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RESEARCH ARTICLE

Preparation and Characterization of a new metal complexes containing p-hydroxyphenyl telluriumtribromide and Triazole compounds

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Abstract

A new series of complexes containing p-hydroxyphenyl tellurium tribromide and triazole compound were prapared, diazonium salts of o-toluidine was reacted with piperidine to offer the corresponding triazole compound. p-hydroxyphenyl tellurium tribromide and triazole compound were reacted with copper bromide, hydrate nickel chloride zinc chloride, hydrate cobalt chloride and iron bromide in 1:2:2 mole ratio to give the transition elements complexes $M[(2\text{-}CH_3\text{-}C_6H_3\text{-}N{=}N)\text{-}N\text{-}C_5H_{10})]_2(4\text{-}OH\text{-}C_6H_4\text{-}TeBr_3)_2$ [where M= Cu (1), Ni (2), Zn (3), Co (4) and Fe (5). The organotellurium compound and triazole were reacted with the same metal halide in 1:1:3 mole ratio to give a new series of complexes complexes $M[(2\text{-}CH_3\text{-}C_6H_3\text{-}N{=}N)\text{-}N\text{-}C_5H_{10})](4\text{-}OH\text{-}C_6H_4\text{-}TeBr_3)_3$ [where M= Cu (6), Ni (7), Zn (8), Co (9) and Fe (10). The I.R, 1H NMR and (C.H.N) data are reported and discussed below.

INTRODUCTION

Triazenes are a class of organic compounds that containing diazoamino functional group (N=N–N) in composition.(1) This kind of compounds is known as just triazene. The coordination properties of the triazene ligand with Aryl groups as 1,3-substituents (Ar–N=N–NH–Ar) have been intensely studied during the past years. Coordination was found to be N1- monodentate, N1,N3-chelating and N1,N3-bridging ligand. These compounds have been used in a variety of ways, ranging from transition metal complexes and polymer synthesis to anticancer drugs.(1)The chemistry of triazene compounds have been the subject of increasing attention in recent years, because of their diverse synthetic applications;(2-4) Moreover, they have been widely applied to obtain triazenido complexes with a large range of metals.(2-4) In the same way organotellurium compounds have been used as a tool in synthetic chemistry(2-4)and ligand chemistry.(5) Thus, the present study will describe the synthesis and characterization of new types of complexes containing of organotellurium compounds and triazenes groups.

Experimental

Chemicals and apparatus

Chemicals obtained from Sigma-Aldrich, Fluka and BDH used without purification. Melting point was determined by using open capillary tube melting point apparatus. 1H NMR spectra was recorded on Bruker 300 MHz spectrometers with TMS as an internal reference using DMSO-d6 solvent. Infra-red spectra were recorded with KBr disks using a FTIR spectrophotometer Shimadzu model 8400 S in range 4000-400 cm-1. Elemental analysis for Carbon, Hydrogen and Nitrogen were performed by using a Euro vector EA 3000A Elemental Analysis (Italy). Melting points of all solid compounds were determined using a MPS10 electrically heated melting point apparatus.

2.2. Synthesis

The following compounds were synthesized as described in literature. (4–hydroxyphenyl)tellurim tribromide (6) and Triazene derivative (1-(o-tolyldiazenyl)piperidine) was prepared by the following general method:(7,8)

(8.7 mmol, 0.93 g) of o-toluidine was added to 4 ml of (6 M) HCl and heated on a water bath to yield a white suspension, which was cooled to 0℃. This suspension was maintained at 0℃, and a solution of (0.61 gm, 8.9 mmol) of NaNO2 in 2 ml of water was added dropwise with stirring over 10 min. A clear orange – brown solution was formed. To this solution (9.3 mmol, 0.79 g) of piperidine was added drop wise with stirring over 10 min. A yellow – orange solid was formed. The mixture was allowed to reach room temperature, and then brought to pH = 8 with saturated aqueous NaHCO3. The red- brown solid was collected by filtration, washed well with H2O and dried in air. The crude product was recrystallized from petroleum ether to offer the final product of triazene .
C12H17N3; formula weight: 203.28 gmol-1; yield: 91 % ; appearance: reddish brown crystals; m.p. = 70-71℃; Elem. analysis: calc. C, 70.90 %; H, 8.43 %; N, 20.67 %; found: C, 70.53 %; H, 8.21 %; N, 20.55 %; FTIR: 3022-3020 ν(C-H) aromatic, 2855 ν(C-H) aliphatic, 1101 ν(C-N) aliphatic, 1356 ν(C-N) aromatic, 1504 ν(C=C) aromatic, 1436 ν(N=N).. 1H NMR (ppm): 6.90- 7.28 (m, 4H, Ar), 2.35 (s, 3H, o-CH3), 3.10 (t, 4H, N-CH2 aliphatic), 1.51-1.59 (m, 6H, (-CH2-)3 aliphatic)

