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# Facile selective synthesis of new furo[3,4-*d*]-1,3-thiazoles

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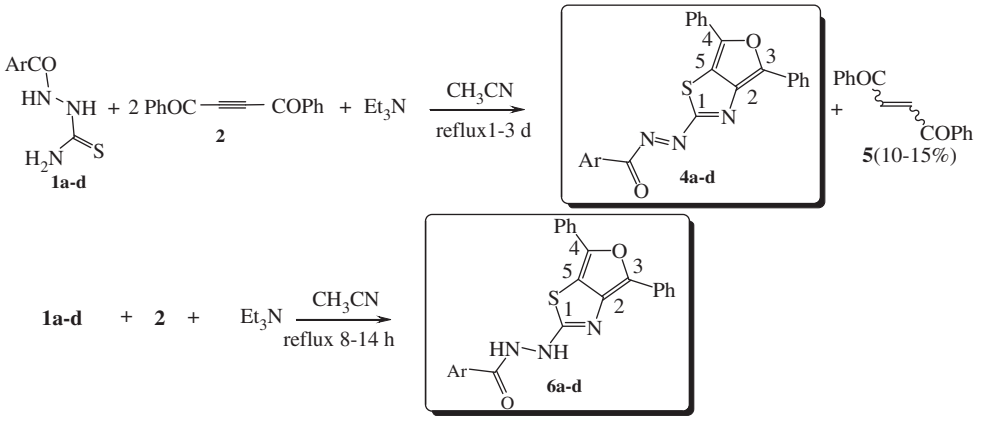
In presence of triethylamine, the reaction of substituted 2-aroyl-hydrazinecarbothioamides with 1,4-Diphenyl-but-2-yne-1,4-dione afforded novel furo[3,4-*d*]-1,3-thiazoles in good yield. The reaction mechanism suggests that the second molecule of 1,4-Diphenyl-but-2-yne-1,4-dione behaved as an oxidizing agent.




**Keywords:** *N*-aryl-hydrazinecarbothioamides; 1,4-Diphenyl-but-2-yne-1,4-dione; triethylamine; bi-nucleophilic attack; furo[3,4-*d*]-1,3-thiazoles



## 1. Introduction

The chemistry of 1,4-diphenylbut-2-yne-1,4-dione (DBD) has been extensively investigated. For example, DBD reacts with benzimidazole-2-thione to produce 2-(acylvinylthio)-benzimidazoles (*1*), while diarylazines react with DBD to produce pyridazines *via* Diels–Alder reactions (*2*). Thioamides and their derivatives occupy a distinctive place among the other *N*,*S*-containing compounds used in the synthesis of heterocyclic systems due to their accessibility

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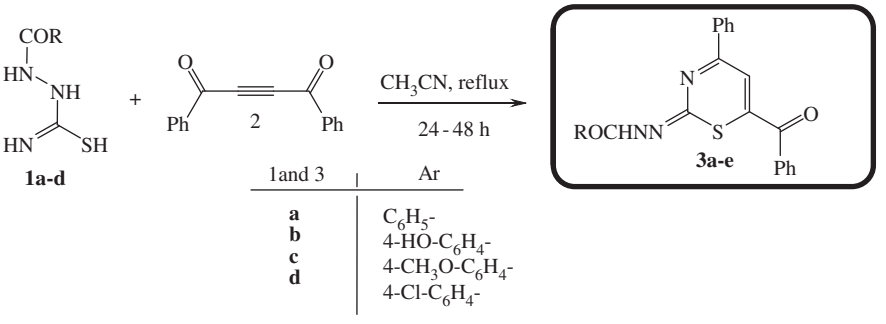



Scheme 1. 1,3-Thiazine formation from the reaction of 2-aroyl-hydrazinecarbothioamides **1a**–**1d** with DBD (**2**).

