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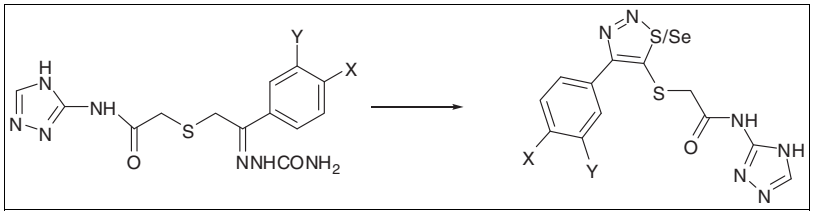

The synthesis of compounds having triazole and selena/thiadiazole rings connected by a chain having sulfur and nitrogen in the link has been described. The resultant *N*-heteroaryl-2-(heteroarylthio)acetamide may be biologically important as related compounds found application in the inhibition of HIV 1 replications.

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INTRODUCTION

The therapeutic importance of 1,2,4-triazoles are well documented. Among these, there are simple molecules having 1,2,4-triazole ring, besides biheterocyclic and polyheterocyclic compounds that contain triazole ring or triazole fused compounds [1–9]. For instance, some biheterocyclic compounds, consisting of 1,2,4-triazole and 1,3,4-thiadiazole rings, have impressive antimicrobial activity [10]. Compounds with a 1,2,3-thiadiazole moiety have been found to exhibit various pharmacological properties such as antifungal, pesticidal, and antibacterial activities [11,12]. Dong *et al*. have synthesized a series of new acrylamide derivatives containing 1,2,3-thiadiazole and evaluated their anti-hepatitis B virus activities *in vitro* [13]. Compounds containing 1,2,3-selenadiazole moieties are well known for their antimicrobial activities [14,15]. Realizing the biological importance of 1,2,4-triazole and the thia/selenadiazole rings, it has been planned to synthesize new compounds containing these skeletons connected by a chain containing a hetero atom, which are expected to have enhanced biological activity as evidenced by related studies [16–18].

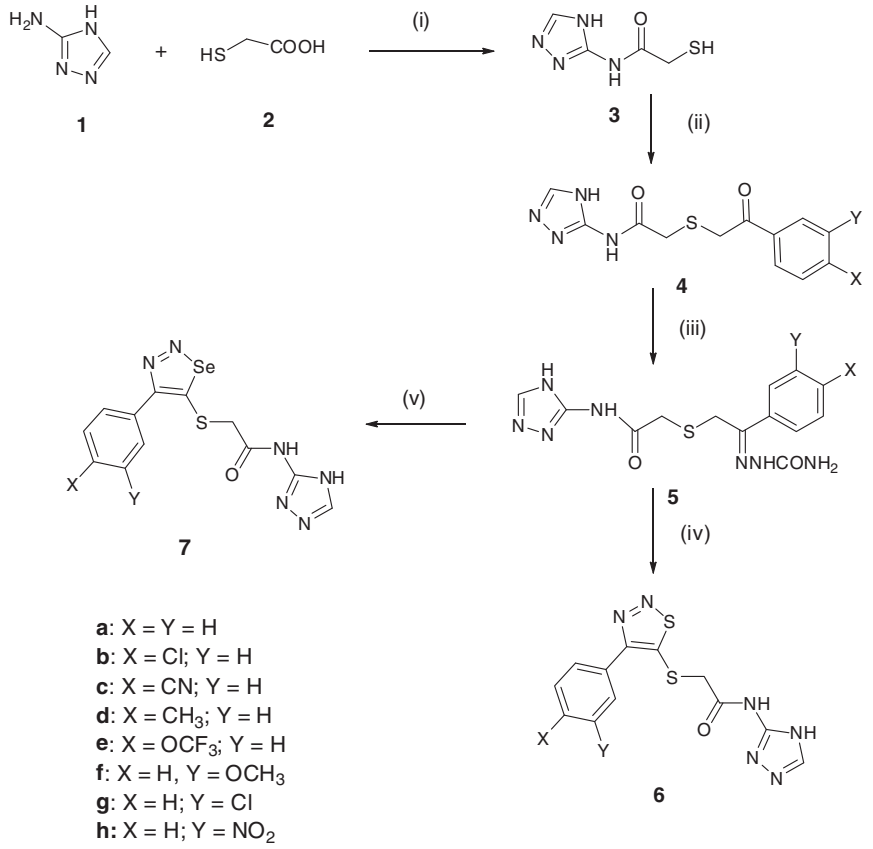
RESULTS AND DISCUSSION

The following scheme has been proposed to generate the targeted linked heterocycles (Scheme 1). The commercially available 3-amino-1,2,4-triazole (**1**) was allowed to react with thioglycolic acid (**2**) under neat condition at room temperature. The amide (**3**) formation occurred quite efficiently. The amide formed was then allowed to react with substituted phenacyl bromides in presence of potassium carbonate to provide a host of carbonyl compounds **4a–4h**. All these ketones are new and fully characterized. These ketones were then converted to their semicarbazones **5a–5h** by the conventional procedure. Although there is a chance for the formation of a mixture of geometrical isomers during this reaction, only one geometrical isomer was isolated in all the cases with no trace of the other isomer suggesting the reaction to be a stereoselective one. There is only one set of hydrogens and carbons in the ^{1}H and ^{13}C NMR spectra of **5f**.

It has been well established that the semicarbazones with active methylene at the α-carbon are capable undergoing ring closure when treated with thionyl chloride [19]. Such a ring closure has been effected on the semicabazones **5** and the reaction proceeded smoothly yielding the respective 1,2,3-thiadiazole **6a–6g** in good yield. It must be admitted the reaction has not gone at all even in poor yield when the *para*-cyano substituted and *meta*-nitro substituted aryl compounds were subjected to this reaction (**5c–6c** and **5h–6h**). This is surprising as the whole of the starting material was recovered unreacted under the reaction conditions employed in the former case, and the mass obtained could not be purified in the latter case. Thus, **6c** and **6h** were not formed.

A related oxidative cyclization process leads to the formation of 1,2,3-selenadiazole from semicarbazone by selenium dioxide. This can be effected in different solvents, and the solvent optimized in the present investigation is ethanol. A range of selenadiazoles **7a–7h** has been attempted by this protocol (Scheme 1). But **7e** and **7h** were not obtained in pure form. It seems that the conversion of the semicarbazone to selenadiazole is relatively difficult compared with thiadiazole as revealed by the yields.

Scheme 1 Reagents and conditions (i) Neat heating (175°C), 15 min; (ii) $ArCOCH_2Br$, EtOH, K_2CO_3, RT, stirring, 5 h; (iii) $NH_2NHCONH_2.HCl$, Anhyd. CH_3COOK, (Methanol : Water (v/v 1:1)), reflux, 8 h; (iv) $SOCl_2$, −15°C to RT, stirring, 12 h; and (v) SeO_2, EtOH, reflux, 12 h.



