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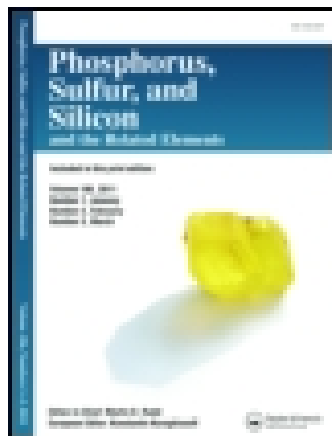
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### Potassium Tellurocyanate Mediated Coupling Reactions of N-(1-Chloroethylidene)Arylamines

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Accepted author version posted online: 11 Jun 2014. Published online: 15 Sep 2014.

To cite this article: Shaker A. Al-Jadaan, Wasfi A. Al-Masoudi, Bahjat A. Saeed & Ali Z. Al-Rubaie (2014): Potassium Tellurocyanate Mediated Coupling Reactions of N-(1-Chloroethylidene)Arylamines, Phosphorus, Sulfur, and Silicon and the Related Elements, DOI: [10.1080/10426507.2014.902830](https://doi.org/10.1080/10426507.2014.902830)

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## POTASSIUM TELLUROCYANATE MEDIATED COUPLING REACTIONS OF *N*-(1-CHLOROETHYLIDENE)ARYLAMINES

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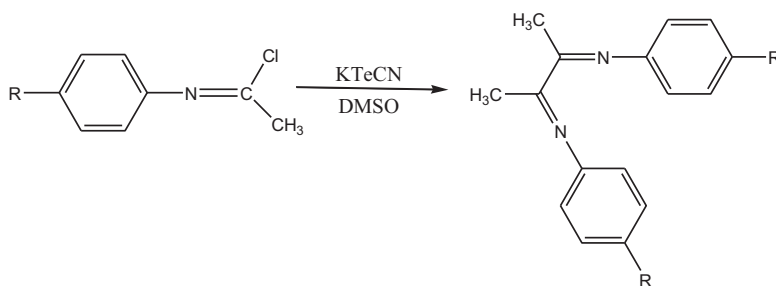
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### GRAPHICAL ABSTRACT



1: R = H; 2: R = Cl; 3: R = CH<sub>3</sub>; 4: R = NO<sub>2</sub>

**Abstract** The reaction of potassium tellurocyanate (prepared in situ) with *N*-(1-chloroethylidene)arylamines (i.e.,  $4\text{-RC}_6\text{H}_4\text{N}=\text{C}(\text{CH}_3)\text{Cl}$ , where R = H, Cl, CH<sub>3</sub> and NO<sub>2</sub>) in DMSO solution gave unexpectedly, after hydrolysis, the corresponding *N*-(3-(arylamino)butan-2-ylidene)arylamines in 63–78%. Reaction of *N*-(3-(arylamino)butan-2-ylidene)arylamines with SOCl<sub>2</sub> or with Br<sub>2</sub> resulted in the substitution of a halogen on the aromatic rings. The presence of a water molecule within the structure of the synthesized diimines was rationalized theoretically by Density Functional Theory (DFT). All compounds were characterized by elemental analysis, IR, NMR and mass spectroscopic data.

**Keywords** Potassium tellurocyanate; coupling reaction; aryl amines; thionyl chloride; bromination

Received 20 November 2013; accepted 6 March 2014.