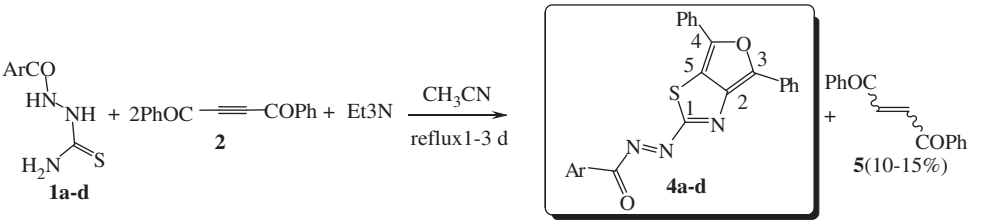


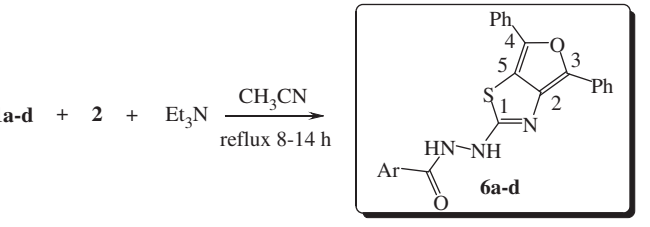
and their ability to act as difunctional nucleophiles. The heterocyclizations of 1,4-disubstituted thiosemicarbazides – in basic or acidic media and under various reaction conditions – have been investigated (*3*, *4*). In addition, four-, five-, six- and seven-membered heterocyclic compounds were prepared by the reaction of thiosemicarbazide derivatives with $\alpha$- and $\beta$-haloketones (*5*, *6*). The $N^2$ of the thiosemicarbazide group is a softer nucleophilic center than the harder and more powerful terminal nitrogen $N^1$. Some time ago, we synthesized a large number of heterocyclic ring systems such as thiazoles, imidazoles, thiazines, thiadiazoles, thiadiazines, pyrazines and indazoles from the reactions of thiosemicarbazides with $\pi$-deficient compounds (*7–10*). Aly *et al.* (*11*) reported that thiosemicarbazides show unusual reactivity toward 2,3-diphenylcyclopropenone, giving a variety of pyridazinethiones and 1,2,4-triazolo[4,3-*b*]-pyridazinethiones. The synthesis of furothiazoles is still of considerable interest in organic chemistry (for a recent review on thiazole synthesis, see (*12*)). Moreover, thiazoles are ubiquitous building blocks in medicinal chemistry and can be found in numerous natural products (*e.g.* epothilone) (*13*, *14*) and biologically important compounds including the anticancer drug dasatinib, antiviral clinical candidate TMC435350 and antidiabetic drug candidate MB06322 (*15*).

Interestingly, numerous manuscripts reporting the selective addition of the nucleophilic-amino group to $\pi$-deficient acetylenic bonds suggest that it predominantly occurs in the presence of Lewis acids and/or under weakly basic conditions (*16–18*). We have recently reported that DBD (**2**) under neutral conditions reacts with *N*-substituted-hydrazino-carbothioamides **1a**–**1d** to form the corresponding *N′*-[6-benzoyl-4-phenyl-2*H*-1, 3-thiazin-2-ylidene]-substituted-hydrazides **3a**–**3d** (Scheme 1) (*19*). In this paper, we repeated the cycloaddition between compounds **1** and **2** in the presence of triethyl amine, with the goal to investigate the selective cyclization process of the adducts.

## 2. Results and discussion

Thus, upon adding a solution of **2** in acetonitrile to an acetonitrile solution of **1a**–**1d** and triethylamine, the reaction proceeds to give furo[3,4-*d*]-1,3-thiazoles **4a**–**4d** in 70–82% yield in addition to dibenzoylethylene (**5**) in 10–15% yield (Scheme 2). We chose compounds **1a**–**1d** having aryl groups with electron donating and withdrawing substituents on the benzene ring in order to examine their effect on reactivity. The structural proof for **4a**–**4d** was based upon mass, $^1$H NMR, $^{13}$C NMR and IR spectral as well as elemental analyses. For example, mass spectrometry and elemental analysis provided the molecular formula of **4a** as $C_{24}H_{15}N_3O_2S$. The IR spectrum did not reveal any absorptions due to C=S, NH or OH groups. However, a sharp band appeared