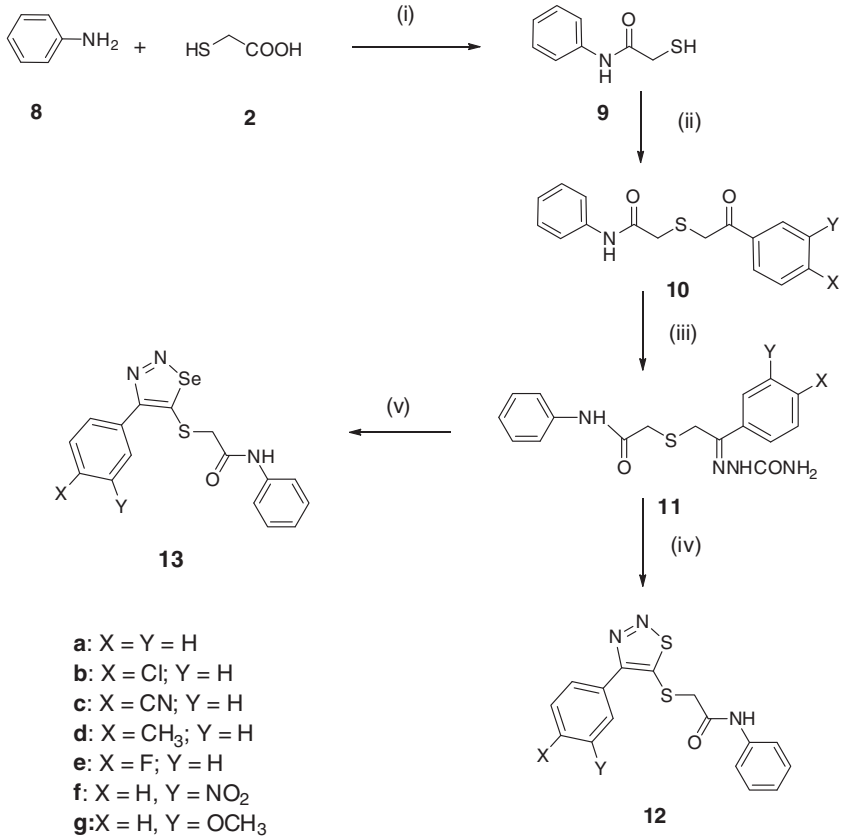
After synthesizing selenadiazoles and thiadiazoles linked to triazole by –S-CH_2-CONH- group, it has been realized that the selena/thiadiazoles connected to –S-CH_2-CONHAr, where Ar is simple aryl, have not been reported in literature. Hence, it has been planned to synthesize such compounds starting from aniline. Aniline reacts with thioglycolic acid to give the amide **9**. The free thiol present in **9** was then functionalized with substituted phenacyl bromides to give a collection of **10**. The keto compounds **10** were then converted to their semicarbazones **11** by the usual procedure. As described earlier, these semicarbazones have been subjected to (i) thinoyl chloride treatment to yield thiadiazoles **12**, and (ii) selenium dioxide treatment to give selenadiazole derivatives **13** (Scheme 2). **11g** could not be obtained in pure form. It must be noted that for the oxidative cyclization of **11** to yield **13** by selenium dioxide, the preferred solvent is THF, whereas for the selenadiazoles **7**, ethanol was preferred. **12c** and **12f** were not formed. The reaction is not at all taking place in −78°C in these cases and at higher temperature, around −10°C, the semicarbazone got hydrolyzed to give back the parent ketone.

At this juncture, it is appropriate to indicate the importance of *N*-heteroaryl-2-(heteroarylthio)acetamide synthesized in the present work. Although this work was being carried out in our laboratory, the same class of compounds has been synthesized and reported, of course by a different route, by Liu and co-workers [16–18]. It has been realized that these compounds of the type X-S-CH_2-CONHAr (where X is a five-membered heterocycle like triazole or tetrazole or thia/selenadiazole), thia acetanilide derivatives, belong to non-nucleoside reverse transcriptase inhibitors group. These are the key targets for inhibition of HIV 1 replications.

EXPERIMENTAL

All chemicals used in this investigation were of reagent grade quality and used without further purification. All melting points were recorded in open capillaries and are uncorrected. The ^1H and ^{13}C NMR spectra were recorded on a Bruker 400 MHz (Bruker, Fallanden, Switzerland) spectrometer at 400 MHz and 100 MHz, respectively in $CDCl_3$/DMSO-d_6 using TMS as internal standard. The chemical shifts are presented in δ-scale. Microanalyses were carried out on a PerkinElmer instrument (PerkinElmer, Lu Vento, The Netherlands).

Scheme 2 Reagents and conditions (i) Neat heating (130°C), 3 h; (ii) $ArCOCH_2Br$, EtOH, K_2CO_3, RT, stirring, 6 h; (iii) $NH_2NHCONH_2.HCl$, Anhyd. CH_3COOK, (Methanol : Water (v/v 1:1)), reflux, 8 h; (iv) $SOCl_2$, −78°C stirring, 4 h ; and (v) SeO_2, THF, reflux, 2 h.



All chromatographic separations were performed on 60–120 mesh silica gel using petroleum ether-ethyl acetate as eluent, unless mentioned otherwise.

***2-Mercapto-N-*(4*H*-1,2,4-triazol-3-yl)acetamide (3).** 4*H*-3-Amino-1,2,4-triazole (**1**) was treated with thioglycolic acid (**2**) in equimolar amount, and the reaction mass was heated at 175°C for 15 min. The resultant mass was worked out to give **3**, which was purified by crystallization. 85% yield as a white solid; ^{1}H NMR (400 MHz, DMSO-d_6) 3.04 (s, 2H), 7.71 (s, 1H), 10.42 (bs, 1H), 11.64 (bs, 1H), 13.44 (bs, 1H).

2-((2-Oxo-2-arylethyl)thio)-N-(4H-1,2,4-triazol-3-yl) acetamide, (4). The thiol **3** was treated with substituted phenacyl bromides and potassium carbonate in equimolar amount taken in ethanol. The mixture was left at room temperature for 5 h, and the resultant mass was worked out to give **4**, which was purified by crystallization. Compounds **4a–h** were prepared by this method.

2-((2-Oxo-2-phenylethyl)thio)-N-(4H-1,2,4-triazol-3-yl)acetamide (4a). White solid; yield: 87%; mp = 220°C; IR (KBr) 3254, 1695, 1681, 1609 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6) 3.44 (s, 2H), 4.25 (s, 2H), 7.54 (t, J = 7.2 Hz, 2H), 7.66 (t, J = 7.2 Hz, 1H), 7.71 (s, 1H), 7.98 (d, J = 7.2 Hz, 2H), 11.67 (bs, 1H), 13.40 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 34.5, 38.5, 128.5, 129.4, 133.9, 135.7, 148.5, 149.1, 168.4, 195.0; ESI-mass (m/z) calcd $[C_{12}H_{12}N_4O_2S]^+$ 276.0, found 276.6.