2.2.1. General procedure for the synthesis of metal complexes (1:2:2) mole ratio:

A solution of the ligand was prepared by preparing the mixture of ligands and metal salts as follows: asolution of 1-(o-tolyldiazenyl)piperidine (otdpip) (1 mmol) in absolute ethanol (10 mL) was added to solution of 4-(hydroxyphenyl)tellurium tribromide (OHPhTetBr) (1 mmol) in absolute ethanol (10 mL) and the mixture was refluxed for 30 min and then $NiCl_2 \cdot 6H_2O$, $CoCl_2 \cdot 6H_2O$, $CuBr_2$, $ZnCl_2$ or $FeBr_2$ (0.5 mmol) in absolute ethanol (10 mL) was added to the ligand solution with stirring and the reaction mixture was further stirred under reflux for 3 hrs. The obtained colored solution was left standing at room temperature to crystallize. The product was removed by filtration, washed with cooled absolute ethanol, recrystallized from methanol and dried. The analytical and physical data of the complexes are:

$[Cu(otdpip)_2(OHPhTetBr)_2]$(1)- Empirical formula: $C_{36}H_{42}CuBr_6N_6O_2Te_2$; formula weight: 1388.59 gmol-1; yield: 66 % ; appearance: brown crystals; m.p. = 162-164℃; Elem. analysis: calc. C, 31.13 %; H, 3.05 %; N, 6.05 %; found: C, 31.02 %; H, 3.01 %; N, 6.00 %; FTIR: 3075-3055 cm-1 ν(C-H) aromatic, 2810 cm-1 ν(C-H) aliphatic, 1024 cm-1 ν(C-N) aliphatic, 1283 cm-1 ν(C-N) aromatic, 1483 cm-1 ν(C=C) aromatic, 1449 cm-1 ν(N=N), 401 cm-1 ν(Te-C); 1279 cm-1 ν(C-O) phenolic. 1H NMR (ppm): 6.99- 7.30 (m, 16H, aromatic), 2.35 (s, 6H, *o*-CH_3), 3.20 (t, 8H, N-CH_2 aliphatic), 1.51-1.57 (m, 12H, (-CH_2-)$_3$ aliphatic).

$[Ni(otdpip)_2(OHPhTetBr)_2]$(2)- Empirical formula: $C_{36}H_{42}NiBr_6N_6O_2Te_2$; formula weight: 1384.59 gmol-1; yield: 73 % ; appearance: light brown crystals; m.p. = 185-186℃; Elem. analysis: calc. C, 31.24 %; H, 3.06 %; N, 6.07 %; found: C, 31.10 %; H, 3.05 %; N, 6.02 %; FTIR: 3075-3050 cm-1 ν(C-H) aromatic, 2830 cm-1 ν(C-H) aliphatic, 1023 cm-1 ν(C-N) aliphatic, 1280 cm-1 ν(C-N) aromatic, 1485 cm-1 ν(C=C) aromatic, 1450 cm-1 ν(N=N), 405 cm-1 ν(Te-C); 1285 cm-1 ν(C-O) phenolic. 1H NMR (ppm): 6.68- 7.20 (m, 16H, aromatic), 2.35 (s, 6H, *o*-CH_3), 3.22 (t, 8H, N-CH_2 aliphatic), 1.52-1.59 (m, 12H, (-CH_2-)$_3$ aliphatic).

$[Zn(otdpip)_2(OHPhTetBr)_2]$(3)- Empirical formula: $C_{36}H_{42}ZnBr_6N_6O_2Te_2$; formula weight: 1390.59 gmol-1; yield: 59 % ; appearance: yellowish - brown crystals; m.p. = 102-105℃; Elem. analysis: calc. C, 31.09 %; H, 3.04 %; N, 6.08 %; found: C, 31.00 %; H, 2.95 %; N, 5.98 %; FTIR: 3065-3050 cm-1 ν(C-H) aromatic, 2830 cm-1 ν(C-H) aliphatic, 1026 cm-1 ν(C-N) aliphatic, 1290 cm-1 ν(C-N) aromatic, 1450 cm-1 ν(C=C) aromatic, 1435 cm-1 ν(N=N), 407 cm-1 ν(Te-C); 1279 cm-1 ν(C-O) phenolic. 1H NMR (ppm): 6.87- 7.22 (m, 16H, aromatic), 2.30 (s, 6H, o-CH_3), 3.22 (t, 8H, N-CH_2 aliphatic), 1.53-1.59 (m, 12H, (-CH_2-)$_3$ aliphatic).