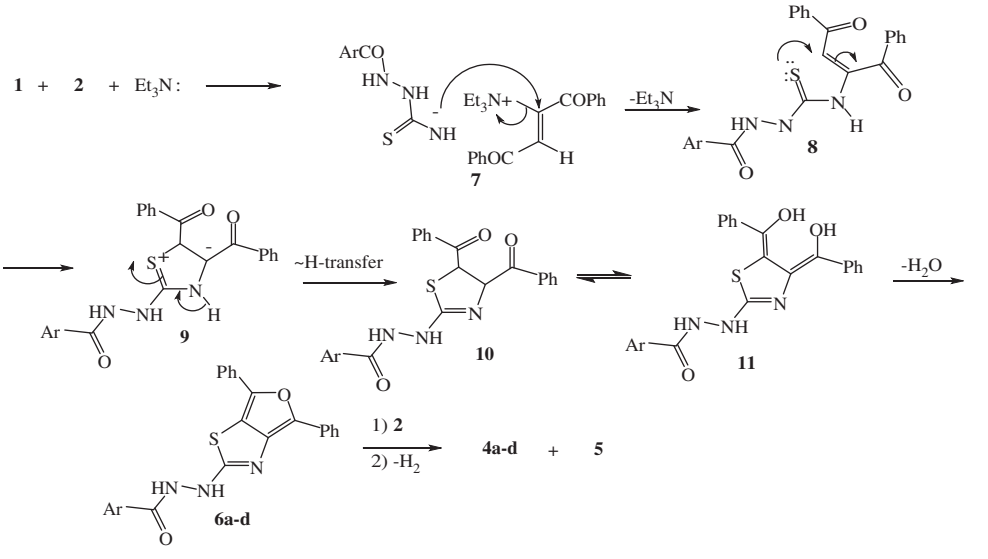

| **1, 4** and **6** | Ar | Yield of **4** (%) | Yield of **6** (%) |
|---|---|---|---|
| **a** | $C_6H_5$- | 75 | 76 |
| **b** | 4-HO-$C_6H_4$- | 77 | 78 |
| **c** | 4-$CH_3O$-$C_6H_4$- | 82 | 83 |
| **d** | 4-Cl-$C_6H_4$- | 70 | 72 |

Scheme 2. Synthesis of various furo[3,4-*d*]-1,3-thiazoles **4a**–**4d** and **6a**–**6d**.

at $\nu_{max} = 1080\,cm^{-1}$ which was assigned to C−O stretching. The $^{13}C$ NMR spectrum revealed carbon signals at $\delta_C = 190.0$, 158.8, 153.2, 153.0, 143.0 and 119.0 corresponding to the C=O, C-1, C-3, C-4, C-2 and C-5 carbons in **4a**, respectively (see Section 4).

The second derivative **4b** was identified as (*E*)-(4,6-Diphenylfuro[3,4-*d*]thiazol-2-yl)diazenyl) (4-hydroxyphenyl)methanone. The IR spectrum of **4b** revealed a broad absorption band at $\nu_{max} =$ 3450–370 $cm^{-1}$ corresponding to the hydroxyl group and another at $\nu_{max} = 1060\,cm^{-1}$ that we assign to a C−O stretch. The carbonyl groups of the dibenzoyl acetylene derivatives disappeared in the $^{13}C$ NMR spectra of the products. The phenolic OH resonated as a broad singlet in the $^1H$ NMR at $\delta_H = 8.80$. Five distinctive carbon signals for the azomethine and furan carbons appeared at $\delta_C = 154.0$ (C-1), 153.4 (C-3), 153.0 (C-4), 143.2 (C-2) and 119.0 (C-5).