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## INTRODUCTION

It has long been recognized that the organotellurium species are efficient and versatile reagent in organic synthesis for reactions including carbon–carbon bond formation, tellurium-metal exchange, and cross-coupling reactions.<sup>1–4</sup>

Some years ago, we demonstrated the use of potassium tellurocyanates in preparing various cyclic tellurides<sup>5–7</sup> and new phenacyl derivatives of tellurium.<sup>8</sup>

Furthermore, the reaction of potassium tellurocyanate with hydrazidoyl bromide in DMSO solution afforded 2,4-disubstituted-4-imino- $\Delta^2$ -1,3,4-telluradiazolines together with  $\alpha$ -tellurocyanatobenzaldehyde arylhydrazone.<sup>5</sup> Thus, while exploring the synthesis of new organotellurocyanate compounds, we reacted *N*-(1-chloroethylidene)arylamines with potassium tellurocyanate in hope to prepare a new series of organotellurocyanate derivatives. We unexpectedly encountered coupling reactions of *N*-(1-chloroethylidene)arylamines rather than the expected tellurocyanate formation. It is worth noting that the reaction of *N*-acylchloroformamide<sup>9</sup> or *N*-vinylchloroformamides<sup>10</sup> with potassium selenocyanate forms cyclic products.

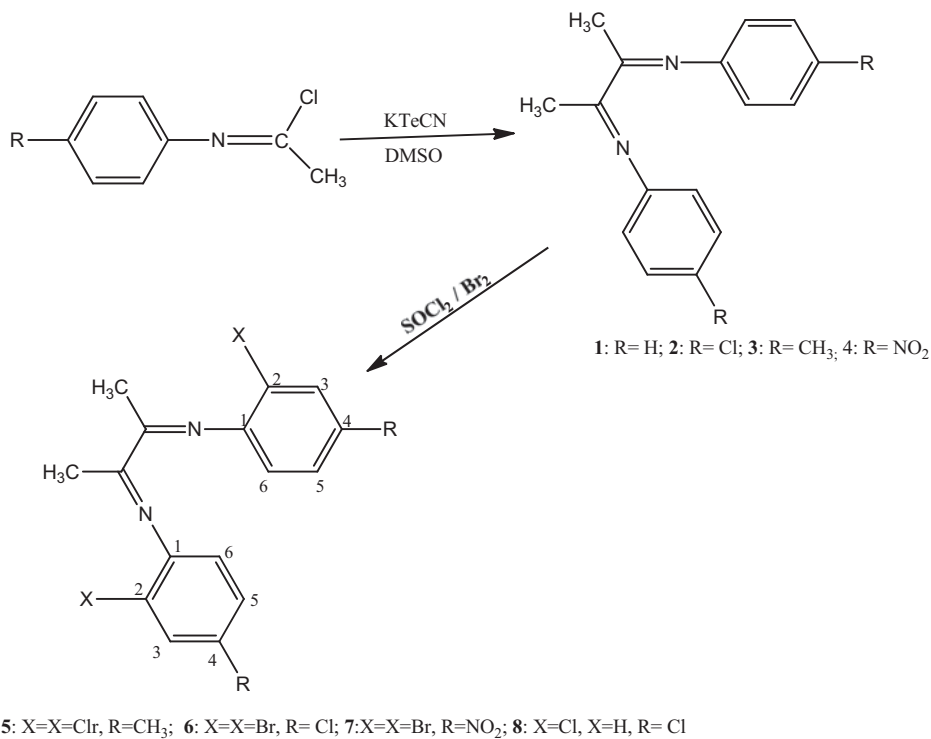
## RESULTS AND DISCUSSION

The present work was motivated by an interest in the use of potassium tellurocyanate as a tellurating agent to prepare new organotellurocyanates based on *N*-(1-chloroethylidene) substituted 4-benzenamines, from which several new organotellurium compounds should be accessible.<sup>5–8</sup> Thus, the reaction of *N*-(1-chloroethylidene)benzenamine, *N*-(1-chloroethylidene)-4-chlorobenzenamine, *N*-(1-chloroethylidene)4-methylbenzenamine, and *N*-(1-chloroethylidene)4-nitrobenzenamine with KTeCN produced *N*-(3-phenylimino)butan-2-ylidene)benzenamine (**1**), *N*-(3-(4-chlorophenylimino)butan-2-ylidene)4-chlorobenzamine (**2**), *N*-(3-(4methylphenylimino)butan-2-ylidene) 4-methylbenzenamine (**3**) and *N*-(3-(4nitrophenylimino)butan-2-ylidene)4-nitrobenzenamine (**4**) in 78%, 63%, 76%, and 73% yield, respectively, (Scheme 1). These compounds (**1–4**) gave satisfactory elemental analyses and spectroscopic data. The formation of these unexpected products could be explained as a result of the formation of organyl tellurocyanates (**A**) as intermediates (Scheme 2). The main pathway may be the disproportionation of the intermediate tellurocyanates (**A**) to the tellurides (**B**) and tellurium dicyanide (**C**), the latter of which decomposed instantly to elemental tellurium and dicyanogen (Scheme 2). Tellurides (**B**) eliminate tellurium thermally<sup>11</sup> or photochemically<sup>12</sup> to give the corresponding products (**1–4**) in 63–78% yields (Scheme 2).

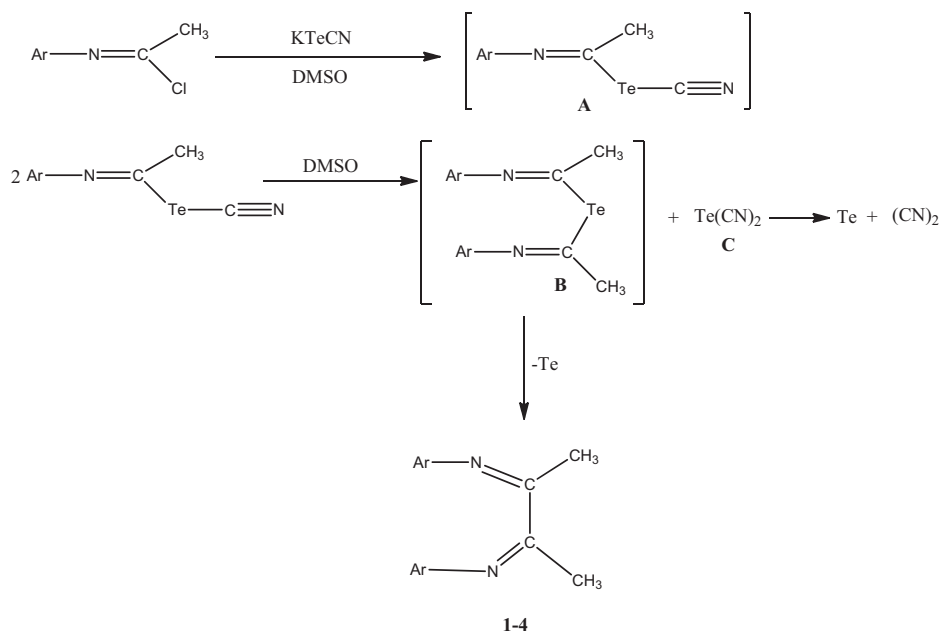
CHN analyses of compounds **1–4** confirm the presence of a water molecule within the structure of these compounds, see Experimental Part.

Treatment of **2** and **4** with excess bromine (3 equivalents) or treatment of compound **3** with three equivalents of SOCl<sub>2</sub> at room temperature resulted in the halogenation of both benzene rings and the formation of compounds **6**, **7**, and **5** in 70%, 77, and 65% yields, respectively (Scheme 1). On the other hand, treatment of compound **2** with one equivalent of SOCl<sub>2</sub> gave a mono *ortho*-chlorinated products (i.e., compound **8**) in good yield (Scheme 1).

The structures of the isolated products (i.e., compounds **5–8**) were confirmed by <sup>1</sup>H and <sup>13</sup>C spectra in addition to elemental analysis. These observations agree with the previous works<sup>13</sup> on the bromination of salicylic aldehyde benzoyl hydrazone with bromine and the chlorination of phenothiazines with thionyl chloride<sup>14</sup> at room temperature.