2-((2-(4-Chlorophenyl)-2-oxoethyl)thio)-N-(4H-1,2,4-triazol-3-yl)acetamide (4b). White solid; yield: 79%; mp = 219°C; IR (KBr) 3233, 1672, 1602 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6) 3.42 (s, 2H), 4.22 (s, 2H), 7.60 (d, J = 8.4 Hz, 2H), 7.70 (s, 1H) 7.98 (d, J = 8.4 Hz, 2H), 11.64 (bs, 1H), 13.40 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 34.6, 38.4, 129.3, 130.8, 134.4, 138.8, 148.5, 149.2, 168.3, 194.0; ESI-mass (m/z) calcd $[C_{12}H_{11}ClN_4O_2S]^+$ 310.03, found 310.9.

2-((2-(4-Cyanophenyl)-2-oxoethyl)thio)-N-(4H-1,2,4-triazol-3-yl)acetamide (4c). Yellow solid; yield: 80%; mp = 231°C; IR (KBr) 3229, 2233, 1675, 1596 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6) 3.41 (s, 2H), 4.24 (s, 2H), 7.82 (s, 1H), 8.00 (d, J = 8.2 Hz, 2H), 8.10 (d, J = 8.2 Hz, 2H), 11.65 (bs, 1H), 13.41 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 34.6, 38.6, 115.7, 118.5, 129.5, 133.1, 139.0, 148.7, 149.2, 168.0, 194.2; ESI-mass (m/z) calcd $[C_{13}H_{11}N_5O_2S + H]^+$ 302.06, found 302.0.

2-((2-Oxo-2-(p-tolyl)ethyl)thio)-N-(4H-1,2,4-triazol-3-yl)acetamide (4d). White solid; yield: 90%; mp = 223–224°C; IR (KBr) 3227, 1672, 1662, 1596 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6) 2.37 (s, 3H), 3.41 (s, 2H), 4.19 (s, 2H), 7.33 (d, J = 8.0 Hz, 2H), 7.70 (s, 1H), 7.87 (d, J = 8.0 Hz, 2H), 11.72 (bs, 1H), 13.48 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 21.6, 34.6, 38.4, 129.0, 129.7, 133.2, 144.4, 148.8, 149.0, 168.2, 194.6; ESI-mass (m/z) calcd $[C_{13}H_{14}N_4O_2S + H]^+$ 291.08, found 291.0.

2-((2-Oxo-2-(4-(trifluoromethoxy)phenyl)ethyl)thio)-N-(4H-1,2,4-triazol-3-yl)acetamide (4e). White solid; yield: 54%; mp = 216°C; IR (KBr) 3230, 1673, 1600, 1257, 1204, 1160 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6) 3.43 (s, 2H), 4.24 (s, 2H), 7.51 (d, J = 8.0 Hz, 2H), 7.70 (s, 1H), 8.10 (d, J = 8.8 Hz, 2H), 11.65 (bs, 1H), 13.40 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 34.6, 38.5, 119.8, 121.4,

131.4, 134.6, 148.5, 149.1, 152.1, 168.3, 193.8; ESI-mass (*m/z*) calcd $[C_{13}H_{11}F_3N_4O_3S]^+$ 360.05, found 360.6.

2-((2-(3-Methoxyphenyl)-2-oxoethyl)thio)-N-(4H-1,2,4-triazol-3-yl)acetamide (4f). White solid; yield: 84%; mp = 205°C; IR (KBr) 3252, 1682, 1615, 1593 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) 3.43 (s, 2H), 3.82 (s, 3H), 4.23 (s, 2H), 7.21–7.24 (m, 1H), 7.43–7.47 (m, 2H), 7.57 (d, *J* = 6.8 Hz, 1H), 7.71 (s, 1H), 11.66 (bs, 1H), 13.42 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 34.6, 38.6, 55.8, 113.4, 119.8, 121.4, 130.3, 137.2, 148.7, 149.2, 159.9, 167.3, 194.8; ESI-mass (*m/z*) calcd $[C_{13}H_{14}N_4O_3S]^+$ 306.08, found 306.6.

2-((2-(3-Chlorophenyl)-2-oxoethyl)thio)-N-(4H-1,2,4-triazol-3-yl)acetamide (4g). White solid; yield: 61%; mp = 204°C; IR (KBr) 3238, 1672, 1600 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) 3.42 (s, 2H), 4.23 (s, 2H), 7.54–7.56 (m, 1H), 7.70–7.73 (m, 2H), 7.95–7.97 (m, 1H), 7.98 (s, 1H), 11.63 (bs, 1H), 13.39 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 34.7, 38.5, 127.5, 128.5, 131.1, 133.5, 134.1, 137.6, 148.8, 149.1, 168.4, 193.9; ESI-mass (*m/z*) calcd $[C_{12}H_{11}ClN_4O_2S]^+$ 310.03, found 310.9.

2-((2-(3-Nitrophenyl)-2-oxoethyl)thio)-N-(4H-1,2,4-triazol-3-yl)acetamide (4h). \Pale yellow solid; yield: 67%; mp = 201°C; IR (KBr) 3083, 1685, 1678, 1605, 1528 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) 3.44 (s, 2H), 4.32 (s, 2H), 7.69 (s, 1H), 7.82 (t, *J* = 7.6 Hz, 1H), 8.39 (d, *J* = 7.6 Hz, 1H), 8.45–8.48 (m, 1H), 8.66 (s, 1H), 11.64 (bs, 1H), 13.38 (bs, 1H); not sufficiently soluble to record ^{13}C NMR spectrum; ESI-mass (*m/z*) calcd $[C_{12}H_{11}N_5O_4S]^+$ 321.05, found 321.9.

2-(2-((2-((4H-1,2,4-Triazol-3-yl)amino)-2-oxoethyl) thio)-1-aryl ethylidene)hydrazinecarboxamide (5). The semicarbazide was prepared by dissolving 0.78 g of semicarbazide hydrochloride in 5 mL of water and adding to this a solution of 1.15 g of potassium acetate in 5 mL of methanol. The potassium chloride thus formed was filtered out after 30 min, leaving the semicarbazide solution as the filtrate. A solution of 0.5 g of ketone **4** in 3 mL of methanol was treated with the prepared semicarbazide reagent. The mixture was refluxed for 8 h. The solvent was removed, and the crude mass was poured onto crushed ice, filtered to give the respective semicarbazones **5**. Compounds **5a–5h** were prepared by this method.

2-(2-((2-((4H-1,2,4-Triazol-3-yl)amino)-2-oxoethyl) thio)-1-phenylethylidene)hydrazinecarboxamide (5a). White solid; yield: 96%; mp = 241°C; IR (KBr) 3477, 3218, 1676, 1601, 1548 cm^{-1}; ^1H NMR (300 MHz, DMSO-d_6) 3.50 (s, 2H), 4.02 (s, 2H), 6.59 (bs, 2H), 7.33 (m, 3H), 7.71 (s, 1H), 7.86 (m, 2H), 9.61 (bs, 1H), 11.70 (bs, 1H), 13.48 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 26.1, 35.1, 126.5, 128.7, 129.0, 137.0, 141.7, 148.8, 149.2, 157.2, 168.8; ESI-mass (*m/z*) calcd $[C_{13}H_{15}N_7O_2S - H]^+$ 333.10, found 332.0.