One pot reaction of **1** and **2** in the presence of triethylamine is initiated by the nucleophilic attack of the nitrogen lone pair in triethylamine on the acetylenic bond leading to salt **7** as suggested by the mechanism shown in Scheme 3. The addition of few drops of triethylamine to the reaction mixtures produced very low percentage yields of the products **4a**–**4d**. Whereas the addition of triethylamine in equimolar quantity together with the starting materials produced the reaction products **4a**–**4d** by the percentage yields shown in Scheme 3. Consequently, this result supports the involvement of triethylamine in the reaction pathway as a catalyst. It is reasonable that the sulfur atom of amido group is the most nucleophilic center in the starting substrates (*11*). However, under basic conditions, the negatively charged imino group rather than the sulfur-thione is more nucleophilic in acylthiocarbohydrazides (*3*, *4*). Consequently, under our weakly basic reaction conditions, the nucleophilic-diethylamino group will predominantly occur at the $\pi$-deficient acetylenic bond in the presence of Lewis acids (*3*, *4*). This would then be followed by a proton transfer to the vinylic carbon followed by the nucleophilic attack of the negatively charged imino at the ethylenic bond that ultimately results in the elimination of the triethylamine molecule to give **8** (Scheme 3). The sulfur lone pair would then complete the cyclization process to form salt **9**, which accompanied by proton transfer from NH-3 to produce intermediate **10**. The tautomerism of **10** to form **11** followed by the elimination of water would ultimately form



Scheme 3. Plausible mechanism of formation of **4a–4d** and **6a–6d**.

compounds **6** (Scheme 3). The oxidation of compounds **6** by the action of a second molecule of **2** would finally afford compounds **4** and **5** (Scheme 3).

The oxidation of **6** by **2** is supported by the observation that upon reacting equal equivalents of **1a–1d**, **2** and triethylamine, the corresponding 4,6-Diphenylfuro[3,4-*d*]thiazol-2-yl) arylhydrazides **6a–6d** were obtained (Scheme 2). The elemental analyses and the spectral data of compounds **6a–6d** proved that they are the reduced form of compounds **6a–6d** (see Section 4). For example, the mass spectra and elemental analysis of **6a** provide its molecular formula as $C_{24}H_{17}N_3O_2S$, whereas the molecular formula for **4a** is $C_{24}H_{15}N_3O_2S$. The IR spectrum of **6a** revealed a broad absorption band at $\nu_{max} = 3350–3260\,cm^{-1}$ for the NH−NH stretch (see Section 4). The NH−NH protons of **6a** were observed as two broad singlets in the $^1H$ NMR spectrum at $\delta_H = 10.50$ and 6.00. The carbon signals of C=O, C-1, C-3, C-4, C-2 and C-5 appeared at $\delta_C = 171.7$, 160.2, 152.8, 152.0, 138.7 and 108.0, respectively.

## 3. Conclusions

Substituted 2-aroyl-hydrazinecarbothioamides can change and can react with acetylenic $\pi$-deficient compounds under slightly basic condition. Moreover and owing to the presence of more than two nucleophilic sites in the starting material, various heterocycles can be obtained.

## 4. Experimental section

### 4.1. *General procedures*

Melting points are uncorrected. $^1H$- and $^{13}C$-NMR spectra were recorded in chloroform-$d_3$ and measured on a Bruker AM 400 (400.134 MHz and 100.60 MHz) instrument. The chemical shifts ($\delta$'s) were measured versus the internal standard TMS. Elemental analyses were performed by the Microanalysis Center of the Institute für Anorganische Chemie, Technische

Universität Braunschweig, Germany. Mass spectra were obtained on a Finnigan MAT 8430 spectrometer at 70 eV. The IR spectra were obtained on a Nicolet 320 FT-IR using KBr pellets. *N*-substituted-hydrazinocarbothioamides **1a**–**1d** were prepared according to the literature (*20*).

### 4.2. *General procedure for preparation of compounds 3*

To a 500 ml two-necked round bottom flask containing a solution of **1a**–**1d** (2 mmol) in acetonitrile (200 ml) and triethylamine (0.202 g, 2 mmol), a solution of **2** (0.688 g, 4 mmol) in acetonitrile (50 ml) was added dropwise with stirring for 30 min at room temperature. The mixture was refluxed for 1–3 days (the reaction was monitored by TLC). The solvent was evaporated under vacuum and the solid residue was dissolved in dry acetone (50 ml) and the solution was chromatographed on thin layer plates (silica gel) using chloroform. The mobile phases containing products **4a**–**4d** as the slowest migrating zones, whereas compound **5** was separated as the fastest migrating zone. Compound **5** was identified by means of an authentic sample. All zones were extracted and the obtained products were recrystallized from the stated solvents.

