See discussions, stats, and author profiles for this publication at: https://www.researchgate.net/publication/230453411

Synthesis, characterization, and computational study of some new organotellurium compounds containing azomethine groups

Article in Heteroatom Chemistry · April 2008

Study of Structures and Electronic Properties View project

Synthesis of Organoselenium Compounds Containing Nitrogen View project

DOI: 10.1002/hc.20437

CITATIONS 7		READS 123			
5 autho	rs, including:				
	Ali Al-Rubaie Al-Ayen University-Thi Qar + University of Basrah 71 PUBLICATIONS 334 CITATIONS		Wasfi A. Al-Masoudi University of Basrah 55 PUBLICATIONS 101 CITATIONS		
	SEE PROFILE		SEE PROFILE		
	Shaker A.N. Al-Jadaan University of Basrah		Ali Jameel Hameed University of Basrah		
	87 PUBLICATIONS 75 CITATIONS SEE PROFILE		38 PUBLICATIONS 367 CITATIONS SEE PROFILE		
C	SEE PROFILE		SEE PROFILE		

All content following this page was uploaded by Ali Jameel Hameed on 30 May 2018.

Synthesis, Characterization, and Computational Study of Some New Organotellurium Compounds Containing Azomethine Groups

Ali Z. Al-Rubaie,¹ Wasfi A. Al-Masoudi,² Shaker A. N. Al-Jadaan,³ Abraham F. Jalbout,⁴ and Ali Jameel Hameed¹

¹Department of Chemistry, College of Science, University of Basrah, Basrah, Iraq

²Department of Chemistry, College of Arts and Science, Tarhona, Almergab University, P.O. Box 48211, Tarhona, Libya

³Department of Chemistry, College of Pharmacy, University of Basrah, Basrah, Iraq

⁴Department of Chemistry, University of Arizona, Tucson, AZ 85721

Received 17 October 2006; revised 21 June 2007

ABSTRACT: The reaction of ortho-mercurated anilines with benzaldehyde gave the ortho-mercurated Schiff bases. The reaction of the mercurated Schiff bases with tellurium tetrabromide in 1:1 and 2:1 mole ratio using dry chloroform as solvent gave the ortho-tellurated Schiff bases compounds ArTeBr₃ and Ar₂TeBr₂, respectively, in good yields (where $Ar = 5 - ClC_6H_3N = CHC_6H_5$, $5 - BrC_6H_3N = CHC_6H_5$, $5 - BrC_6H_5$, 5 - BrC $CH_3OC_6H_3N = CHC_6H_5$, and $5 - NO_2C_6H_3N = CHC_6H_5$). The reduction of ArTeBr₃ by hydrazine hydrate gave the corresponding ditelluride (i.e., $Ar_{2}Te_{2}$). Treatment of Ar₂TeBr₂ with hydrazine hydrate afforded tellurides (Ar_2Te) in good yields. Attempts to prepare the corresponding aryl tellurenyl bromides, ArTeBr, by partial reduction of ArTeBr₃ with various reducing agents were unsuccessful. All these new compounds were characterized by microanalysis, ¹H, and ¹³C NMR, IR, and mass spectroscopic data. A computational study for the $Te \rightarrow N$ interactions of all compounds was calculated using the GAUSSIAN 03 program pack-

© 2008 Wiley Periodicals, Inc.



age. © 2008 Wiley Periodicals, Inc. Heteroatom Chem 19:307–315, 2008; Published online in Wiley InterScience (www.interscience.wiley.com). DOI 10.1002/hc.20437

INTRODUCTION

Direct telluration of Schiff bases by tellurium tetrahalides resulted in formation of ionic products, arising from hydrolysis of tellurium tetrahalides [1–3]. Mercuration of Schiff bases followed by a trans-metallation reaction with tellurium tetrabromide provided a satisfactory method to prepare [4-methoxy-2-(phenyliminomethyl)phenyl] tellurium tribromide in 42% yield, which on reduction gave the stable tellurium(II) derivative in 30% yield [3]. The mercurated Schiff bases also reacted with *p*-ethoxyphenyl tellurium trichloride to produce the corresponding dichloride [3], namely, (4-ethoxyphenyl)[4-methoxy-2-(phenyliminomethyl)-phenyl] tellurium dichloride, in 70% yield [3].