$[Co(otdpip)_2(OHPhTetBr)_2]$(4)- Empirical formula: $C_{36}H_{42}CoBr_6N_6O_2Te_2$; formula weight: 1384.59 gmol-1; yield: 68 % ; appearance: reddish - brown crystals; m.p. = 180-181℃; Elem. analysis: calc. C, 31.23 %; H, 3.06 %; N, 6.07 %; found: C, 31.05 %; H, 3.04 %; N, 5.98 %; FTIR: 3070-3052 cm-1 ν(C-H) aromatic, 2830 cm-1 ν(C-H) aliphatic, 1026 cm-1 ν(C-N) aliphatic, 1280 cm-1 ν(C-N) aromatic, 1481 cm-1 ν(C=C) aromatic, 1445 cm-1 ν(N=N), 406 cm-1 ν(Te-C); 1280 cm-1 ν(C-O) phenolic. 1H NMR (ppm): 6.70- 7.60 (m, 16H, aromatic), 2.31 (s, 6H, o-CH_3), 3.22 (t, 8H, N-CH_2 aliphatic), 1.52-1.56 (m, 12H, (-CH_2-)$_3$ aliphatic).

[Fe(otdpip)$_2$(OHPhTetBr)$_2$](5)- Empirical formula: $C_{36}H_{42}FeBr_6N_6O_2Te_2$; formula weight: 1381.59 gmol-1; yield: 71 % ; appearance: light brown crystals; m.p. = 155-157°C; Elem. analysis: calc. C, 31.30 %; H, 3.06 %; N, 6.08 %; found: C, 31.02 %; H, 3.05 %; N, 6.05 %; FTIR: 3095-3075 cm-1 ν(C-H) aromatic, 2820 cm-1 ν(C-H) aliphatic, 1026 cm-1 ν(C-N) aliphatic, 1286 cm-1 ν(C-N) aromatic, 1485 cm-1 ν(C=C) aromatic, 1449 cm-1 ν(N=N), 405 cm-1 ν(Te-C); 1280 cm-1 ν(C-O) phenolic. 1H NMR (ppm): 6.77- 7.20 (m, 16H, aromatic), 2.31 (s, 6H, o-CH_3), 3.25 (t, 8H, N-CH_2 aliphatic), 1.53-1.59 (m, 12H, (-CH_2-)$_3$ aliphatic).

2.2.2. General procedure for the synthesis of metal complexes (1:1:3) mole ratio :

A solution of the ligand was prepared by preparing the mixture of ligands and metal salts as follows: asolution of 1-(o-tolyldiazenyl)piperidine (otdpip) (0.5 mmol) in absolute ethanol (10 mL) was added to solution of 4-(hydroxyphenyl)tellurium tribromide (OHPhTetBr) (1.5 mmol) in absolute ethanol (10 mL) and the mixture was refluxed for 30 min and then NiCl2·6H2O, CoCl2·6H2O, CuBr2, ZnCl2 or FeBr2 (0.5 mmol) in absolute ethanol (10 mL) was added to the ligand solution with stirring and the reaction mixture was further stirred under reflux for 3 hrs. The obtained colored solution was left standing at room temperature to crystallize. The product was removed by filtration, washed with cooled absolute ethanol, recrystallized from methanol and dried. The analytical and physical data of the complexes are:

[Cu(otdpip)(OHPhTetBr)3](6)- Empirical formula: C30H29CuBr9N3O3Te3; formula weight: 1645.05 gmol-1; yield: 75 % ; appearance: yellowish - brown crystals; m.p. = 150-152°C; Elem. analysis: calc. C, 21.90 %; H, 1.78 %; N, 2.55 %; found: C, 21.60 %; H, 1.55%; N, 2.05 %; FTIR: 3090-3065 cm-1 ν(C-H) aromatic, 2825 cm-1 ν(C-H) aliphatic, 1020 cm-1 ν(C-N) aliphatic, 1284 cm-1 ν(C-N) aromatic, 1490 cm-1 ν(C=C) aromatic, 1449 cm-1 ν(N=N), 406 cm-1 ν(Te-C); 1275 cm-1 ν(C-O) phenolic. 1H NMR (ppm): 6.80- 6.98 (m, 7H, aromatic), 7.20- 7.29 (m, 9H, aromatic), 2.33 (s, 3H, o-CH3), 3.22 (t, 4H, N-CH2 aliphatic), 1.51-1.57 (m, 6H, (-CH2-)3 aliphatic).