Scheme 1



Scheme 2

**Table 1** The calculated energies and structural properties of the diimine and the diimine–water complex calculated at two levels of theory

B3LYP/6-31+G(d)		B3LYP/6-31+G(d,p)		B3LYP/6-311+G(d,p)	
Ligand-water	Ligand	Ligand-water	ligand	Ligand-water	Ligand
−805.27523275	−728.85189006	−805.31006432	−728.87604857	−805.47174066	−729.01314089

The IR spectra of compounds **1–8** show a broad band around  $3300\text{ cm}^{-1}$  due to the presence of a water molecule and a band due to  $\text{C}=\text{N}$  that appears as medium to strong peak in the range  $1660\text{--}1677\text{ cm}^{-1}$ . For compounds **4** and **8**, the bands at  $1330$  and  $1363\text{ cm}^{-1}$  and at  $1530$  and  $1573\text{ cm}^{-1}$  were assigned to the symmetrical and asymmetrical stretching vibration of the  $\text{NO}_2$  group, respectively. All Ar-Cl and Ar-Br compounds show a weak band around  $770\text{ cm}^{-1}$  and around  $660\text{ cm}^{-1}$ , respectively, Experimental Section. Thus, in order to rationalize the presence of a water molecule within the structure of the synthesized diimines, a theoretical Density Functional Theory (DFT) study was undertaken. The free ligand, water, and the ligand–water 1:1 complex were fully optimized at several levels of theory. Several conformations for the complex were examined and the geometry of the most stable one identified. The structure was obtained from the B3LYP/6-311+G(d,p) calculations. It has  $C_1$  symmetry where the  $\text{H}_2\text{O}$  molecule bridges the nitrogen atoms of the  $-\text{CN}-\text{CN}-$ linkage in the ligand via two hydrogen bonds of  $2.256\text{ \AA}$  lengths for each. The bridge forces the ligand to adopt a twisted conformation about this linkage with a torsion angle equal to  $63.6^\circ$ . On the other hand, the corresponding angle in the ligand is  $179.1^\circ$ . The binding energy of the ligand–water complex was calculated as follows: Binding energy = Total energy of the complex – (Total energy of water + Total energy of the ligand). At the B3LYP/6-11+G(d,p) level of theory, the calculated binding energy is  $0.271\text{ kJ}\cdot\text{mol}^{-1}$ . The negative value of the binding energy suggests a stabilizing effect of the binding of a water molecule to the ligand. The fully optimized parameters of the diimine and the diimine–water complex calculated at different levels of theory are presented in Table 1 and shown in Figure 1.