Sadekov et al. [4-8] made considerable contributions to the chemistry of tellurated azomethines, and several tellurated Schiff bases were

Correspondence to: Ali Z. Al-Rubaie; e-mail: alrubaie49@ yahoo.com.

		М.Р. (°С)	Yield (%)				
Compound	ınd Color			С	Н	N	ν(C == N)
1	Yellow	137–139	83	26.42 (26.81)	1.39 (1.45)	2.16 (2.40)	1555s
2	Yellow	155–157	75	24.74 (24.93)	1.47 (1.59)	2.06 (2.23)	1560s
3	Yellow	39–141	72	28.73 (29.01)	1.94 (2.07)	2.11 (2.42)	1553s
4	Brown	171–173	80	25.92 (26.35)	1.38 (1.53)	4.3 (4 .73) [′]	1545s
5	Yellow	137–139	52	43.24 (43.57)	2.48 (2.53)	3.59 (3.91)	1535s
6	Yellow	174–176 ^b	65	46.78 (47.04)	2.96 (3.04)	3.85 (4.22)	1555s
7	Yellow	112–114 ^b	77	47.46 (47.51)	3.21 (3.42)	3.92 (3.96)	1559s
8	Yellow	172–174	74	41.97 (42.32)	2.37 (2.46)	7.27 (7.59)	1554s
9	Orange	122-124	64	45.31 (45.60)	2.51 (2.63)	3.76 (4.09)	1557s
10	Red-brown	134–136	60	43.42 (42.32)	2.39 (2.57)	7.65 (7.59)	1552s
11	Pale Yellow	202–204	52	54.04 (54.31)	3.43 (3.51)	4.55 (4.87)	1550s
12	Yellow	184–186	45	47.98 (48.34)	2.51 (2.78)	4.02 (4.33)	1560s
13	Yellow	163–165	73	57.68 (57.57)	4.03 (4.41)	3.98 (4.79)	1545s
14	Yellow	149–151	76	51.12 (51.39)	3.26 (̀3.38)́	9.06 (9.40)	1548s

TABLE 1 Some Physical and Analytical Data for Compounds 1–14

^aCalculated values are given in parentheses.

^bDecomposed.

prepared and characterized [4–8]. The ability of β -bromotellurenylvinylaldehydes [8–11] and monohalotellurobenzaldehydes [12] to form Schiff bases enabled this methodology to synthesize a wide range of organotellurium compounds containing –CH=N groups [12]. Such compounds were used as precursors or proposed as intermediates for the synthesis of heterocyclic tellurium compounds, such as 5*H*-1.2-oxatelluroles and their benzo analogs, isotellurazoles, 1,2,3-telluradiazines, 1,2,6-oxatellurazocines, and other related tellurium heterocycles [8]. Molecular structures of some of these tellurated azomethines in the solid state indicated the existence of the intramolecular Te \leftarrow N bond [5,6,12–15].

Recently, Singh and coworkers [16] reported the preparation of some tellurated Schiff bases 4-MeOC₆H₄TeCH₂CH₂N=C(CH₃)C₆H₄-2-OH and $2-HOC_6H_4(CH_3)C=NCH_2CH_2TeCH_2CH_2N=C(CH_3)$ C₆H₄-2-OH from the reaction of {2-[(4methoxyphenyl)telluro]ethyl}amine bis(2and aminoethyl) telluride with o-hydroxyacetophenone, respectively [16]. Furthermore, the condensation of bis(o-formylphenyl) diselenide with (R)-(+)-1-phenylethylamine afforded the chiral azomethine diselenide. (1-phenylethylimino)methyl) phenyl)diselanyl)benzylidene-1-phenylethanamine [17]. On the other hand, a recent study described the synthesis of monomeric tellurides of type $4-RC_{6}H_{4}(2-(4,4'-NO_{2}C_{6}H_{4}CH=NC_{6}H_{3}-Me)Te$ as the first example of a telluride with $1,4-\text{Te}\cdots N$ intramolecular interaction [18].

Some years ago, we prepared several organic tellurium compounds containing amino groups by reacting aminoarylmercury chlorides with tellurium tetrabromide in glacial acetic acid [19,20]. Thus, we decided to use these mercurated anilines as precursors to prepare a new series of mercurated Schiff bases, from which a number of hitherto unavailable organotellurium compounds containing azomethine groups were prepared.