[Ni(otdpip)(OHPhTetBr)3](7)- Empirical formula: C30H29NiBr9N3O3Te3; formula weight: 1640.2 gmol-1; yield: 65 % ; appearance: light brown crystals; m.p. = 193-195°C; Elem. analysis: calc. C, 21.97 %; H, 1.78 %; N, 2.56 %; found: C, 21.77 %; H, 1.58%; N, 2.15 %; FTIR: 3095-3068 cm-1 ν(C-H) aromatic, 2820 cm-1 ν(C-H) aliphatic, 1040 cm-1 ν(C-N) aliphatic, 1280 cm-1 ν(C-N) aromatic, 1495 cm-1 ν(C=C) aromatic, 1449 cm-1 ν(N=N), 406 cm-1 ν(Te-C); 1270 cm-1 ν(C-O) phenolic. 1H NMR (ppm): 6.85- 6.90 (m, 7H, aromatic), 7.25- 7.28 (m, 9H, aromatic), 2.34 (s, 3H, o-CH3), 3.21 (t, 4H, N-CH2 aliphatic), 1.52-1.59 (m, 6H, (-CH2-)3 aliphatic).

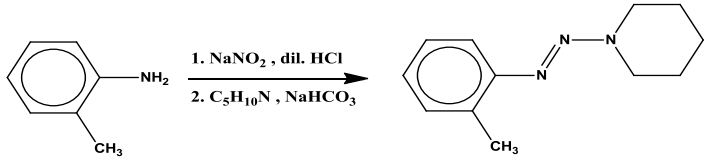
[Zn(otdpip)(OHPhTetBr)3](8)- Empirical formula: C30H29ZnBr9N3O3Te3; formula weight: 1646.89 gmol-1; yield: 76 % ; appearance: reddish - brown crystals; m.p. = 165-167°C; Elem. analysis: calc. C, 21.88 %; H, 1.77 %; N, 2.55 %; found: C, 21.68 %; H, 1.65%; N, 2.35 %; FTIR: 3085-3060 cm-1 ν(C-H) aromatic, 2830 cm-1 ν(C-H) aliphatic, 1025 cm-1 ν(C-N) aliphatic, 1282 cm-1 ν(C-N) aromatic, 1497 cm-1 ν(C=C) aromatic, 1445 cm-1 ν(N=N), 405 cm-1 ν(Te-C); 1270 cm-1 ν(C-O) phenolic. 1H NMR (ppm): 6.85 - 6.99 (m, 7H, aromatic), 7.22 - 7.28 (m, 9H, aromatic), 2.35 (s, 3H, o-CH3), 3.20 (t, 4H, N-CH2 aliphatic), 1.52-1.59 (m, 6H, (-CH2-)3 aliphatic).

[Co(otdpip)(OHPhTetBr)3](9)- Empirical formula: C30H29CoBr9N3O3Te3; formula weight: 1640.44 gmol-1; yield: 60 % ; appearance: yellowish - brown crystals; m.p. = 115-117°C; Elem. analysis: calc. C, 21.96 %; H, 1.78 %; N, 2.56 %; found: C, 21.65 %; H, 1.75%; N, 2.15 %; FTIR: 3095-3075 cm-1 ν(C-H) aromatic, 2820 cm-1 ν(C-H) aliphatic, 1027 cm-1 ν(C-N) aliphatic, 1280 cm-1 ν(C-N) aromatic, 1495 cm-1 ν(C=C) aromatic, 1445 cm-1 ν(N=N), 405 cm-1 ν(Te-C); 1270 cm-1 ν(C-O) phenolic. 1H NMR (ppm): 6.85- 6.90 (m, 7H, aromatic), 7.25- 7.30 (m, 9H, aromatic), 2.31 (s, 3H, o-CH3), 3.20 (t, 4H, N-CH2 aliphatic), 1.51-1.59 (m, 6H, (-CH2-)3 aliphatic).

[Fe(otdpip)(OHPhTetBr)3](10)- Empirical formula: C30H29FeBr9N3O3Te3; formula weight: 1637.35 gmol-1; yield: 62 % ; appearance: dark brown crystals; m.p. = 177-179°C; Elem. analysis: calc. C, 22.01 %; H, 1.79 %; N, 2.57 %; found: C, 21.95 %; H, 1.65%; N, 2.35 %; FTIR: 3092-3066 cm-1 ν(C-H) aromatic, 2830 cm-1 ν(C-H) aliphatic, 1025 cm-1 ν(C-N) aliphatic, 1287 cm-1 ν(C-N) aromatic, 1496 cm-1 ν(C=C) aromatic, 1450 cm-1 ν(N=N), 403 cm-1 ν(Te-C); 1270 cm-1 ν(C-O) phenolic. 1H NMR (ppm): 6.83- 6.90 (m, 7H, aromatic), 7.25- 7.30 (m, 9H, aromatic), 2.34 (s, 3H, o-CH3), 3.20 (t, 4H, N-CH2 aliphatic), 1.52-1.59 (m, 6H, (-CH2-)3 aliphatic).