### 4.3. *(E)-((4,6-Diphenylfuro[3,4-d]thiazol-2-yl)diazenyl)(phenyl)methanone (4a)*

Orange crystals (ethanol), mp 222 °C, yield: 0.64 g (75%). IR ($\nu_{max}$, $cm^{-1}$): 3010–3000 (Ar−CH), 1690 (C=O), 1080 (C−O). $^1$H NMR (400 MHz, $CDCl_3$): $\delta_H$ 8.30 (d, 2H, $J$=8.0 Hz, Ar−H), 8.00 (d, 2H, $J$=7.8 Hz, Ar−H), 7.82 (d, 2H, $J$=1.0 Hz, Ar−H), 7.70–7.50 (m, 4H, Ar−H), 7.40 (t, 2H, $J$=8.0, 1 Hz, Ar−H), 7.38–7.20 (m, 3H, Ar−H). $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta_C$ 190.0 (C=O), 158.8 (C-1), 153.2 (C-3), 153.0 (C-4), 143.0 (C-2), 135.3 (Ar−C), 129.90 (*o*-Ar−2CH), 129.0 (*m*-Ar−2CH), 128.2 (Ar−C), 128.0 (*o*-Ar−2CH), 127.2 (*m*-Ar−4CH), 127.0 (*p*-Ar−2CH), 126.3 (Ar−C), 126.0 (*o*-Ar−2CH), 125.8 (*p*-Ar−CH), 119.0 (C-5); $m/z$ (70 eV, %): 409 [$M^+$] (100), 366 (20), 332 (30), 324 (50), 304 (24), 276 (24), 76 (54); $C_{24}H_{15}N_3O_2S$ (409.46): Calcd: C, 70.40; H, 3.69; N, 10.26; S, 7.83. Found: C, 70.20; H, 3.60; N, 10.15; S, 7.75%.

### 4.4. *(E)-((4,6-Diphenylfuro[3,4-d]thiazol-2-yl)diazenyl)(4-hydroxyphenyl)methanone (4b)*

Orange crystals (methanol), mp 250 °C, yield: 0.66 g (77%). IR ($\nu_{max}$, $cm^{-1}$): 3450–3370 (OH), 3012–3008 (Ar−CH), 1692 (C=O), 1060 (C−O). $^1$H NMR (400 MHz, $CDCl_3$): $\delta_H$ 8.80 (s, 1H, OH), 8.20 (d, 2H, $J$=8.0 Hz, Ar−H), 7.96 (d, 2H, $J$=7.8 Hz, Ar−H), 7.80 (d, 2H, $J$=1.0 Hz, Ar−H), 7.75–7.62 (m, 3H, Ar−H), 7.40–7.22 (m, 5H, Ar−H). $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta_C$ 188.9 (CO), 162.0 (Ar−C−OH), 160.0 (C-1), 153.4 (C-3), 153.0 (C-4), 143.2 (C-2), 127.9 (Ar−C), 131.6 (*o*-Ar−2CH), 130.8 (Ar−2C), 129.4 (Ar−C), 129.2 (*m*-Ar−2CH), 128.6 (*m*-Ar−2CH), 128.2 (*o*-Ar−2CH), 126.7 (*p*-Ar−CH), 125.0 (*o*-Ar−2CH), 122.4 (*m*-Ar−2CH), 119.0 (C-5); $m/z$ (70 eV, %): 425 [$M^+$] (100), 408 (28), 350 (22), 332 (18), 304 (40), 276 (16), 272 (16), 258 (34), 216 (24), 92 (30), 76 (54); $C_{24}H_{15}N_3O_3S$ (425.46): Calcd: C, 67.75; H, 3.55; N, 9.88; S, 7.54. Found: C, 67.65; H, 3.50; N, 9.74; S, 7.50%.