RESULTS AND DISCUSSION

Synthesis

Isolated yields, melting points, color and carbon, hydrogen, and nitrogen analytical data for all new organotellurium compounds are given in Table 1. The present work describes the synthesis of new tellurated Schiff bases by reaction of the corresponding mercurated Schiff bases, which in turn prepared from the reaction of aryl mercury chloride with benzaldehyde, with tellurium tetrabromide in 1:1 ratio to produce the required tellurium containing materials (ArTeBr₃), that is, compounds **1–4** (Scheme 1). On the other hand, when tellurated Schiff bases and tellurium tetrabromide brought together in 2:1 ratio gave the dioranyltellurium dibromides (compounds **5–8**) in fair yields (Table 1).

Reduction of compounds 1 and 4 by ethanolic solution of hydrazine hydrate gave the corresponding ditellurides (9 and 10) in reasonable yields (Table 1). Furthermore, the diorganyl tellurides 11, 12, 13, and 14 were prepared by the reduction of compounds 5, 6, 7, and 8, respectively by hydrazine hydrate in boiling ethanol. Attempt to prepare the corresponding tellurenyl bromides (i.e., Ar-TeBr) from partial reduction of compounds 1 or 4 by various reducing agents, such as Na₂S·9H₂O, NaBH₄, and hydrazine hydrate or sodium metabisulfite, were unsuccessful and yielded the corresponding



SCHEME 1 Preparative methods for compounds 1–14.

ditellurides. This is probably due to the absence of the interaction between nitrogen atom of -CH=N- group and tellurium atom.

IR spectra for all compounds displayed common features in certain regions and characteristic bands in the fingerprint and other regions. The IR spectra confirm the presence of the azomethine group (-CH=N-) stretching with a sharp region around 1550 cm⁻¹ (Table 1). Furthermore, a complete condensation of primary amino groups is confirmed by the lack of N-H stretching bands in the IR spectra of all mercurated and tellurated Schiff bases. This conclusion is also supported by ¹H NMR data, which show the presence of CH=N hydrogen resonances

in the region 8.42–9.22 ppm (Table 2). In general, the ¹H NMR spectra of all compounds show all the expected peaks in proper intensity ratio (Table 2). It is worthy to note that the proton of azomethine resonate as a singlet between 9.17–9.22 ppm for compounds **1–8** and **11–14**, whereas for the ditellurides **9** and **10** appeared at 8.72 and 8.42 ppm, respectively. The main reason for this shift toward a stronger field as compared with compounds **1–8** and **11–14** may be attributed to the coordination of both azomethine groups through their nitrogen atoms with both tellurium atoms forming two fivemembered chelate ring structures in solution, as shown below:

Compounds	¹ H NMR (DMSO-d ₆); TMS = 0 ppm	^{13}C NMR (DMSO-d ₆); TMS = 0 ppm
1	7.38 (d, 1H, H-6); 7.45 (dd, 1H, H-5); 7.56 (d, 1H, H-3); 7.62–7.73 (m, 3H, H-3', H-4', H-5'); 7.81–7.91 (m, 2H, H-2', H-6'); 9.19 (s, 1H,CH=N).	122.8 (C-2); 123.3 (C-6): 128.5 (C-3', C-5'); 129.0 (C-2', C-6'); 130.0 (C-3); 130.2 (C-5); 131.0 (C-4'); 132.1 (C-4); 133.5 (C-1'); 151.1 (C-1); 160.0 (-CH=N-)
3	3.51 (s, 3H, OCH ₃); 7.02 (d, 1H, H-6); 7.11–7.20 (m, 2H, H-3, H-5); 7.62–7.74 (m, 2H, H-1, H-3); 7.62–7.74 (m, 3H, H-3', H-4', H-5'); 7.82–8.19 (m, 2H, H-2', H-6'); 9.17 (s, 1H, -CH=N–).	55.4 (OCH ₃); 116.0 (C-3); 115.2 (C-5); 121.0 (C-2); 123.1 (C-6); 128.2 (C-3', C-5'); 129.0 (C-2', C-6'); 130.8 (C-4'); 133,6 (C-1'); 146.2 (C-1); 159.0 (C-4); 160.0 (-CH=N-).
5	7.35 (d, 2H, H-6), 7.44 (dd, 2H, H-5); 7.58 (d, 2H, H-3); 7.52–7.61 (m, 6H, H-3', H-4', H-5'); 7.79–8.14 (m, 4H, H-3', H-6'); 9.21 (s, 2H,CH=N).	
6	7.39 (dd, 2H, H-5); 7.54 (d, 2H, H-6); 7.62 (d, 2H, H-3); 7.72–7.78 (m, 6H, H-3', H-4', H-5'); 7.82–8.02 (m, 4H, H-2', H-6'); 9.15 (s, 2H,CH=N-).	121.2 (C-4); 122.7 (C-2); 124.3 (C-6); 128.7 (C-3', C-5'); 129.1 (C-2', C-6'); 131.2 (C-4'); 132.8 (C-3); 133.1 (C-5); 133.8 (C-1'); 146.2 (C-1); 160.1 (-CH=N-).
8	7.53 (d, 2H, H-6); 7.38–7.47 (m, 6H, H-3′, H-4′, H-5′); 7.78–8.12 (m, 8H, H-2′, H-6′, H-3, H-5); 9.22 (S, 2H, —CH=N–).	122.4 (C-5); 122.9 (C-2); 123.2 (C-6); 124.8 (C-3); 128.8 (C-3, C-5'); 129.1 (C-2', C-6'); 131.2 (C-4'); 133.6 (C-1'); 147.2 (C-4); 158.8 (C-1); 160.1 (-CH=N-)
9	7.35 (d, 2H, H-6); 7.44 (dd, 2H, H-5); 7.56 (d, 2H, H-3); 7.61–7.72 (m, 6H, H-3', H-4', H-5'); 7.79–7.94 (m, 4H, H-2', H-6'); 8.72 (s, 2H, —CH=N—).	123.0 (C-2); 123.3 (C-6): 128.8 (C-3', C-5'); 129.8 (C-2', C-6'); 130.1 (C-3); 131.0 (C-5); 132.0 (C-4'); 132.5 (C-4); 133.6 (C-1'); 150.2 (C-1); 160.2 (-CH=N-)
10	7.42 (d, 2H, H-6); 7.52–7.60 (m, 6H, H-3, H-4', H-5'); 7.80–8.15(m, 8H, H-3', H-6, H-2', H-6'); 8.42 (s, 2H, —CH=N–).	121.8 (C-5); 122.3 (C-2); 123.0 (C-6); 124.6 (C-3); 128.8 (C-3', C-5'); 129.2 (C-2', C-6'); 131.2 (C-4'); 133.3 (C-1'); 148.1 (C-4); 157.4 (C-1); 160.1 (-CH=N)
13	3.54 (s, 6H, OCH ₃); 6.92 (d, 2H, H-6); 7.20–7.28 (m, 4H, H-3, H-5); 7.62–7.74 (m, 6H, H-3', H-4', H-5'); 7.92–8.24 (m, 4H, H-2', H-6'); 9.22 (s, 2H, −CH=N–)	
14	7.38 (d, 2H, H-6); 7.42–7.55 (m, 6H, H-3', H-4', H-5'); 7.82–8.18 (m, 8H, H-3, H-6, H-2', H-6'); 9.20 (s, 2H, —CH=N–).	

TABLE 2 ¹H and ¹³C NMR Spectral Data for Selected Compounds^a

^aHydrogen and carbon assignment as illustrated below:





 $R = Cl (9); R = NO_2 (10)$

This supports our argument on the inability to prepare the corresponding tellurenyl halides from tribromides by partial reduction, though the theoretical data on the solid compounds disagree with this (see Computational Studies section).

Carbon-13 NMR spectra gave further support to the formulation of these new compounds (Table 2). The spectra revealed the presence of -CH=N- group around 160 ppm. The large variation for carbon atoms bearing tellurium may be attributed to the polarity of Te-C bond [22].

In the mass spectra (EI) of compounds 4, 5, 9, and 10, the molecular ion peaks [M+] were

observed with low-relative intensity at 592 (5%), 688 (11%), 710 (4.4%), and 718 (3%). Important fragments were also observed, confirming the proposed structures.