2-(2-((2-((4H-1,2,4-Triazol-3-yl)amino)-2-oxoethyl)thio)-1-(4-chlorophenyl)ethylidene) hydrazine carboxamide (5b). White solid; yield: 88%; mp = 250°C; IR (KBr) 3479, 3228, 1680, 1603, 1552 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) 3.50 (s, 2H), 4.02 (s, 2H), 6.63 (bs, 2H), 7.38 (d, *J* = 8.4 Hz, 2H) 7.71 (s, 1H), 7.91 (d, *J* = 8.4 Hz, 2H), 9.67 (bs, 1H), 11.68 (bs, 1H), 13.46 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 25.5, 34.7, 127.9, 128.2, 133.2, 135.4, 140.2, 148.0, 148.7, 156.7, 168.2; ESI-mass (*m/z*) calcd $[C_{13}H_{14}ClN_7O_2S - H]^+$ 367.06, found 366.0.

2-(2-((2-((4H-1,2,4-Triazol-3-yl)amino)-2-oxoethyl)thio)-1-(4-cyanophenyl)ethylidene) hydrazinecarboxamide (5c). Pale yellow solid; yield: 86%; mp = 226°C; IR (KBr) 3467, 3238, 2223, 1688, 1588 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) 3.46 (s, 2H), 4.05 (s, 2H), 6.72 (bs, 2H), 7.73 (s, 1H), 7.78 (d, *J* = 8.4 Hz, 2H), 8.08 (d, *J* = 8.4 Hz, 2H), 9.88 (bs, 1H), 11.69 (bs, 1H), 13.52 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 25.3, 34.7, 110.6, 118.8, 126.8, 132.0, 139.6, 140.9, 148.4, 149.7, 156.5, 167.9; ESI-mass (*m/z*) calcd $[C_{14}H_{14}N_8O_2S]^+$ 358.10, found 358.8.

2-(2-((2-((4H-1,2,4-Triazol-3-yl)amino)-2-oxoethyl) thio)-1-(p-tolyl)ethylidene)hydrazinecarboxamide (5d). White solid; yield: 96%; mp = 248°C; IR (KBr) 3471, 3214, 1678, 1678, 1604, 1552 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) 2.29 (s, 3H), 3.49 (s, 2H), 4.00 (s, 2H), 6.56 (bs, 2H), 7.14 (d, *J* = 7.8 Hz, 2H), 7.71 (s, 1H), 7.75 (d, *J* = 7.8 Hz, 2H), 9.52 (bs, 1H), 11.67 (bs, 1H), 13.47 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 21.2, 26.1, 35.1, 126.4, 129.3, 134.2, 138.5, 141.8, 148.4, 149.2, 157.2, 168.8; ESI-mass (*m/z*) calcd $[C_{14}H_{17}N_7O_2S]^+$ 347.12, found 347.8.

2-(2-((2-((4H-1,2,4-Triazol-3-yl)amino)-2-oxoethyl) thio)-1-(4-(trifluoromethoxy)phenyl)ethylidene) hydrazinecarboxamide (5e). White solid; yield: 70%; mp = 218–219°C; IR (KBr) 3479, 3233, 1680, 1557, 1256, 1210 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) 3.50 (s, 2H), 4.04 (s, 2H), 6.63 (bs, 2H), 7.30 (d, *J* = 8.5 Hz, 2H), 7.71 (s, 1H), 8.00 (d, *J* = 8.5 Hz, 2H), 9.70 (bs, 1H), 11.69 (bs, 1H), 13.45 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 26.1, 35.1, 121.0, 121.4, 121.8, 124.1, 131.2, 131.4. 148.8, 149.1, 152.2, 168.4; ESI-mass (*m/z*) calcd $[C_{14}H_{14}F_3N_7O_3S - H]^+$ 417.08, found 416.0.

2-(2-((2-((4H-1,2,4-Triazol-3-yl)amino)-2-oxoethyl)thio)-1-(3-methoxyphenyl)ethylidene) hydrazinecarboxamide (5f). White solid; yield: 94%; mp = 224°C; IR (KBr) 3474, 3225, 1677, 1614, 1586, 1556 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) 3.50 (s, 2H), 3.76 (s, 3H), 4.01 (s, 2H), 6.58 (bs, 2H), 6.89–6.94 (m, 1H), 7.25 (t, *J* = 7.9 Hz, 1H), 7.38–7.53 (m, 2H), 7.71 (s, 1H), 9.61 (bs, 1H), 11.68 (bs, 1H), 13.45 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 26.0, 34.7, 55.1, 111.5, 114.4, 118.6, 129.3, 138.1, 141.4, 148.1, 148.8, 156.7, 159.3, 168.8; ESI-mass (*m/z*) calcd $[C_{14}H_{17}N_7O_3S - H]^+$ 363.11, found 361.8.

2-(2-((2-((4H-1,2,4-Triazol-3-yl)amino)-2-oxoethyl)thio)-1-(3-chlorophenyl)ethylidene) hydrazinecarboxamide (5g). White solid; yield: 84%; mp = 262°C; IR (KBr) 3478, 3215, 1679, 1606, 1551 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) 3.50 (s, 2H), 4.02 (s, 2H), 6.69 (bs, 2H), 7.33–7.37 (m, 2H), 7.71 (s, 1H), 7.79–7.81 (m, 1H), 7.97 (s, 1H), 9.69 (bs, 1H), 11.69 (bs, 1H), 13.46 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 26.0, 35.1, 125.2, 126.1, 128.7, 130.4, 133.8, 139.1, 140.4, 148.6, 149.2, 157.1, 168.5; ESI-mass (*m/z*) calcd $[C_{13}H_{14}ClN_7O_2S]^+$ 367.06, found 367.9.

2-(2-((2-((4H-1,2,4-Triazol-3-yl)amino)-2-oxoethyl)thio)-1-(3-nitrophenyl)ethylidene) hydrazinecarboxamide (5h). Yellow solid; yield: 89%; mp = 264°C; IR (KBr) 3482, 3216, 1686, 1610, 1558, 1529 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) 3.50 (s, 2H), 4.32 (s, 2H), 6.71 (bs, 2H), 7.63 (d, *J* = 7.2 Hz, 1H), 7.70 (s, 1H), 8.15 (d, *J* = 7.3 Hz, 1H), 8.35–8.40 (m, 1H), 8.57 (s, 1H), 9.85 (bs, 1H), 11.66 (bs, 1H), 13.42 (bs, 1H); not quite soluble to record ^{13}C NMR spectrum; ESI-mass (*m/z*) calcd $[C_{13}H_{14}N_8O_4S - H]^+$ 378.09, found 377.0.

2-((4-Aryl-1,2,3-thiadiazol-5-yl)thio)-N-(4H-1,2,4- triazol-3-yl)acetamide (6). The semicarbazone **5** (0.001 mol) was added to 10 mL of thionyl chloride and cooled to −15°C. The mixture was then slowly stirred at RT for 12 h. The excess thionyl chloride was decomposed by pouring an aqueous solution of sodium carbonate. The product was filtered and dried. Compounds **6** were prepared by this method.