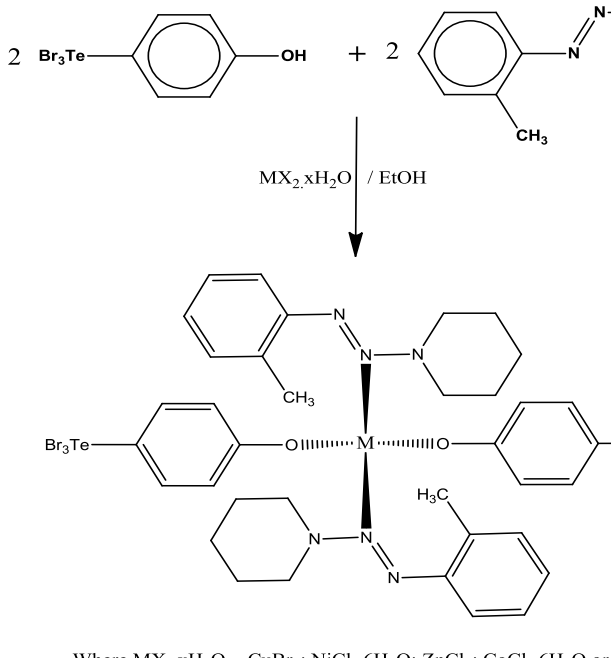
Results and discussion

The synthesis of triazene compound was performed by the reaction of diazotization of o- toluidine and piperidine based on literature with minor modification, to give the corresponding triazene 1-(o-tolyldiazenyl)piperidine, [Scheme 1.].



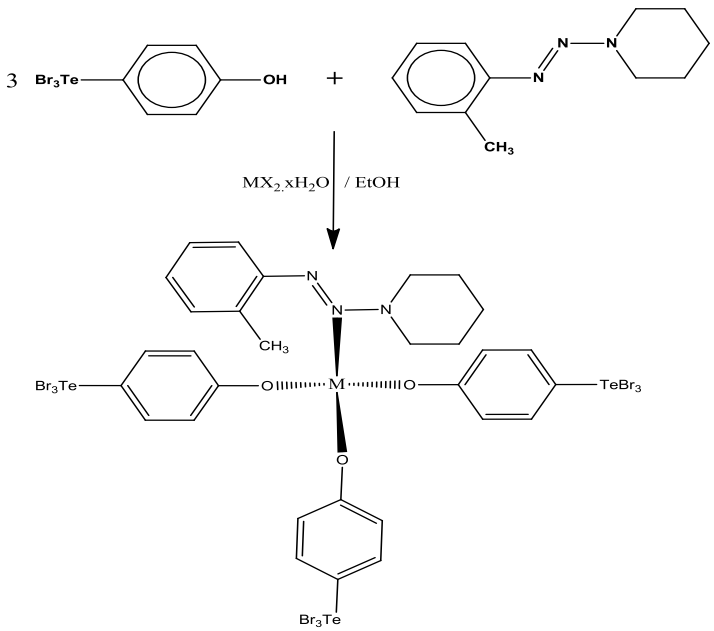
SCHEME 1. Preparative methods for 1-(o-tolyldiazenyl)piperidine

The metal – ligand complexes were synthesized at 1:2:2 and 1:1:3 molar ratio and obtained in pure form. Reactions of (Cu-, Ni-, Zn-, Co- and Fe- halides) with 1-(o-tolyldiazenyl)piperidine (otdpip) and 4-(hydroxyphenyl)tellurium tribromide (OHPhTetBr) in 1:2:2 molar ratio in ethanol are represented in Scheme 2.



SCHEME 2. Preparative methods for complexes (1:2:2) molar ratio

Reactions of (Cu-, Ni-, Zn-, Co- and Fe- halides) with 1-(o-tolyldiazenyl)piperidine (otdpip) and 4-(hydroxyphenyl)tellurium tribromide (OHPhTetBr) in 1:1:3 molar ratio in ethanol are represented in Scheme 3.



Where $MX_2.xH_2O$ = $CuBr_2$; $NiCl_2.6H_2O$; $ZnCl_2$; $CoCl_2.6H_2O$ or $FeBr_2$

SCHEME 3. Preparative methods for complexes (1:1:3) molar ratio

The complexes are air- stable, insoluble in water, partly soluble in ethanol and soluble in DMSO and DMF, non hygroscopic and colored solids crystals. The elemental analysis data (CHN) of ligands and all their complexes are in a good agreement with the calculated values as shown in Table 1.

Table 1.