COMPUTATIONAL STUDY

To simulate the geometric structures of 9 and 10 and to reveal the $Te \cdots N$ interactions in such a kind of ditellurides, we employed theoretical calculations to achieve a better understanding. All the calculations were performed by using the GAUSSIAN 03 program package [23]. The B3LYP method of DFT was used to optimize all the structures reported. A basis set of lanl2dz [24] was chosen for Te atoms and 6-31G^{*} for all other atoms. Frequency calculations were performed at the same level of theory as geometry optimizations. All the structures reported are confirmed to be local minima from frequency analysis. The optimized structures of different conformers of our computational models are given in Fig. 1, and the distances of Te and N as well as the energy values of the different conformers of each model are depicted in Table 3.

The three different conformers of model 1 are shown in Fig. 1, and in this model all the benzene rings are omitted. Conformer A has the lowest energy. The Te1···N1 and Te2···N2 distances are both 3.178 Å. Although this distance is longer than the reported $Te \cdots N$ secondary interactions that are all lower than 3.0 Å [25–28], it is still much shorter than 3.7 Å [29], which is the sum of van der Waals radii of Te and N. The structure of **B** shows two five-membered rings can be formed because the distances of Te2···N1 and Te1···N2 are both only 3.022 A. However, the higher energy of **B** than **A** indicates the formations of five-membered rings though Te \cdots N secondary interactions are not favored. In **C**, there has no interaction between the Te atoms and N atoms at all; thus, there is no surprise and this conformer has the highest energy. From this computational model, we can conclude that in such kind of ditellurides the formation of five-membered rings through Te2...N1 and Te1...N2 interactions is not favored, and the formation of four-membered rings through Te1...N1 and Te2...N2 interactions is preferred. This conclusion is confirmed in the calculation of model 2. In this model, two benzene rings connected to the Te atoms are introduced. Although in conformer E, there may exist interactions between all the Te atoms and all the N atoms and two fivemembered rings can be formed, the more stable conformer is **D** that only has two secondary interactions of Te1···N1 and Te2···N2.

To further confirm our conclusion and to reveal the true situation in 9 and 10, we carried out calculations with model 3. In this model, the Cl and NO_2 groups connected to the benzene rings in 9 and **10** are omitted, and we suppose that these simplifications will not induce dramatic change in the geometric structures and stabilities of the conformers. Table 3 shows **F** is the most stable conformer, and there is no exception that in this conformer Te1...N1 and Te2...N2 secondary interactions are existed to form two four-membered rings. In conformer I, there may exist certain interactions of Te2···N1 and Te1···N2; however, higher energy is calculated. From the structures of the five conformers, we can understand this matters influence the stabilities of different conformers. G is the second stable conformer because in this conformer the two benzene rings connected to the imine moieties are both located at the antipositions of the C=N double bond. Thus, the repulsions between the two benzene rings connected at the two ends of the same imine moiety are relieved, and such kind of repulsion can be detected in following conformers H, I, and J. It is reasonable that there may have repulsion between the two benzene rings, which are both connected at the C atoms of the two imine moieties in conformer **G**, whereas in conformer **F** this is not any kind of repulsion between the benzene rings at all.

From calculations, we know the structures of 9 and 10 must be similar to conformer F in model 3. There are secondary interactions between the Te atom and N atoms, which are connected at two neighboring carbons of the same benzene ring, and four-membered rings are expected to be formed as a result of secondary interactions, whereas the formation of five-membered rings is not favorable. The repulsions between the benzene rings in 9 and 10 influence the stabilities of the conformers mostly, and the structures in the case of F are ideal.

On the basis of our conclusion on the structures of **9** and **10**, we believe that there have also been such kinds of secondary Te—N interactions in all other organotellurium compounds in our experiment. As shown in Fig. 2, in the optimized skeletal structures of **1–4**, **5–8**, and **11–14**, the Te—N secondary interactions are obvious. In compounds **1–4**, the secondary interaction between Te and N is supposed to be the strongest, since the Te—N distance is only 2.561 Å, which is the shortest. In compounds **5–8** and **11–14**, the distance between Te and N is longer, which is all around 3.0 Å and still in the range of secondary interaction. These observations agree well with the X-ray crystallography study on analogous compounds [18].



FIGURE 1 Different conformers of the three computational models. **A**, **B**, and **C** are different conformers of model 1; **D** and **E** are two conformers of model 2; **F**, **G**, **H**, **I**, and **J** are different conformers of model 3.