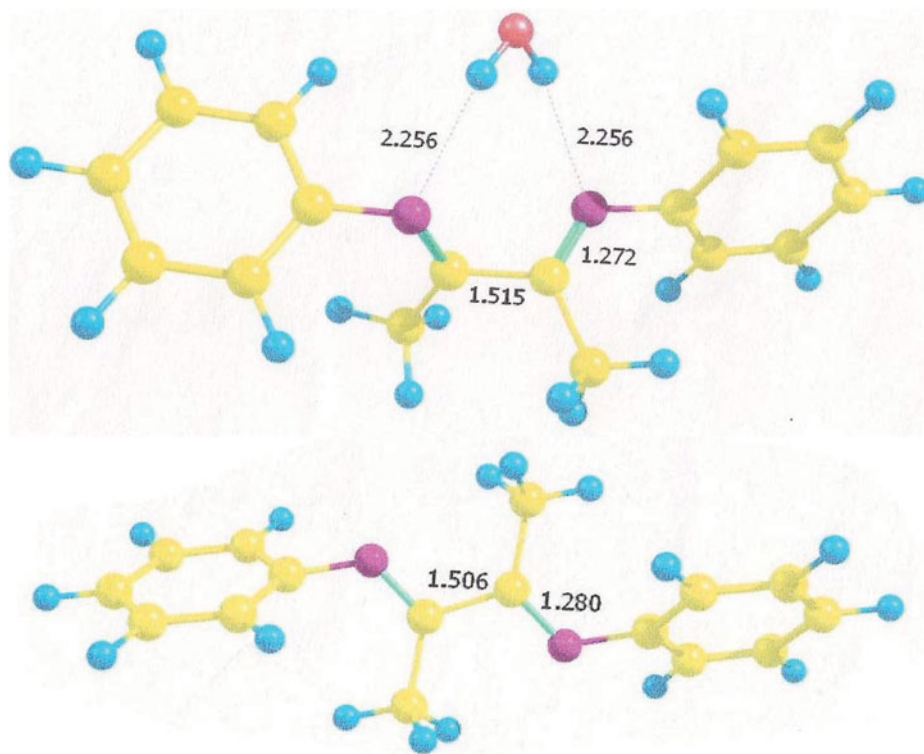
In the  $^1\text{H}$  NMR spectra of compound **1–8**, the resonances of the aromatic ring were observed in the range  $8.23\text{--}6.58\text{ ppm}$  while the  $\text{CH}_3$  groups resonate in the range  $2.05\text{--}2.51\text{ ppm}$ . The  $^1\text{H}$  NMR spectra of all compounds show all the expected proton resonances and integration values.  $^{13}\text{C}$  NMR spectra of compounds **1**, **2**, **3**, **4**, **6**, and **8** provided further support for the formation of these compounds, see Experimental Section.

In conclusion, this work shows the ability of potassium tellurocyanate to act as a coupling reagent for imidoyl chlorides, although the diimines have been well known as ligands for many years.

## EXPERIMENTAL

### Physical Measurements

$^1\text{H}$  and  $^{13}\text{C}$  NMR spectra were obtained with a Bruker 300 spectrometer instrument. They were recorded in  $\text{DMSO-d}_6$  or  $\text{CDCl}_3$  solutions containing TMS as internal standard. The chemical shifts for all  $^1\text{H}$  and  $^{13}\text{C}$  NMR spectra were reported in  $\delta$  units downfield from the internal reference  $\text{Me}_4\text{Si}$ . Elemental analyses (C, H, and N) were performed



**Figure 1** The optimized structures of the diimine and the diimine–water complex.

by Analytical Laboratories of Konstanz University, Germany. Mass spectra (EI) were determined on a Finnigan MAT 321 spectrometer at 70 eV (Konstanz University, Germany).

### Synthesis

All reactions were carried out under an argon atmosphere. All solvents were dried and freshly distilled under nitrogen before use. *N*-(1-chloroethylidene)benzenamine, *N*-(1-chloroethylidene)-4-chlorobenzenamine, *N*-(1-chloroethylidene)-4-methylbenzenamine and *N*-(1-chloroethylidene)-4-nitrobenzenamine were prepared by literature methods<sup>15</sup>.

***N*-(3-(Phenylimino)butan-2-ylidene)benzenamine (1)**. A mixture of finely ground tellurium powder (1.91 g; 15 mmol) and dry powdered potassium cyanide (0.97 g; 15 mmol) in dry DMSO (25 mL) was refluxed under an argon atmosphere with stirring until most tellurium was dissolved (~2 h). To the resulting solution, diluted with dry DMSO (20 mL) and cooled to room temperature, was added a solution of *N*-(1-chloroethylidene)benzenamine (2.30 g; 15 mmol) in dry DMSO (25 mL) dropwise over period of 30 min. The mixture was stirred for 2 h at room temperature; the pale yellow solution was filtered, poured into distilled water (500 mL) and set aside overnight. Tellurium deposited upon standing from the solution. Extraction with diethyl ether (3 × 100 mL) gave a pale-yellow solution, which was dried (MgSO<sub>4</sub>), filtered, and evaporated under reduced pressure to give a pale-yellow precipitate. Recrystallization from ether/hexane (3/2) gave a white solid product of compound **1** in 78% yield, m.p. 128–130°C. (Lit.<sup>16</sup> 130–131°C).