Physical and analytical data for ligand and compounds 1-10

Comp.	color	Melting point (℃)	Analysis (%)a			Yield %
			C	H	N	
C12H17N3 (ligand)	Reddish - brown	70-71	70.90 (70.53)	8.43 (8.21)	20.67 (20.55)	91
C36H42CuBr6N6O2Te2 (1)	Brown	162- 164	31.13 (31.02)	3.05 (3.01)	6.05 (6.00)	66
C36H42NiBr6N6O2Te2 (2)	Light brown	185-186	31.24 (31.10)	3.06 (3.05)	6.07 (6.02)	73
C36H42ZnBr6N6O2Te2 (3)	Yellowish - Brown	102-105	31.09 (31.00)	3.04 (2.95)	6.08 (5.98)	59

C36H42CoBr6N6O2Te2(4)	Reddish - Brown	180 -181	31.23 (31.05)	3.06 (3.04)	6.07 (5.98)	68
C36H42FeBr6N6O2Te2 (5)	Light - brown	155-157	31.30 (31.02)	3.06 (3.05)	6.08 (6.05)	71
C30H29CuBr9N3O3Te3(6)	Yellowish - Brown	150 - 152	21.90 (21.60)	1.78 (1.55)	2.55 (2.05)	75
C30H29NiBr9N3O3Te3(7)	Light - brown	193 - 195	21.97 (21.77)	1.78 (1.58)	2.56 (2.15)	65
C30H29ZnBr9N3O3Te3 (8)	Reddish - brown	165-167	21.88 (21.68)	1.77 (1.65)	2.55 (2.35)	76
C30H29CoBr9N3O3Te3(9)	Yellowish - Brown	115 - 117	21.96 (21.65)	1.78 (1.75)	2.56 (2.15)	60
C30H29FeBr9N3O3Te3(10)	Dark brown	177-179	22.01 (21.95)	1.79 (1.65)	2.57 (2.35)	62

a calculated values are in parentheses

The IR spactra of the ligands and complexes synthesized are shows two strong bands appeared at range (1020-1101) cm-1 and (1280-1356) cm-1 due to stretching aliphatic (C-N) and aromatic (C-N) respectively.(10, 11) The IR spactra of compounds 1-11 showed a strong band in range (1435-1450) cm-1 can be attributed to (N=N) bond.(10, 11) The sharp band of medium intensity occurred at range (1450-1504) cm-1 is attributed to (C=C) aromatic. The strong as well as sharp band at range (1270-1285) cm-1 is due to the (C-O phenolic) stretching vibrations.(10, 11) The (Te-C) stretching vibrations appeared at range (401-407) cm-1 in p-hydroxyphenyl telluriumtribromide, this shift confirms the participation of oxygen on the C-O-M bond.

All the IR data suggest that the metal was bonded to the ligand through the phenolic oxygen and azo – groups in triazole. As shown in Table 2.

Table 2.

IR Spectroscopic data for Ligand and compounds 1-10

Ligand/ complexes IR (cm-1)	λmax in nm (ε in M-1cm-1)
C12H17N3 (ligand)	3022-3020 ν(C-H) aromatic, 2855 ν(C-H) aliphatic, 1101 ν(C-N) aliphatic, 1356 ν(C-N) aromatic, 1504 ν(C=C) aromatic, 1436 ν(N=N).
C36H42CuBr6N6O2Te2 (1)	3075-3055 ν(C-H) aromatic, 2810 ν(C-H) aliphatic, 1024 ν(C-N) aliphatic, 1283 ν(C-N) aromatic, 1483 ν(C=C) aromatic, 1449 ν(N=N), 401 ν(Te-C); 1279 ν(C-O) phenolic
C36H42NiBr6N6O2Te2 (2)	3075-3050 ν(C-H) aromatic, 2830 ν(C-H) aliphatic, 1023 ν(C-N) aliphatic, 1280 ν(C-N) aromatic, 1485 ν(C=C) aromatic, 1450 ν(N=N), 405 ν(Te-C); 1285 ν(C-O) phenolic