2-((4-Phenyl-1,2,3-thiadiazol-5-yl)thio)-N-(4H-1,2,4-triazol-3-yl)acetamide (6a). White solid; yield: 73%; mp = 199°C; IR

(KBr) 3234, 2898, 1681, 1603, 1549 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) 4.25 (s, 2H), 7.52 (t, J = 7.9 Hz, 1H), 7.59 (t, J = 7.9 Hz, 2H), 7.73 (bs, 1H), 7.87 (d, J = 7.9 Hz, 2H), 12.01 (bs, 1H), 13.53 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 40.7, 127.1, 128.4, 129.2, 130.3, 147.2, 148.2, 149.4, 156.9, 166.7; ESI-mass (*m/z*) calcd $[C_{12}H_{10}N_6OS_2 - H]^+$ 318.04, found 317.0.

Anal. Calcd for $C_{12}H_{10}N_6OS$: C, 45.27; H, 3.17; N, 26.40. Found: C, 45.32; H, 2.99; N, 26.58%.

2-((4-(4-Chlorophenyl)-1,2,3-thiadiazol-5-yl)thio)-N-(4H-1,2,4-triazol-3-yl)acetamide (6b). Pale yellow solid; yield: 63%; mp = 202°C; IR (KBr) 3113, 2904, 1690, 1627, 1594 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) 4.24 (s, 2H), 7.65 (d, J = 8.4 Hz, 2H), 7.90 (d, J = 8.4 Hz, 2H), 8.00 (bs, 1H), 11.63 (bs, 1H), 13.51 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 41.2, 129.4,* 129.6, 130.5, 134.3, 147.0, 149.0, 156.3, 166.0; ESI-mass (*m/z*) calcd $[C_{12}H_9ClN_6OS_2 - H]^+$ 352.0, found 351.0. *Two carbon signals merged here.

Anal. Calcd for $C_{12}H_9ClN_6OS_2$: C, 40.85; H, 2.57; N, 23.82. Found: C, 40.78; H, 2.62; N, 23.79%.

2-((4-(p-Tolyl)-1,2,3-thiadiazol-5-yl)thio)-N-(4H-1,2,4-triazol-3-yl)acetamide (6d). White solid; yield: 78%; mp = 217–218°C; IR (KBr) 3232, 2951, 1681, 1601, 1552 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) 2.38 (s, 3H), 4.22 (s, 2H), 7.38 (d, J = 7.9 Hz, 2H), 7.75 (d, J = 7.9 Hz, 2H), 7.75 (s, 1H), 11.94 (bs, 1H), 13.51 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 20.9, 40.6, 127.4, 128.2, 128.8, 129.0, 138.8, 147.5, 148.9, 157.4, 166.1; ESI-mass (*m/z*) calcd $[C_{13}H_{12}N_6OS_2 - H]^+$ 332.05, found 330.8.

Anal. Calcd for $C_{13}H_{12}N_6OS_2$: C, 46.97; H, 3.64; N, 25.28. Found: C, 46.83; H, 3.58; N, 25.32%.

N-(4H-1,2,4-Triazol-3-yl)-2-((4-(4-(trifluoromethoxy) phenyl)-1,2,3-thiadiazol-5-yl)thio)acetamide (6e). White solid; yield: 52%; mp = 197°C; IR (KBr) 3233, 2906, 1682, 1609, 1259 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) 4.25 (s, 2H), 7.59 (d, J = 8.4 Hz, 2H), 7.72 (s, 1H), 7.80 (d, J = 8.4 Hz, 2H), 11.96 (bs, 1H), 13.51 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 41.2, 116.6, 119.2, 121.8, 122.2, 129.5, 130.0, 130.9, 149.1, 156.2, 166.2; ESI-mass (*m/z*) calcd $[C_{13}H_9F_3N_6O_2S_2 - H]^+$ 402.02, found 401.0.

Anal. Calcd for $C_{13}H_9F_3N_6O_2S_2$: C, 38.80; H, 2.25; N, 20.89. Found: C, 38.72; H, 2.18; N, 20.93%.

2-((4-(3-Methoxyphenyl)-1,2,3-thiadiazol-5-yl)thio)-N-(4H-1,2,4-triazol-3-yl)acetamide (6f). White solid; yield: 78%; mp = 200°C; IR (KBr) 3248, 2953, 1681, 1597, 1268 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) 3.82 (s, 3H), 4.25 (s, 2H), 7.09 (d, J = 7.9 Hz, 1H), 7.40–7.51 (m, 3H), 7.78 (s, 1H), 11.96 (bs, 1H), 13.51 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 40.9, 55.7, 114.1, 115.3, 121.0, 130.5, * 131.9, 148.1, 148.9, 157.0, 159.8, 166.1; ESI-mass (*m/z*) calcd $[C_{13}H_{12}N_6O_2S_2]^+$ 348.05, found 348.7. *Two carbon signals merged here.

Anal. Calcd for $C_{13}H_{12}N_6O_2S_2$: C, 44.82; H, 3.47; N, 24.12. Found: C, 44.76; H, 3.38; N, 24.23%.

2-((4-(3-Chlorophenyl)-1,2,3-thiadiazol-5-yl)thio)-N-(4H-1,2,4-triazol-3-yl)acetamide (6g). White solid; yield: 57%; mp = 218°C; IR (KBr) 3231, 2955, 1682, 1602, 1292 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) 4.25 (s, 2H), 7.58–7.64 (m, 3H), 7.84 (d, J = 6.9 Hz, 1H), 7.91(s, 1H), 12.01 (bs, 1H), 13.62 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 41.1, 127.4, 128.3, 129.5, 130.1, 131.3, 131.4, 132.7, 134.0, 149.7, 155.9, 166.1; ESI-mass (*m/z*) calcd $[C_{12}H_9ClN_6OS_2 - H]^+$ 352.0, found 351.0.

Anal. Calcd for $C_{12}H_9ClN_6OS_2$: C, 40.85; H, 2.57; N, 23.82. Found: C, 40.77; H, 2.48; N, 23.86%.

2-((4-Aryl-1,2,3-selenadiazol-5-yl)thio)-N-(4H-1,2,4-triazol-3-yl)acetamide (7). To a solution of 0.001 mol of the appropriate semicarbazone dissolved in absolute alcohol, 0.01 mol of powdered selenium dioxide was added portion wise with stirring. The reaction mixture was heated to reflux at 95°C for 12 h. The deposited selenium was removed by filtration, and the filtrate was poured into crushed ice and the solid filtered. Compounds **7** were prepared by this method.

2-((4-Phenyl-1,2,3-selenadiazol-5-yl)thio)-N-(4H-1,2,4-triazol-3-yl)acetamide (7a). Brown solid; yield: 42%; mp = 156°C; IR (KBr) 3231, 2894, 1681, 1601, 1548 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) 4.23 (s, 2H), 7.48–7.51 (m, 1H), 7.56 (t, J = 7.4 Hz, 2H), 7.79 (bs, 1H), 7.80 (d, J = 7.4 Hz, 2H), 11.94 (bs, 1H), 13.51 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 44.0, 129.1, 129.2, 129.3, 129.4, 131.6, 149.3, 156.6, 157.0, 168.2; ESI-mass (*m/z*) calcd $[C_{12}H_{10}N_6OSSe - H]^+$ 365.98, found 364.9.