In conclusion, a new series of organotellurium compounds containing -CH=N- group was prepared. It was disappointing that on reduction compounds **1** and **4** by various reducing agents gave ultimately the corresponding ditellurides instead of organotellurium(II) halides (RTeX). Evidence is available to suggest that some of these organotellurium compounds (i.e., telluride and ditellurides) form complexes with a range of metal salts.

EXPERIMENTAL

Physical Measurements

¹H and ¹³C NMR spectra were obtained with Bruker LA-250 (250 MHz) and Bruker 300 spectrometer instruments. They were recorded in DMSO- d_6 solutions containing TMS as internal standard. Elemental analyses (C, H, and N) were performed by Analytical Laboratories of Konstanz University,

		<i>Te2</i> …N1	Te1·…N1	Te1…N2	<i>Te2</i> · · · <i>N</i> 2	E _{HF}	Е _{НF} (0 К)	ΔE_{HF}	Δ <i>Ε_{ΗF} (0 K</i>)
1	Α		3.178		3.178	-358.959209	-358.831186	0.0	0.0
	В	3.022	3.521	3.022	3.521	-358.954853	-358.826404	2.7	3.0
	С					-358.952871	-358.824927	4.0	3.9
2	D		2.991		2.991	-666.258749	-666.035436	0.0	0.0
	Е	3.233	3.288	3.288	3.287	-666.251605	-666.028030	4.5	4.6
3	F		2.997	2.996		-1128.385494	-1127.998883	0.0	0.0
	G	3.602	3.277	3.562	3.277	-1128.376383	-1127.989693	5.7	5.8
	н	3.536	3.284	3.462	3.290	-1128.367427	-1127.980484	11.3	11.5
	1	3.331	3.284	3.367	3.292	-1128.358196	-1127.971187	16.9	17.4
	J	5.030	3.325	4.324	3.325	-1128.357960	-1127.971006	17.3	17.5

TABLE 3 The Te...N Interactions, Total Energies (in Hartree) and Sum of the Electronic and Zero-Point Energies (in Hartree) of Different Conformers of the Three Computational Models

Distances are in Å and relative energies are in kcal/mol.

Germany. Mass spectra were determined on a Finnigan MAT-321 spectrometer at 70 eV, and measurements were carried out on ¹³⁰Te isotope.

Synthesis

All reactions were carried out under dry nitrogen atmosphere. All solvents were dried and freshly distilled under nitrogen before use. 4-Chloro-2-aminophenyl-tellurium(IV)tribromide, 4-bromo-2-aminophenyl tellurium(IV) tribromide [19,20], 4-methoxy-2-aminophenyltellurium(IV) tribromide [19,20], and 4-nitro-2-aminophenyltellurium(IV) tribromide [19,20], and tellurium tetrabromide [21] were prepared by following the literature methods.

Synthesis of 2-(Benzylideneamino)-5chlorophenyl Mercuric Chloride

A solution of 2-amino-5-chlorophenyl mercuric chloride (1.08 g; 3 mmol) and benzaldehyde (0.32 g; 3 mmol) in ethanol (60 cm³) containing three drops of sulfuric acid was refluxed for 1 h. After cooling, the precipitate was collected by filtration. The solid compound was washed several times with ethanol and dried in vacuo.

Compounds 2-(benzylideneamino)-5-bromophenyl mercuric chloride, 2-(benzylideneamino)-5methoxyphenyl mercuric chloride, and 2-(benzylideneamino)-5-nitrophenyl mercuric chloride were prepared by following the above general synthetic procedure using appropriate reactants. All mecurated Schiff bases gave satisfactory analyses and are not reported.

Synthesis of 2-(Benzylideneamino)-5chlorophenyltellurium Tribromide (1)

A mixture of 2-(benzylideneamino)-5-chlorophenyl mercuric chloride (0.81 g; 1.79 mmol) and tellurium tetrabromide (0.80 g; 1.79 mmol) in dry chloroform (30 cm³) was stirred and refluxed for 4 h under nitrogen atmosphere. The resulting solution was filtered hot and cooled to room temperature. The resulting precipitate was collected by filtration. Recrystallization of the solid product from ether/hexane (7/3) gave compound **1** as yellow solid in 83% yield (1.96 g), mp 144-146°C. 2-(Benzylideneamino)-5bromophenyltellurium tribromide (2), 2-(benzylideneamino)-5-methoxyphenyltellurium tribromide (3), and 2-(benzylideneamino)-5-nitrophenyltellurium tribromide (5) were prepared by following the above method.