C36H42ZnBr6N6O2Te2 (3)	3065-3050 ν(C-H) aromatic, 2830 ν(C-H) aliphatic, 1026 ν(C-N) aliphatic, 1290 ν(C-N) aromatic, 1450 ν(C=C) aromatic, 1435 ν(N=N), 407 ν(Te-C); 1279 ν(C-O) phenolic
C36H42CoBr6N6O2Te2(4)	3070-3052 ν(C-H) aromatic, 2830 ν(C-H) aliphatic, 1026 ν(C-N) aliphatic, 1280 ν(C-N) aromatic, 1481 ν(C=C) aromatic, 1445 ν(N=N), 406 ν(Te-C); 1280 ν(C-O) phenolic
C36H42FeBr6N6O2Te2 (5)	3095-3075 ν(C-H) aromatic, 2820 ν(C-H) aliphatic, 1026 ν(C-N) aliphatic, 1286 ν(C-N) aromatic, 1485 ν(C=C) aromatic, 1449 ν(N=N), 405 ν(Te-C); 1280 ν(C-O) phenolic
C30H29CuBr9N3O3Te3(6)	3090-3065 ν(C-H) aromatic, 2825 ν(C-H) aliphatic, 1020 ν(C-N) aliphatic, 1284 ν(C-N) aromatic, 1490 ν(C=C) aromatic, 1449 ν(N=N), 406 ν(Te-C); 1275 ν(C-O) phenolic
C30H29NiBr9N3O3Te3(7)	3095-3068 ν(C-H) aromatic, 2820 ν(C-H) aliphatic, 1040 ν(C-N) aliphatic, 1280 ν(C-N) aromatic, 1495 ν(C=C) aromatic, 1449 ν(N=N), 406 ν(Te-C); 1270 ν(C-O) phenolic
C30H29ZnBr9N3O3Te3 (8)	3085-3060 ν(C-H) aromatic, 2830 ν(C-H) aliphatic, 1025 ν(C-N) aliphatic, 1282 ν(C-N) aromatic, 1497 ν(C=C) aromatic, 1445 ν(N=N), 405 ν(Te-C); 1270 ν(C-O) phenolic
C30H29CoBr9N3O3Te3(9)	3095-3075 ν(C-H) aromatic, 2820 ν(C-H) aliphatic, 1027 ν(C-N) aliphatic, 1280 ν(C-N) aromatic, 1495 ν(C=C) aromatic, 1445 ν(N=N), 405 ν(Te-C); 1270 ν(C-O) phenolic.
C30H29FeBr9N3O3Te3(10)	3092-3066 ν(C-H) aromatic, 2830 ν(C-H) aliphatic, 1025 ν(C-N) aliphatic, 1287 ν(C-N) aromatic, 1496 ν(C=C) aromatic, 1450 ν(N=N), 403 ν(Te-C); 1270 ν(C-O) phenolic

1H NMR spectra of compounds 1-11 were recorded in DMSO-d6 and shows in Table 3. The 1H NMR spectrum of compounds shows a multiple broad signal at the range 6.68-7.30 ppm can be assigned to protons in aromatic ring of phenyl groups. The protons in N–(CH2)2 can be obtained at range (3.20-3.25) ppm as triplet signal, also a singlet signal can be appeared at range (2.30-2.35) ppm due to o-methyl group in triazene. (10, 11, 12-14) The protons in (-CH2-)3 can be appeared at the range (1.51 – 1.59) ppm as a multiple signal, (10, 12-16).

Table 3.
1H NMR Spectroscopic data for Ligand and compounds 1-10

Ligand/ complexes	Chemical shift (ppm)
C12H17N3 (ligand)	6.90- 7.28 (m, 4H, Ar), 2.35 (s, 3H, o-CH3), 3.10 (t, 4H, N-CH2 aliphatic), 1.51-1.59 (m, 6H, (-CH2-)3 aliphatic)
C36H42CuBr6N6O2Te2 (1)	6.99- 7.30 (m, 16H, Ar), 2.35 (s, 6H, o-CH3), 3.20 (t, 8H, N-CH2 aliphatic), 1.51-1.57 (m, 12H, (-CH2-)3 aliphatic)
C36H42NiBr6N6O2Te2 (2)	6.68- 7.20 (m, 16H, Ar), 2.35 (s, 6H, o-CH3), 3.22 (t, 8H, N-CH2 aliphatic), 1.52-1.59 (m, 12H, (-CH2-)3 aliphatic)
C36H42ZnBr6N6O2Te2 (3)	6.87- 7.22 (m, 16H, Ar), 2.30 (s, 6H, o-CH3), 3.22 (t, 8H, N-CH2 aliphatic), 1.53-1.59 (m, 12H, (-CH2-)3 aliphatic)
C36H42CoBr6N6O2Te2(4)	6.70- 7.60 (m, 16H, Ar), 2.31 (s, 6H, o-CH3), 3.22 (t, 8H, N-CH2 aliphatic), 1.52-1.56 (m, 12H, (-CH2-)3 aliphatic)
C36H42FeBr6N6O2Te2 (5)	6.77- 7.20 (m, 16H, Ar), 2.31 (s, 6H, o-CH3), 3.25 (t, 8H, N-CH2 aliphatic), 1.53-1.59 (m, 12H, (-CH2-)3 aliphatic)
C30H29CuBr9N3O3Te3(6)	6.80- 6.98 (m, 7H, Ar), 7.20- 7.29 (m, 9H, Ar), 2.33 (s, 3H, o-CH3), 3.22 (t, 4H, N-CH2 aliphatic), 1.51-1.57 (m, 6H, (-CH2-)3 aliphatic).
C30H29NiBr9N3O3Te3(7)	6.85- 6.90 (m, 7H, Ar), 7.25- 7.28 (m, 9H, Ar), 2.34 (s, 3H, o-CH3), 3.21 (t, 4H, N-CH2 aliphatic), 1.52-1.59 (m, 6H, (-CH2-)3 aliphatic)
C30H29ZnBr9N3O3Te3 (8)	6.85 - 6.99 (m, 7H, Ar), 7.22 - 7.28 (m, 9H, Ar), 2.35 (s, 3H, o-CH3), 3.20 (t, 4H, N-CH2 aliphatic), 1.52-1.59 (m, 6H, (-CH2-)3 aliphatic)