Anal. Calcd for $C_{12}H_{10}N_6OSSe$: C, 39.46; H, 2.76; N, 23.01. Found: C, 39.32; H, 2.71; N, 23.17%.

2-((4-(4-Chlorophenyl)-1,2,3-selenadiazol-5-yl)thio)-N-(4H-1,2,4-triazol-3-yl)acetamide (7b). White solid; yield: 33%; mp = 198–200°C; IR (KBr) 3096, 2852, 1677, 1583, 1531 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) 4.22 (s, 2H), 7.63 (d, J = 8.4 Hz, 2H), 7.84 (d, J = 8.4 Hz, 2H), 8.02 (bs, 1H), 11.58 (bs,1H); δ_C (100 MHz, DMSO-d_6) 43.6, 128.9, 130.3, 130.5, 133.3, 148.2, 149.4, 154.9, 157.1, 165.5; ESI-mass (*m/z*) calcd $[C_{12}H_9ClN_6OSSe - H]^+$ 399.94, found 398.8.

Anal. Calcd for $C_{12}H_9ClN_6OSSe$: C, 36.06; H, 2.27; N, 21.02. Found: C, 35.98; H, 2.19; N, 21.41%.

2-((4-(4-Cyanophenyl)-1,2,3-selenadiazol-5-yl)thio)-N-(4H-1,2,4-triazol-3-yl)acetamide (7c). Yellow solid; yield: 28%; mp = 168°C; IR (KBr) 3230, 2916, 1680, 1599 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) 4.26 (s, 2H), 7.98 (s, 1H), 8.03 (s, 4H), 11.60 (bs,1H), 1H signal not visible; ^{13}C NMR (100 MHz, DMSO-d_6) 44.4, 112.4, 119.0, 128.2, 128.5, 133.2, 133.6, 136.4, 154.8, 159.6, 165.9; ESI-mass (*m/z*) calcd for $[C_{13}H_9N_7OSSe - H]^+$ 390.98, found 389.8.

Anal. Calcd for $C_{13}H_9N_7OSSe$: C, 40.01; H, 2.32; N, 25.12. Found: C, 39.92; H, 2.30; N, 25.43%.

2-((4-(p-Tolyl)-1,2,3-selenadiazol-5-yl)thio)-N-(4H-1,2,4-triazol-3-yl)acetamide (7d). Brown solid; yield: 38%; mp = 154°C; IR (KBr) 3237, 2920, 1681, 1593, 1536 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) 2.39 (s, 3H), 4.20 (s, 2H), 7.20 (s, 1H), 7.28 (d, J = 8.4 Hz, 2H), 7.71 (d, J = 8.4 Hz, 2H), 11.90 (bs,1H), 13.53 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 20.9, 43.6, 128.8, 129.2, 129.3, 131.2, 138.3, 155.9, 156.1, 160.1, 166.2; ESI-mass (*m/z*) calcd $[C_{13}H_{12}N_6OSSe - H]^+$ 380.0, found 379.0.

Anal. Calcd for $C_{13}H_{12}N_6OSSe$: C, 41.17; H, 3.19; N, 22.16. Found: C, 41.10; H, 3.12; N, 22.20%.

]2-((4-(3-Methoxyphenyl)-1,2,3-selenadiazol-5-yl)thio)-N-(4H-1,2,4-triazol-3-yl)acetamide (7f). Brown solid; yield: 42%; mp = 160°C; IR (KBr) 3244, 2903, 1678, 1592, 1553 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) 3.81 (s, 3H), 4.21 (s, 2H), 7.05–7.07 (m, 1H), 7.34–7.38 (m, 2H), 7.48 (t, J = 7.9 Hz, 1H), 7.78 (s, 1H), 11.93 (bs,1H), 13.48 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 44.0, 55.7, 114.7, 114.9, 121.5, 130.4, 133.1, 148.1, 148.9, 156.2, 157.4, 159.8, 166.1; ESI-mass (*m/z*) calcd $[C_{13}H_{12}N_6O_2SSe - H]^+$ 395.99, found 397.0.

Anal. Calcd for $C_{13}H_{12}N_6O_2SSe$: C, 39.50; H, 3.06; N, 21.26. Found: C, 39.38; H, 2.28; N, 21.41%.

2-((4-(3-Chlorophenyl)-1,2,3-selenadiazol-5-yl)thio)-N-(4H-1,2,4-triazol-3-yl)acetamide (7g). Brown solid; yield 28%; mp 192°C; IR (KBr) 3231, 2910, 1681, 1604, 1559 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) 4.23 (s, 2H), 7.55–7.62 (m, 3H), 7.79 (d, J = 7.2 Hz, 1H), 7.86 (s, 1H), 11.92

(bs,1H), 13.52 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 44.0, 127.9, 128.8, 129.1, 129.3, 131.2, 133.8, 133.9, 155.0, 158.3, 166.0; ESI-mass (*m*/*z*) calcd $[C_{12}H_9ClN_6OSSe - H]^+$ 399.94, found 398.7.

Anal. Calcd for $C_{12}H_9ClN_6OSSe$: C, 36.06; H, 2.27; N, 21.02. Found: C, 35.98; H, 2.21; N, 20.97%.

2-Mercapto-N-phenylacetamide (9). Aniline (**8**) was treated with thioglycolic acid (**2**) in equimolar amount, and the reaction mixture was heated at 130°C for 3 h, and the resultant mass was worked out to give **9**, which was purified by crystallization.

2-((2-Oxo-2-arylethyl)thio)-N-phenylacetamide (10). The thiol **9** was treated with substituted phenacyl bromides and potassium carbonate in equimolar amount taken in ethanol. The mixture was left at room temperature for 6 h, and the resultant mass was worked out to give **10**, which was purified by crystallization. Compounds **10a–g** were prepared by this method.

2-(2-Oxo-2-phenylethyl)thio)-N-phenylacetamide (10a). White solid; yield: 92%; mp = 72°C; IR (KBr) 3300, 3048, 1692, 1597 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6) 3.37 (s, 2H), 4.21 (s, 2H), 7.04 (t, *J* = 7.1 Hz, 1H), 7.29 (t, *J* = 7.1 Hz, 2H), 7.54 (m, 4H), 7.66 (t, *J* = 7.3 Hz, 1H), 7.98 (d, *J* = 7.1 Hz, 2H), 10.09 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 36.0, 38.3, 119.6, 123.8, 128.9, 129.2, 133.9, 134.1, 135.7, 139.3, 167.7, 195.1; ESI-mass (*m*/*z*) calcd $[C_{16}H_{15}NO_2S]^+$ 285.08, found 285.7.