Bis(2-(benzylideneamino)-5-chlorophenyl)tellurium Dibromide (**5**)

A mixture of 2-(benzylideneamino)-5-chlorophenyl mercuric chloride (1.62 g; 3.58 mmol) and tellurium tetrabromide (0.80 g; 1.79 mmol) in dry chloroform (40 cm³) was stirred and refluxed for 5 h under nitrogen atmosphere. The resulting solution was filtered hot and cooled to room temperature. The resulting precipitate was collected by filtration. Recrystallization of the brown solid product from ethanol/chloroform (4/1) gave compound **5** as yellowish-brown solid in 52% yield (mp 137– 139°C).

Bis(2-(benzylideneamino)-5-bromophenyl)tellurium dibromide (**6**), 2-(benzylideneamino)-5-methoxyphenyl)tellurium dibromide (**7**), and bis(2-



FIGURE 2 Optimized skeleton structures for compounds 1-4 (a), 5-8 (b), and 11-14 (c). Distances are in Å.

(benzylideneamino)-5-nitrophenyl)tellurium dibromide (8) were prepared by reacting 2 mol of the corresponding mercurated Schiff base with 1 mol of tellurium tetrabromide in dry chloroform under the same conditions for the preparation of compound 5.

Synthesis of Bis(2-(benzylideneamino)-5chlorophenyl)ditelluride (**9**)

Compound 1 (1.16 g; 2 mmol) was dissolved in 25 cm^3 of hot ethanol and heated under reflux.

An ethanolic solution of hydrazine hydrate (0.26 g; 8 mmol) was added dropwise to the refluxing solution until the evolution of nitrogen ceased. The resulting solution was cooled to room temperature, poured into 1 dm³ of water, and extracted with ether ($3 \times 100 \text{ cm}^3$). The ether extracts were dried over anhydrous calcium chloride, and evaporation of the solvent afforded an orange precipitate. Recrystallization from ethanol gave an orange solid in 64% (mp 122–124°C). Compound bis(2-(benzylideneamino)-5-nitrophenyl)ditelluride (**10**) was prepared by following the above method for the preparation of compound **9**.

Synthesis of Bis(2-(*benzylideneamino*)-5*chlorophenyl*)*telluride* (**11**)

Bis(2-(benzylideneamino)-5-chlorophenyl)tellurium dibromide (**5**) (0.72 g; 1 mmol) was dissolved in 25 cm³ of hot ethanol. Hydrazine hydrate in hot ethanol was added dropwise until evolution of nitrogen ceased. The resulting solution was poured into 500 cm³ of water, and a pale yellow precipitate was formed. The product was recrystallized from chloroform to give yellow precipitate in 64% yield (0.42 g, mp 134–136°C).

Compounds bis(2-(benzylideneamino)-5-bromophenyl)telluride (12), bis(2-(benzylideneamino)-5-methoxyphenyl)telluride (13), and bis(2-(benzylideneamino)-5-nitrophenyl)telluride (14) were prepared by the reduction of compounds **6–8** by hydrazine hydrate in boiling ethanol and as for compound 11.

ACKNOWLEDGMENTS

Authors thank Prof. N. A. Al-Masoudi (Konstanz University, Germany) for providing some of CHN analyses, NMR, and mass spectroscopic data. We are also thankful to Department of Chemistry, College of Arts and Science, Tarhona, Almergab University, Libya for providing facilities and Petroleum Research Center, Tripoli, Libya for providing some of the IR and NMR spectroscopic data.

REFERENCES

- Rolling, P. V.; Kirt, D. D.; Dill, J. L.; Hall, S.; Holtstrom C. J Organomet Chem 1976, 116, 39.
- [2] Bruce, M. I.; Goodall, B. L.; Stone, F. A. A. J Chem Soc, Chem Commun 1973, 558.
- [3] Singh, H. B.; McWhinnie, W. R. J Chem Soc, Dalton Trans 1985, 821.
- [4] Sadekov, I. D.; Maksimenko, A. A.; Maslakov, A. G.; Minkin, V. I. J Organomet Chem 1990, 391, 179.
- [5] Maksimenko, A. A.; Sadekov, I. D.; Maslakov, A. G.; Mehrotra, G. K.; Kompan, O. E.; Struchkov, Yu. T.; Lindeman, S. V.; Minkin, V. I. Metalloorg Khim 1988, 1, 1151.