Anal. Calc. for  $C_{16}H_{16}N_2 \cdot H_2O$ : C, 75.57; H, 7.13; N, 11.01. Found: C, 75.47; H, 7.05; N, 10.87%. IR (KBr,  $cm^{-1}$ ): 3310b, 3075w, 2888w, 2845w, 1660s, 1615s, 1560s, 1265s, 752s.  $^1H$  NMR (DMSO- $d_6$ ,  $\delta$  / ppm): 2.21 (s, 6H,  $CH_3$ ); 7.26(t,  $J = 8.8, 1.0$  Hz, Ar-H); 7.33(t, 4H,  $J = 8.5, 1.1$  Hz, Ar-H); 7.48(d, 4H,  $J = 7.9$  Hz, Ar-H).  $^{13}C$  NMR (DMSO- $d_6$ ,  $\delta$  / ppm): 23.9( $CH_3$ ), 118.5 (C-2,6), 122.0 (C-3,5), 128.0(C-4), 139.0(C-1), 168.1(C=N).

The following compounds were prepared similarly using the appropriate *N*-(1chloroethylidene)arylamine:

***N*-(3-(4-Chlorophenylimino)butan-2-ylidene)-4-chlorobenzamine (2).**

White solid, 63% yield, m.p. 169–171°C (Lit.[16] 175°C, Lit.<sup>17</sup> 177°C). Anal. Calc. for  $C_{16}H_{14}Cl_2N_2 \cdot H_2O$ : C, 59.46; H, 4.99; N, 8.67. Found: C, 59.01; H, 4.98; N, 8.21%. IR(KBr,  $cm^{-1}$ ): 3300b, 3080w, 2950w, 2850w, 1671s, 1620m, 1553s, 1268s, 770m.  $^1H$  NMR(DMSO- $d_6$ ,  $\delta$  / ppm): 2.05(s, 6H,  $CH_3$ ); 7.29 (d, 4H,  $J = 7.6$  Hz, Ar-H); 7.66 (d, 4H,  $J = 7.6$  Hz, Ar-H).  $^{13}C$  NMR(DMSO- $d_6$ ,  $\delta$  / ppm): 23.9 ( $CH_3$ ), 120.0 (C-2,6), 126.0(C-3,5), 128.1(C-4), 138.2(C-1), 168.3(C=N). EI-MS  $m/z$  ( $M^+$ ) 304.

***N*-(3-(4-Methylphenylimino)butan-2-ylidene)-4-methylbenzenamine (3).**

White solid, 76%, m.p. 140–141°C. Anal. Calc. for  $C_{18}H_{20}N_2 \cdot H_2O$ : C, 76.56; H, 7.85; N, 9.92. Found: C, 76.28; H, 7.66; N, 10.27%. IR(KBr,  $cm^{-1}$ ): 3085w, 2900w, 2800w, 1663s, 1615m, 1555s, 1260s, 750s.  $^1H$  NMR(DMSO- $d_6$ ,  $\delta$  / ppm): 2.20(s, 6H,  $CH_3$ -Ar); 2.34(s, 6H,  $CH_3$ ); 7.12(d, 4H,  $J = 7.7$  Hz, Ar-H); 7.37(d, 4H,  $J = 7.5$  Hz, Ar-H).  $^{13}C$  NMR(DMSO- $d_6$ ,  $\delta$  / ppm): 20.4 ( $CH_3$ ), 23.83( $CH_3$ -Ar), 118.0(C-2,6), 128.1(C-3,5), 131.2(C-4), 136.0(C-1), 167.9(C=N). EI-MS  $m/z$  ( $M^+$ ) 264.