2-((2-(4-Chlorophenyl)-2-oxoethyl)thio)-N-phenylacetamide (10b). White solid; yield: 86%; mp = 121–122°C; IR (KBr) 3325, 1661, 1588, 1512 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6) 3.36 (s, 2H), 4.20 (s, 2H), 7.04 (t, *J* = 7.2 Hz, 1H), 7.29 (t, *J* = 7.2 Hz, 2H), 7.53–7.60 (m, 4H), 7.99 (d, *J* = 8.4 Hz, 2H), 10.09 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 35.5, 37.9, 119.2, 123.4, 128.7, 128.8, 130.4, 134.0, 138.3, 138.9, 167.2, 193.7; ESI-mass (*m*/*z*) calcd $[C_{16}H_{14}ClNO_2S]^+$ 320.04, found 320.0.

2-((2-(4-Cyanophenyl)-2-oxoethyl)thio)-N-phenylacetamide (10c). White solid; yield 88%; mp 94–95°C; IR (KBr) 3292, 2231, 1681, 1596 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6) 3.37 (s, 2H), 4.25 (s, 2H), 7.05 (t, *J* = 7.3 Hz, 1H), 7.30 (t, *J* = 7.3 Hz, 2H), 7.54 (d, *J* = 7.3 Hz, 2H), 7.99 (d, *J* = 8.2 Hz), 8.01 (d, *J* = 8.2 Hz, 2H), 10.09 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 35.5, 38.0, 115.2, 118.0, 119.2, 123.3, 128.6, 129.0, 132.7, 138.6, 138.8, 167.0, 193.9; ESI-mass (*m*/*z*) calcd $[C_{17}H_{14}N_2O_2S - H]^+$ 310.08, found 309.0.

2-(2-Oxo-2-(p-tolyl)ethyl)thio)-N-phenylacetamide (10d). White solid; yield: 90%; mp = 129–130°C; IR (KBr) 3300, 1656, 1599, 1518 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6) 2.37 (s, 3H), 3.36 (s, 2H), 4.17 (s, 2H), 7.04 (t, *J* = 7.4 Hz, 1H), 7.27–7.33 (m, 4H), 7.55 (d, *J* = 7.4 Hz, 2H), 7.87 (d, *J* = 8.2 Hz, 2H), 10.08 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 21.6, 36.0, 38.3, 119.6, 123.8, 129.0, 129.1, 129.7, 133.3, 139.3, 144.3, 167.7, 194.7; ESI-mass (*m*/*z*) calcd $[C_{17}H_{17}NO_2S]^+$ 299.10, found 299.7.

2-(2-(4-Fluorophenyl)-2-oxoethyl)thio)-N-phenylacetamide (10e). White solid; yield: 90%; mp = 117–118°C; IR (KBr) 3326, 1665, 1593, 1517 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6) 3.38 (s, 2H), 4.21 (s, 2H), 7.05 (t, *J* = 7.3 Hz, 1H), 7.28–7.39 (m, 4H), 7.55 (d, *J* = 7.3 Hz, 2H), 8.06–8.09 (m, 2H), 10.10 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 35.6, 37.9, 115.8, 119.2, 123.4, 128.7, 131.6, 132.0, 138.9, 163.9, 164.4, 193.4; ESI-mass (*m*/*z*) calcd $[C_{16}H_{14}FNO_2S + H]^+$ 303.07, found 304.2.

2-(2-(3-Nitrophenyl)-2-oxoethyl)thio)-N-phenyl acetamide (10f). Viscous liquid; yield: 84%; ^{1}H NMR (400 MHz, DMSO-d_6) 3.38 (s, 2H), 4.31 (s, 2H), 7.03 (t, *J* = 7.4 Hz, 1H), 7.28 (t, *J* = 7.4 Hz, 2H), 7.52 (d, *J* = 7.4 Hz, 2H), 7.83 (t, *J* = 7.8 Hz, 1H), 8.40–8.42 (m, 2H), 8.45–8.48 (m, 1H), 10.07 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 36.0, 38.5, 119.5, 123.3, 123.8, 128.0, 129.1, 131.0, 135.2, 137.0, 139.3, 148.4, 167.5, 193.6; ESI-mass (*m*/*z*) calcd $[C_{16}H_{14}N_2O_4S - H]^+$ 330.07, found 328.8.

2-(2-(3-Methoxyphenyl)-2-oxoethyl)thio)-N-phenylacetamide (10g). Colorless viscous liquid; yield: 84%; ^{1}H NMR (400 MHz, DMSO-d_6) 3.36 (s, 2H), 3.80 (s, 3H), 4.20 (s, 2H), 7.04 (t, *J* = 7.6 Hz, 1H), 7.20–7.23 (m, 1H), 7.29 (t, *J* = 7.6 Hz, 2H), 7.44 (m, 2H), 7.53–7.58 (m, 3H), 10.07 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 35.5, 37.9, 55.3, 113.0, 119.1, 119.4, 120.9, 123.3, 128.7, 129.8, 136.7, 138.8, 159.3, 167.2, 194.5; ESI-mass (*m*/*z*) calcd $[C_{17}H_{17}NO_3S + H]^+$ 315.09, found 316.0.

2-(2-(2-Oxo-2-(phenylamino)ethyl)thio)-1-arylethylidene) hydrazinecarboxamide (11). The semicarbazide was prepared by dissolving 0.78 g of semicarbazide hydrochloride in 5 mL of water; and adding to this, a solution of 1.15 g of potassium acetate in 5 mL of methanol. The potassium chloride thus formed was filtered out after 30 min, leaving the semicarbazide solution as the filtrate. A solution of 0.5 g of ketone **10** in 3 mL of methanol was treated with the prepared semicarbazide reagent. The mixture was refluxed for 8 h. The solvent was removed and the crude mass was poured onto crushed ice, filtered to give the respective semicarbazones **5**. Compounds **11a–11f** were prepared by this method.

2-(2-((2-Oxo-2-(phenylamino)ethyl)thio)-1-phenylethylidene) hydrazine carboxamide (11a). White solid; yield: 92%; mp = 145–146°C; IR (KBr) 3475, 3303, 1685, 1657 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6) 3.41 (s, 2H), 4.05 (s, 2H), 6.58 (bs, 2 H), 7.07 (t, *J* = 7.3 Hz, 1H), 7.27–7.41 (m, 5H), 7.59 (d, *J* = 7.9 Hz, 2H), 7.89 (d, *J* = 7.8 Hz, 2H), 9.69 (bs, 1H), 10.18 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 26.1, 35.8, 119.7, 119.8, 124.0, 126.5, 128.1, 129.0, 129.1, 129.2, 137.0, 139.2, 157.2, 168.3; ESI-mass (*m*/*z*) calcd $[C_{17}H_{18}N_4O_2S + H]^+$ 342.12, found 343.0.

