby resulting in overall prolonged structure, hexanuclear species containing Te_2I_2 square.

$\text{C}_5\text{H}_{10}\text{TeI}_4$ (formed through reaction of $\text{C}_5\text{H}_{10}\text{TeI}_2$ with I_2), an analogue of I_2 adduct of acyclic dimethyl diiodotellurane Me_2TeI_4 (obtained by their action of Me_2TeI_2 with ICl) corresponded to complex of different structural motif where iodine molecules are on both sides bonded to iodine atom of hypervalent Te-I bond of $\text{C}_5\text{H}_{10}\text{TeI}_2$ molecule. The supramolecular



Scheme-II

associations in cyclic tellurane[1,1,2,3,4,5,6-heptahydro-1,1-dichlorotellurane] $C_5H_{10}TeCl_2$, where tetrameric molecular aggregates assisted by Te---Cl secondary bonds are observed [39]. These tetramers are further associated through long Cl---Cl contacts to yield octameric molecular aggregates.

Srivastava *et al.* [40] synthesized several cyclic telluranes *viz.* $C_4H_8TeCl_2$, $C_4H_8TeBr_2$, $C_5H_{10}TeBr_2$, $C_8H_8TeCl_2$, $C_8H_8TeBr_2$ and described the formation of 2D stairs in $C_4H_8TeCl_2$, two dimensional ribbons in $C_4H_8TeBr_2$, trimeric molecular aggregates in $C_5H_{10}TeBr_2$ and three dimensional supramolecular networks in $C_8H_8TeCl_2$ and $C_8H_8TeBr_2$, formed through intermolecular

Te---Cl and Te---Br secondary non-covalent interactions. Further secondary bonding interactions in *para*-substituted diphenyl tellurium dichlorides *viz.* $(p-XC_6H_4)_2TeCl_2$ [X = H, CH₃, OCH₃] confirmed by $^{13}C/^{125}Te$ NMR, CP-MAS spectroscopy along with the crystal and molecular structure of $(p-MeC_6H_4)_2TeCl_2$ have been reported [41]. Supramolecular self-assembly (through Te---X secondary bonds) in the solid state structure of 1,1,3-trichloro-2,4,5,6-tetrahydro-1*H*-1,4-benzotellurophene has been reported [42]. The structure unveils a hexagonal framework made up of four membered Te_2Cl_2 and 12-membered Te_6Cl_6 rings formed by self-assembly of Cl-Te-Cl rods through Te---Cl

supramolecular associations. By replacing iodine of $C_4H_7(CH_3)TeI_2$ with benzoate group (C_6H_5OCO group), polymeric supramolecular associations of $C_4H_7(CH_3)TeI_2$ are remodelled to trimeric supramolecular associations and when iodine of $C_4H_7(CH_3)TeI_2$ is replaced by 4-nitrobenzoate group ($4-NO_2C_6H_4OCO$ group), the polymeric supramolecular association of $C_4H_7(CH_3)TeI_2$ are transformed to tetrameric supramolecular association. This report demonstrates the cooperative participation of intermolecular I---I, Te---I, Te---O secondary bonds and $C(sp^3)H$ ---O and $C(sp^2)H$ ---O hydrogen bonds.

The supramolecular self-organization of tetraorgano-telluroxanes *viz.* $[(CH_3)_2Te(NO_3)_2]_2O$, $[(CH_3)_2TeOC_6H_2(NO_2)_3]_2O$ and $[C_4H_8TeO-C_6H_2(NO_2)_3]_2O$ are explained through cooperative participation of Te---O secondary bonds, C-H---O hydrogen bonds and π -stacking of organic substituents [54]. Supramolecular assembly of diverse structural types of organotellurium compounds in the reactions of $(4-OCH_3-C_6H_4)_2TeO$ has been reported by Chandrasekha and Kumar [55].

Recent reports from Srivastava *et al.* [56,57] envisages the supramolecular associations in various acyclic and cyclic organotellurium(IV) derivatives R_2TeX_2 , R_2TeIX and R_2TeCIX [$R_2 = (C_2H_5)_2$, $(n-C_3H_7)_2$, C_4H_8 , $C_4H_7(CH_3)$, C_5H_{10} , C_8H_8 ; $X = OCO-C_6H_5$, $OCOC_6H_3(NO_2)_2-3,5$, $OCOCH=C_6H_5$, $OCOC_6H_4(NO_2)-4$, $S_2CN(C_2H_5)_2$, $S_2CNC_4H_8O$ and $S_2CNC_5H_{10}$] formed through intramolecular Te---S secondary bonds, inter-molecular Te---X ($X = I, S, O$) secondary bonds and C-H---O hydrogen bonds. The third order non-linear optical studies of select organotellurium compounds by degenerate four wave mixing technique using a pico-second laser was also done which inferred the presence of efficient third-order non-linear optical susceptibility. $C_4H_7(CH_3)Te[S_2CN(C_2H_5)_2]_2$ shows an efficient third order NLO susceptibility $\{\chi^{(3)}\}$ of the order of 10^{-14} esu and second order hyperpolarizability (γ) of the order of 10^{-32} esu and it opens a new field of organotellurium derivatives to be applied in the field of photonics.

Conclusion

In general, the presence of supramolecular associations in organotellurium compounds is governed by both organic group and ligands attached to tellurium. They contribute and largely determine the nature and pattern of the supramolecular associations in organotellurium compounds. This emerging area is stimulated by two parallel developments (a) synthesis of novel tellurium agents with specialized properties and (b) the detailed evaluation of their reactivity and mechanisms associated with these compounds. The supramolecular associations in organotellurium compounds generated through cooperative Te---S, Te---I secondary bonds and C-H---O hydrogen bonds may be utilized as 'rescue agents'. In future, the unique properties of tellurium compounds will definitely place them in various technological applications.

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