### 4.5. *(E)-((4,6-Diphenylfuro[3,4-d]thiazol-2-yl)diazenyl)(4-methoxyphenyl)methanone (4c)*

Pale orange crystals (methanol), mp 198 °C, yield: 0.72 g (82%). IR ($\nu_{max}$, $cm^{-1}$): 3020–3001 (Ar−CH), 1694 (C=O), 1044 (C−O).$^1$H NMR (400 MHz, $CDCl_3$): $\delta_H$ 8.20 (d, 2H, Ar−H), 3.85 (s, 3H, $CH_3$), 8.00 (d, 2H, Ar−H), 7.74 (d, 2H, Ar−H), 7.45 (t, 2H, Ar−H), 7.40 (t, 2H, $J$=7.8, 1.0 Hz, Ar−H), 7.36 (m, 1H, Ar−H), 7.30–7.10 (m, 3H, Ar−H); $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta_C$ 188.8 (CO), 164.2 (C−O), 160.4 (C-1), 153.4 (C-4), 152.8 (C-3), 140.8 (C-2), 131.0 (*o*-Ar−2CH), 130.0 (Ar−C), 129.6 (Ar−C), 129.2 (*m*-Ar−4CH), 128.8 (*p*-Ar−CH), 128.6 (*o*-Ar−4CH), 127.6

(Ar−C), 125.7 (*p*-Ar−CH), 116.0 (*m*-Ar−2CH), 119.0 (C-4), 54.6 ($CH_3$−O); *m/z* (70 eV, %): 439 [$M^+$] (100), 408 (18), 376 (24), 362 (16), 346 (20), 332 (24), 304 (32), 296 (16), 270 (24), 220 (26), 128 (32), 274 (26), 106 (36), 92 (26), 76 (34); $C_{25}H_{17}N_3O_3S$ (439.49): Calcd: C, 68.32; H, 3.90; N, 9.56; S, 7.30. Found: C, 68.20; H, 3.90; N, 9.62; S, 7.24%.

### 4.6. *(E)-(4-Chlorophenyl) ((4,6-Diphenylfuro[3,4-d]thiazol-2-yl)diazenyl)methanone (4d)*

Pale orange crystals (ethanol), mp 210 °C, yield: 0.62 g (70%). IR ($\nu_{max}$, $cm^{-1}$): 3060–3008 (Ar−CH), 1692 (C=O), 1035 (C−O). $^1$H NMR (400 MHz, $CDCl_3$): $\delta_H$ 8.12–8.08 (m, 2H, Ar−H), 7.80–7.64 (m, 4H, Ar−H), 7.50 (t, 2H, *J*=7.8, 1 Hz, Ar−H), 7.42 (t, 2H, *J*=7.8, 1.0 Hz, Ar−H), 7.34–7.26 (m, 2H, Ar−H), 7.10–7.08 (m, 2H, Ar−H); $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta_C$ 188.0 (CO), 160.4 (C-1), 154.0 (C-4), 152.8 (C-3), 142.4 (C-2), 139.6 (Ar−C−Cl), 133.2 (Ar−2C), 131.0 (*o*-Ar−4CH), 130.3 (*m*-Ar−2CH), 129.2 (*m*-Ar−4CH), 128.6 (Ar−C), 127.2 (*o*-Ar−2CH), 126.4 (*p*-Ar−CH), 125.6 (*p*-Ar−CH), 118.7 (C-5); *m/z* (70 eV, %): 445 [M+2] (12), 444 [M+1] (38), 443 [$M^+$] (100), 410 (18), 408 (22), 382 (16), 380 (24), 368 (24), 352 (16), 350 (26), 290 (12), 284 (18), 282 (18), 242 (18), 240 (22), 112 (32), 76 (46); $C_{24}H_{14}ClN_3O_2S$ (443.90): Calcd: C, 64.94; H, 3.18; Cl, 7.99; N, 9.47; S, 7.22. Found: C, 64.80; H, 3.08; Cl, 8.02; S, 7.12%.

### 4.7. *General procedure*

By applying the same procedure mentioned before, to a 500 ml two-necked round bottom flask containing a solution of **1a**–**1d** (2 mmol) in acetonitrile (200 ml) and triethylamine (0.202 g, 2 mmol), a solution of **2** (0.344 g, 2 mmol) in acetonitrile (30 mL) was added dropwise with stirring for 30 min at room temperature. The mixture was refluxed for 8–14 h (the reaction was monitored by TLC). The solvent was evaporated under vacuum and the solid residues were treated with absolute ethanol (200 ml); compounds **6a**–**6d** were separated and recrystallized from the stated solvents.