- [6] Maksimenko, A. A.; Sadekov, I. D.; Maslakov, A. G.; Mehrotra, G. K.; Borodkin, G. S.; Kompan,O. E.; Struchkov, Yu. I.; Fedotov, M. A.; Minkin,V. I. Metallorg Khim 1989, 2, 298.
- [7] Sadekov, I. D.; Maksimenko, A. A.; Mehrotra, G. K; Minkin, V. I. Zh Org Khim 1987, 23, 657.
- [8] Sadekov, I. D; Minkin, V. I. ARKiVOC 2001, 125, and references therein.
- [9] Sadekov, I. D.; Nivorozhkin, V. L.; Zakharov, A. V.; Minkin, V. I. Zh Org Khim 1996, 32, 1434.
- [10] Minkin, V. I.; Sadekov, I. D.; Zakharov, A. V.; Nivorozhkin, V. L.; Kompan, O.E.; Struchkov, Yu. T. J Organomet Chem 1997, 233, 536.
- [11] Nivorozhkin, V. L.; Zakharov, A. V.; Sadekov, I. D.; Minkin, V. I. Zh Org Khim 2000, 36, 631.
- [12] Abed, Y.; Al-Salim, N. I.; Greaves, M. R.; McWhinnie, W. R.; West, A. A.; Hamor, T. A. J Chem Soc, Dalton Trans 1989, 1697.
- [13] Al-Salim, N. I; McWhinnie, W. R.; Hamor, T. A. J Chem Soc, Chem Commun 1989, 453.
- [14] Al-Salim, N. I; McWhinnie, W. R.; Hamor, T. A. J Organomet Chem 1988, 356, 159.
- [15] Sadekov, I. D.; Maksimenko, A. A.; Kompan, O. E.; Maslakov, A. G.; Struchkov, Yu. I.; Minkin, V. I. Metallorg Khim 1989, 2, 1260.
- [16] Kumar, R. P.; Singh, A. K.; Butcher, R. J.; Sharma, P.; Toscano, R. A. Eur J Inorg Chem 2004, 1107.
- [17] Panda, S.; Zade, S. S.; Singh, H. B.; Wolmershäuser, G. J Organomet Chem 2005, 690, 4142.
- [18] Ashk, K. S.; Anamika; Kumar, A.; Singh, P.; Srivastava, R. C.; Bucher, R. J.; Beckmann, J.; Duthine, A. J Organomet Chem 2006, 691, 1954.
- [19] Al-Rubaie, A. Z.; Al-Salim, N. I.; Al-Jadaan, S. A. N. J Organomet Chem 1993, 443, 67.
- [20] Al-Rubaie, A. Z.; Al-Jadaan, S. A. N. Appl Organomet Chem 1998, 12, 79.
- [21] Cocksedge, H. E. J Chem Soc 1908, 2175.
- [22] Al-Rubaie, A, Z.; Al-Derawi, A. K. Heteroatom Chem 2007, 18, 93 and references therein.
- [23] Frisch, M. J., et. al. Gaussian, Inc., GAUSSIAN03, Revision C.02, Wallingford CT, 2004.
- [24] (a) Hay, P. J.; Wadt, W. R. J Chem Phys 1985, 82, 275;
 (b) Wadt, W. R.; Hay, P. J. J Chem Phys 1985, 82, 284;
 (c) Hay, P. J; Wadt, W. R. J Chem Phys 1985, 82, 299.
- [25] Majeed, Z.; McWhinnie, W. R.; Hamor, T. A. J Organomet Chem 1997, 549, 257.
- [26] Cozzolino, A. F.; Britten, J. F.; Vargas-Baca, I. Cryst Growth Des 2006, 6, 181.
- [27] Cozzolino, A. F.; Vargas-Baca, I.; Mansour, S.; Mahmoudkhani, A. H. J Am Chem Soc 2005, 127, 3184.
- [28] Chauhan, A. K. S.; Anamika; Kumar, A.; Srivastava, A. C.; Butcher, R. J. J Organomet Chem 2005, 690, 313.
- [29] Pauling, L. The Nature of the Chemical Bond; Cornell University Press: New York, 1960.