***N*-(3-(4-Nitrophenylimino)butan-2-ylidene)-4-nitrobenzenamine (4).**

White solid, 73% yield, m.p. 118–120°C. Anal. Calc. for  $C_{16}H_{14}N_4O_4 \cdot 2H_2O$ : C, 53.04; H, 5.01; N, 15.46. Found: C, 52.69; H, 4.93; N, 15.35%. IR(KBr,  $cm^{-1}$ ): 3305b, 3090w, 2900w, 2850w, 1677s, 1617m, 1562s, 1266s, 748s.  $^1H$  NMR(DMSO- $d_6$ ,  $\delta$  / ppm): 2.16 (s, 6H,  $CH_3$ ) 7.81(d, 4H,  $J = 8.8$  Hz, Ar-H); 8.21(d, 4H,  $J = 8.8$  Hz, Ar-H).  $^{13}C$  NMR(DMSO- $d_6$ ,  $\delta$  / ppm): 24.2( $CH_3$ ), 112.0(C-2,6), 125.1(C-3,5), 135.2 (C-1), 155.0(C-1), 169.0(C=N).

***N*-(3-(2-Chloro-4-methylphenylimino)butan-2-ylidene)-2-chloro-4-methylbenzenamine (5).** To a solution of *N*-(3-(4-methylphenylimino)butan-2-ylidene)-4-methylbenzenamine (3) (0.27 g; 1 mmol) in dry diethyl ether (15 mL) was added slowly with stirring a solution of thionyl chloride (0.36 g; 3 mmol) in diethyl ether (10 mL) at 5°C. The mixture was stirred further for 1 h at room temperature; the pale yellow solution then filtered. The solvent was allowed to evaporate at room temperature and a pale yellow precipitate was obtained. The product was recrystallized from chloroform and hexane (3:2) to give compound 5 as a very pale yellow solid in 67% yield; m.p. 117–119°C. Anal. Calc. for  $C_{18}H_{18}Cl_2N_2 \cdot H_2O$ : C, 61.56; H, 7.74; N, 7.97. Found: C, 61.09; H, 5.65; N, 7.63%. IR(KBr,  $cm^{-1}$ ): 3320b, 3092w, 2970w, 2845w, 1677s, 1658m, 1565s, 1270s, 765s.  $^1H$  NMR(DMSO- $d_6$ ,  $\delta$  / ppm): 2.15(s, 6H,  $CH_3$ -Ar); 2.34(s, 6H,  $CH_3$ ); 7.12 (d, 2H,  $J = 7.6$  Hz, ArH); 7.25(s, 2H, ArH); 7.36 (d, 2H,  $J = 1.2$  Hz, Ar-H).

***N*-(3-(2-Bromo-4chlorophenylimino)butan-2-ylidene)-2-bromo-4-chloro benzenamine (6).** To a solution of *N*(3(4chlorophenylimino)butan2ylidene)4 chlorobenzamine (2) (0.30 g; 1 mmol) in dry diethyl ether(20 mL) was added dropwise a solution of bromine (0.48 g; 3 mmol) in dry diethyl ether (20 mL). The yellow solution was stirred for 2 h at room temperature. Evaporation of the solvent at room temperature yielded a yellow precipitate of compound 6. Recrystallization of the product from chloroform and ether (7:3) gave a yellow solid in 70% yield.

Anal. Calc. for  $C_{16}H_{12}Br_2Cl_2N_2 \cdot H_2O$ : C, 39.85; H, 2.93; N, 5.82. Found: C, 39.64; H, 2.91; N, 5.77%. IR (KBr,  $cm^{-1}$ ): 3315b, 3076w, 2900w, 2840w, 1667s, 1620s, 1566s, 1265s, 775s, 665w  $^1H$  NMR(DMSO- $d_6$ ,  $\delta$  / ppm): 2.13(s, 6H,  $CH_3$ ); 7.02 (d, 2H,  $J$  = 7.8 Hz, Ar-H); 7.47 (d, 2H,  $J$  = 7.6 Hz, Ar-H); 8.12(s, 2H, Ar-H).  $^{13}C$  NMR(DMSO- $d_6$ ,  $\delta$  / ppm): 23.4( $CH_3$ ), 112.3(C-2), 125.4(C-6), 130.1(C-4), 131.7 (C-5), 133.4(C-3), 154.8(C-1), 168.7(C=N). EI-MS  $m/z$  ( $M^+$ ) 462.