### 4.8. *N-(4,6-Diphenylfuro[3,4-d]thiazol-2-yl)benzohydrazide (6a)*

Yellow crystals (methanol), mp 322 °C, yield: 0.63 g (76%). IR ($\nu_{max}$, $cm^{-1}$): 3350–3260 (NH−NH), 3040–3012 (Ar−CH), 1690 (C=O), 1040 (C−O); $^1$H NMR (400 MHz, $CDCl_3$): $\delta_H$ 10.50 (s, 1H, CONH), 8.20 (d, 2H, *J*=7.8 Hz, Ar−H), 8.10–8.04 (m, 4H, Ar−H), 7.60–7.65 (m, 2H, Ar−H), 7.60 (t, 2H, *J*=7.8, 1.0 Hz, Ar−H), 7.50–7.36 (m, 5H, Ar−H), 6.00 (s, 1H, N−H).$^{13}$C NMR (100 MHz, $CDCl_3$): $\delta_C$ 171.1 (CO), 160.2 (C-1), 152.8 (C-3), 152.0 (C-4), 138.7 (C-2), 130.0 (Ar−2C), 128.6 (Ar−C), 128.4 (*o*-Ar−2CH), 127.6 (*o*-Ar−4CH), 126.8 (*m*-Ar−2CH), 127.0 (*m*-Ar−4CH), 126.2 (*p*-Ar−2CH), 125.8 (*p*-Ar−CH), 108.0 (C-5); *m/z* (70 eV, %): 411 [$M^+$] (100), 338 (28), 326 (30), 306 (22), 278 (20), 248 (24), 242 (12), 78 (42); $C_{24}H_{17}N_3O_2S$ (411.48): Calcd: C, 70.05; H, 4.16; N, 10.21; S, 7.79. Found: C, 70.12; H, 4.08; N, 10.11; S, 7.65%.

### 4.9. *N-(4,6-Diphenylfuro[3,4-d]thiazol-2-yl)-4-hydroxybenzohydrazide (6b)*

Yellow crystals (methanol), mp 340 °C (decomp.), yield: 0.67 g (78%). IR ($\nu_{max}$, $cm^{-1}$): 3450–3230 (OH, NH−NH), 3090–3010 (Ar−CH), 1686 (C=O), 1040 (C−O); $^1$H NMR (400 MHz, $CDCl_3$): $\delta_H$ 10.50 (s, 1H, CONH), 9.40 (s, 1H, OH), 8.20–8.10 (m, 4H, Ar−H), 7.84 (d, 2H, *J*=8.0, 1.0 Hz, Ar−H), 7.48 (d, 2H, *J*=7.8, 1.0 Hz, Ar−H), 7.42–7.34 (m, 4H, Ar−H), 6.72–6.65 (m, 2H, Ar−H), 6.10 (s, 1H, N−H); $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta_C$ 164.8 (CO), 162.4 (C−OH), 160.0 (C-1), 152.6 (C-3), 151.8 (C-4), 138.7 (C-2), 130.8 (Ar−C), 130.3 (Ar−C), 127.8

(*o*-Ar−2CH), 127.6 (*o*-Ar−2CH), 127.2 (*o*−Ar−2CH), 126.8 (*m*-Ar−2CH), 126.4 (*m*-Ar−2CH), 125.8 (*p*-Ar−CH), 125.6 (Ar−C), 125.3 (*p*-Ar−CH), 116.0 (*m*-Ar−2CH), 107.8 (C-5); *m/z* (70 eV, %): 427 [$M^+$] (100), 410 (24), 334 (38), 306 (32), 274 (16), 260 (20), 218 (24), 200 (18), 93 (32), 76 (48); $C_{24}H_{17}N_3O_3S$ (427.48): Calcd: C, 67.43; H, 4.01; N, 9.83; S, 7.50. Found: C, 67.30; H, 3.98; N, 9.85; S, 7.40%.