C30H29CoBr9N3O3Te3(9)	6.85- 6.90 (m, 7H, Ar), 7.25- 7.30 (m, 9H, Ar), 2.31 (s, 3H, o-CH3), 3.20 (t, 4H, N-CH2 aliphatic), 1.51-1.59 (m, 6H, (-CH2-)3 aliphatic)
C30H29FeBr9N3O3Te3(10)	6.83- 6.90 (m, 7H, Ar), 7.25- 7.30 (m, 9H, Ar), 2.34 (s, 3H, o-CH3), 3.20 (t, 4H, N-CH2 aliphatic), 1.52-1.59 (m, 6H, (-CH2-)3 aliphatic)

All complexes were contained at the same ligands but with different molar ratio therefore; seem converged results for 1H NMR and disappearance of the OH band from 1H NMR spectra refer to coordinate ligand (OHPhTetBr) from OH group and composing the bond (M-O-C) in this complexes. In conclusion, several new coordination compounds containing triazene and organotellurium were prepared in this study. These new compounds may be used as dyes for industrial processes.

References:

1- A- M. K. Rofouel, M. R. Melardi, M. Barkhi and H. R. Khalili Ghaydar; Anal. Sci.; 24, 81x, (2008). B- H. Kantekin , G. Sarkı, A. Koca, O. Bekircan , A. Aktas, R.Z. Uslu Kobak, and M.B.Sa_glam, Journal of Organometallic Chemistry; 789, 53, (2015)
2- I. D. Sadekov, A. A. Maksimenko, A. G. Malakov, G. M. Abakrov and V. I. Minkin, Khim. Grterotsiki. Soedin, 1426 (1988).
3- H. Zollinger, Angew. Chem. Int. Ed., 41, 3338 (2002).
4- J. J. Escobar-Nuricumbo, C. Campos-Alvarado, G. Ríos-Moreno, D.Morales-Morales, P. J. Walsh, M. Parra-Hake, Inorg. Chem., 46, 6182 (2007).
5- A-R. Shanmugakala, P. Tharmaraj, C. D. Sheela, and C. Anitha; Inter. Jour. of Inor. Chemistry; 1, (2012).B- L. Álvarez-Rodríguez, J. A. Cabeza∗, P. García-Álvarez∗and D. Polo; Coordination Chemistry Reviews, 300, 1, (2015).
6- S. A. Al-Jadaan, M.Sc. Thesis, University of Basrah, (1990).
7- T. Chin, A. Phipps, F. R. Fronczek and R. Isovitsch, J. Heterocyclic Chem., 48, 215 (2011).
8- S. Sengupta and S. K. Sadhukhan, Organic Syntheses, Coll., 10, 263, (2004) , 79, 52 (2002).
9- A. Z. Al-Rubaie, A. M. Fingan, N. I. Al-Salim and S. A. Al-Jadaan, Polyhedron , 14 , 2575 (1995).
10- R. M. Silerstien, F. X. Webster and D. J. Kiemle, "Spectrometric Identification of Organic Chemistry Compounds ", 6th Ed. , John Wiley & Sons , N. Y, (2005).
11- R. I. Shriner and C. K. Hermann," Spectroscopic Techniques for Organic Chemistry ", John Wiley & Sons, N. Y, (2004).
12- M. Shanmugapriya, A. Abdul Jameel and M. Syed Ali Padusha, Int. J. PharmTech Res., 4, 85 (2012).
13- M. J. Fernández-Torres, International Journal of Chemistry; 2, 32 (2013).
14- P. Panneerselvam and M. Karikalan, International Journal of Pharma and Bio Sciences, 2, 267 (2011).
15- P. Perumal, T. Sivakkumart, N. Kannappan and R. Manavalan, International Journal of Pharmacy and Pharmaceutical Sciences, 5, 317 (2013).
16- A. Thangamani, J. Jayabharathi and A. Manimekalai, J. Chem. Sci., 122, 579 (2010).