**N-(3-(2-Bromo-4-nitrophenylimino)butan-2-ylidene)2-bromo-4-nitrobenzenamine (7)**. This compound was prepared by the same above method using *N*-(3-(4nitrophenylimino)butan-2-ylidene)-4-nitrobenzamine (**4**) as starting material. Compound **7** was obtained as a brown solid in 77% yield, m.p. 126–128°C. Anal. Calc. for  $C_{16}H_{12}Br_2N_4O_4 \cdot H_2O$ : C, 38.27; H, 2.81; N, 11.16. Found: C, 37.92; H, 2.69; N, 10.87%. IR (KBr,  $cm^{-1}$ ): 3310b, 3095w, 2950w, 2867w, 1665s, 1615m, 1564s, 1267s, 660m.

$^1H$  NMR(DMSO- $d_6$ ,  $\delta$  / ppm): 2.51(s, 6H,  $CH_3$ ); 6.62 (d, 2H,  $J$  = 7.7 Hz, Ar-H); 7.94(d, 2H,  $J$  = 7.5 Hz, Ar-H); 8.11(s, 2H, Ar-H).  $^{13}C$  NMR(DMSO- $d_6$ ,  $\delta$  / ppm): 23.6 ( $CH_3$ ), 112.0(C-2), 123.1(C-6), 126.0 (C-5), 131.0 (C-3), 135.2(C-4), 155.1(C-1), 168.4 (C=N). EI-MS  $m/z$  ( $M^+$ ) 482.

**N-(3-(2,4-Dichlorophenylimino)butan-2-ylidene)4-chlorobenzeneamine (8)**. This compounds was prepared by the same method described for the preparation of compound **5** by using equimolar ratio of *N*-(3-(4-chlorophenylimino)butan-2ylidene)-4-chlorobenzeneamine (**2**) (0.27 g; 1 mmol) and thionyl chloride (0.12 g; 1 mmol). Compound **8** was obtained as a pale yellow solid in 65% yield, m.p. 152–154°C. Anal. Calc. for  $C_{16}H_{13}Cl_3N_2 \cdot H_2O$ : C, 53.73; H, 4.23; N, 7.83. Found: C, 53.36; H, 4.38; N, 7.47%. IR(KBr,  $cm^{-1}$ ): 3320b, 3070w, 2887w, 2796w, 1670s, 1615m, 1564s, 1267s, 751, 776m.  $^1H$  NMR(DMSO- $d_6$ ,  $\delta$  / ppm): 2.03(s, 6H,  $CH_3$ ); 7.01–7.08(m, 3H, Ar-H); 7.36(d, 1H,  $J$  = 6.8 Hz, Ar-H); 7.53-7.59(m, 3H, Ar-H).  $^{13}C$  NMR(DMSO- $d_6$ ,  $\delta$  / ppm): 23.4 ( $CH_3$ ), 122.4 (C-6), 124.8 (C',6'), 128.1(C-2), 129.1(C-5), 130.2(C-4),131.6 (C-4'), 132.89C-3'), 135,8 (C3), 147,1(C-1'), 149.6 (C-1), 168.8(C=N). EI-MS  $m/z$  ( $M^+$ ) 338.

## FUNDING

The authors thank Prof. Dr N. A. Al-Masoudi, Department of Chemistry, Konstanz University, Germany for providing some of the CHN analyses, NMR and mass spectroscopic data.

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