### 4.10. *N-(4,6-Diphenylfuro[3,4-d]thiazol-2-yl)-4-methoxybenzohydrazide (6c)*

Yellow crystals (methanol), mp 316 °C, yield: 0.73 g (83%). IR ($\nu_{max}$, $cm^{-1}$): 3340–3260 (NH−NH), 3060–3012 (Ar−CH), 1688 (C=O), 1030 (C−O); $^1$H NMR (400 MHz, $CDCl_3$): $\delta_H$ 10.40 (s, 1H, CONH), 8.18–8.12 (m, 4H, Ar−H), 7.90 (d, 2H, *J*=7.8 Hz, Ar−H), 7.50 (t, 2H, *J*=7.7, 0.8 = Hz, Ar−H), 7.45 (t, 2H, *J*=7.7, 0.8 Hz, Ar−H), 7.38–7.32 (m, 2H, Ar−H), 7.17 (d, 2H, *J*=8.0, 1.0 Hz, Ar−H), 6.01 (s, 1H, N−H), 3.92 (s, 3H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta_C$ 165.2, 163.0 (Ar−O−C), 160.2 (C-1), 152.7 (C-3), 151.7 (C-4), 138.6 (C-2), 130.3 (Ar−C), 129.0 (Ar−C), 128.5 (*o*-Ar−4CH), 128.2 (*m*-Ar−2CH), 127.8 (*m*-Ar−2CH), 126.2 (*p*-Ar−CH), 125.2 (*o*-Ar−2CH), 124.7 (Ar−C), 124.4 (*p*-Ar−CH), 114.4 (*m*-Ar−2CH), 108.0 (C-5), 55.4 ($CH_3$−O); *m/z* (70 eV, %): 441 [$M^+$] (100), 426 (28), 410 (22), 384 (34), 364 (30), 348 (18), 272 (26), 106 (34), 92 (24), 76 (54); $C_{25}H_{19}N_3O_3S$ (441.50): Calcd: C, 68.01; H, 4.34; N, 9.52; S, 7.26. Found: C, 68.10; H, 4.30; N, 9.43; S, 7.20%.

### 4.11. *4-Chloro-N-((4,6-Diphenylfuro[3,4-d]thiazol-2-yl)diazenyl)benzohydrazide (6d)*

Yellow crystals (ethanol), mp 308 °C, yield: 0.64 g (72%); IR ($\nu_{max}$, $cm^{-1}$): 3330–3250 (NH−NH), 3050–3010 (Ar−CH), 1690 (C=O); $^1$H NMR (400 MHz, $CDCl_3$): $\delta_H$ 10.38 (s, 1H, CONH), 8.22 (d, 2H, Ar−H), 8.10 (d, 2H, Ar−H), 7.80–7.68 (m, 4H, Ar−H), 7.48 (t, 2H, *J*=8.0, 1.0 Hz, Ar−H), 7.42 (t, 2H, *J*=8.0, 1.0 Hz, Ar−H), 7.35–7.32 (m, 2H, Ar−H), 6.00 (s, 1H, N−H); $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta_C$ 164.8 (CO), 160.4 (C-1), 152.6 (C-3), 151.4 (C-4), 138.8 (C-2), 136.6 (Ar−C−Cl), 130.1 (Ar−C), 129.8 (Ar−C), 128.6 (*o*-Ar−2CH), 128.4 (Ar−C), 128.0 (*m*-Ar−2CH), 127.4 (*o*-Ar−2CH), 127.0 (*m*-Ar−4CH), 126.3 (*p*-Ar−CH), 125.8 (*o*-Ar−2CH), 124.8 (*p*-Ar−CH), 106.8 (C-5); *m/z* (70 eV, %): 447 [M+2] (15), 446 [M+1] (36), 445 [$M^+$] (100), 412 (22), 410 (24), 384 (18), 382 (20), 370 (26), 354 (18), 352 (24), 290 (18), 286 (16), 284 (24), 244 (20), 242 (22), 112 (34), 76 (50); $C_{24}H_{16}ClN_3O_2S$ (445.92): Calcd: C, 64.64; H, 3.62; Cl, 7.95; N, 9.42; S, 7.19. Found: C, 64.50; H, 3.60; Cl, 8.04; N, 9.54; S, 7.04%.



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