2-(1-(4-Chlorophenyl)-2-(2-oxo-2-(phenylamino) ethyl)thio) ethylidene)hydrazinecarboxamide (11b). White solid; yield: 88%; mp = 208–209°C; IR (KBr) 3473, 3303, 1682, 1657, 1560 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6) 3.38 (s, 2H), 4.03 (s, 2H), 6.62 (bs, 2H), 7.06 (t, *J* = 7.8 Hz, 1H), 7.31 (t, *J* = 7.8 Hz, 2H), 7.38 (d, *J* = 8.6 Hz, 2H), 7.57 (d, *J* = 7.8 Hz, 2H), 7.92 (d, *J* = 8.6 Hz, 2H), 9.75 (bs, 1H), 10.14 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 25.8, 35.8, 119.8, 124.0, 128.4, 128.6, 129.2, 133.6, 135.8, 139.2, 140.9, 157.1, 168.2; ESI-mass (*m*/*z*) calcd $[C_{17}H_{17}ClN_4O_2S - H]^+$ 376.08, found 375.0.

2-(1-(4-Cyanophenyl)-2-((2-oxo-2-(phenylamino)ethyl) thio) ethylidene)hydrazinecarboxamide (11c). White solid; yield: 84%; mp = 189–190°C; IR (KBr) 3476, 3306, 2233, 1690, 1657 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6) 3.39 (s, 2H), 4.07 (s, 2H), 6.73 (bs, 2H), 7.06 (t, *J* = 7.4 Hz, 1H), 7.31 (t, *J* = 7.4 Hz, 2H), 7.56 (d, *J* = 7.4 Hz, 2H), 7.78 (d, *J* = 8.4 Hz, 2H), 8.10 (d, *J* = 8.4 Hz, 2H), 9.94 (bs, 1H), 10.14 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 25.7, 35.8, 111.0, 119.3, 119.8, 124.0, 127.3, 129.2, 132.5, 133.2, 139.1, 141.4, 157.0, 168.2; ESI-mass (*m*/*z*) calcd $[C_{18}H_{17}N_5O_2S - H]^+$ 367.11, found 366.0.

2-(2-(2-Oxo-2-(phenylamino)ethyl)thio)-1-(p-tolyl)ethylidene) hydrazine carboxamide (11d). White solid; yield: 90%; mp = 213–214°C; IR (KBr) 3476, 3304, 1661, 1562 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6) 2.30 (s, 3H), 3.40 (s, 2H), 4.02 (s, 2H), 6.58 (bs, 2H), 7.08 (t, *J* = 7.4 Hz, 1H), 7.16 (d, *J* = 7.4 Hz, 2H), 7.33 (t, *J* = 7.4 Hz, 2H), 7.58 (d, *J* = 8.2 Hz, 2H), 7.79 (d, *J* = 8.2 Hz, 2H), 9.64 (bs, 1H), 10.17 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 21.2, 26.0, 35.8, 119.7, 124.0, 126.5,

129.2, 129.3, 134.2, 138.5, 139.2, 142.1, 157.3, 168.3; ESI-mass (*m/z*) calcd $[C_{18}H_{20}N_4O_2S + H]^+$ 356.13, found 357.1.

2-(1-(4-Fluorophenyl)-2-((2-oxo-2-(phenylamino)ethyl) thio) ethylidene)hydrazinecarboxamide (11e). White solid; yield: 86%; mp = 179–180°C; IR (KBr) 3475, 3306, 1685, 1657, 1563 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) 3.39 (s, 2H), 4.03 (s, 2H), 6.61 (bs, 2H), 7.06 (t, *J* = 7.8 Hz, 1H), 7.16 (t, *J* = 7.8 Hz, 2H), 7.31 (t, *J* = 7.8 Hz, 2H), 7.58 (d, *J* = 8.0 Hz, 2H), 7.93–7.96 (m, 2H), 9.69 (bs, 1H), 10.15 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 26.0, 35.7, 115.4, 119.7, 124.0, 128.8, 129.1, 133.4, 139.2, 141.1, 157.2, 163.0, 168.3; ESI-mass (*m/z*) calcd $[C_{17}H_{17}FN_4O_2S + H]^+$ 360.11, found 361.2.

2-(1-(3-Nitrophenyl)-2-(2-oxo-2-(phenylamino)ethyl) thio) ethylidene)hydrazinecarboxamide (11f). Yellow solid; yield: 82%; mp = 160–161°C; IR (KBr) 3489, 3187, 1672, 1602, 1521 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) 3.39 (s, 2H), 4.12 (s, 2H), 6.71 (bs, 2H), 7.05 (t, *J* = 7.4 Hz, 1H), 7.28–7.32 (m, 2H), 7.55–7.64 (m, 3H), 8.13–8.16 (m, 1H), 8.38 (d, *J* = 7.2 Hz, 1H), 8.60 (s, 1H), 9.93 (bs, 1H), 10.12 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 25.5, 35.2, 119.2, 120.6, 122.9, 123.5, 128.7, 129.6, 132.5, 138.3, 138.7, 139.6, 148.1, 156.6, 167.6; ESI-mass (*m/z*) calcd $[C_{17}H_{17}N_5O_4S + H]^+$ 387.10, found 388.1.

N-Phenyl-2-((4-aryl-1,2,3-thiadiazol-5-yl)thio) acetamide (12). The semicarbazone **11** (0.001 mol) was added to 10 mL of thionyl chloride and cooled to −78°C and stirred at that temperature for 4 h. Then the excess thionyl chloride was decomposed by pouring an aqueous solution of sodium carbonate. The product was filtered and dried. Compounds **12** were prepared by this method.

N-Phenyl-2-(4-phenyl-1,2,3-thiadiazol-5-yl)thio)acetamide (12a). White solid; yield: 62%; mp = 52–53°C; IR (KBr) 3265, 3051, 1662, 1597, 1539 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$) 3.85 (s, 2H), 7.15 (t, *J* = 7.1 Hz, 1H), 7.30–7.37 (m, 4H), 7.49–7.58 (m, 4H), 7.95 (d, *J* = 7.1 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) 41.9, 120.1, 125.2, 128.7, 128.8, 129.0, 129.4, 130.1, 136.6, 145.2, 158.7, 163.8; ESI-mass (*m/z*) calcd $[C_{16}H_{13}N_3OS_2 - H]^+$ 327.05, found 326.0.

Anal. Calcd for $C_{16}H_{13}N_3OS_2$: C, 58.69; H, 4.00; N, 12.83. Found: C, 58.62; H, 4.13; N, 12.88%.

2-(4-(4-Chlorophenyl)-1,2,3-thiadiazol-5-yl)thio)-N-phenylacetamide (12b). Viscous liquid; yield: 65%; ^1H NMR (400 MHz, $CDCl_3$) 3.86 (s, 2H), 7.16 (t, *J* = 7.2 Hz, 1H), 7.31 (t, *J* = 7.2 Hz, 2H), 7.39 (d, *J* = 7.2 Hz, 2H), 7.52 (d, *J* = 8.4 Hz, 2H), 7.90 (d, *J* = 8.4 Hz, 2H); NH not seen; ^{13}C NMR (100 MHz, $CDCl_3$) 41.9, 120.0, 125.3, 128.6, 128.7, 129.1, 130.0, 135.6, 136.6, 145.2, 157.7, 163.5; ESI-mass (*m/z*) calcd $[C_{16}H_{12}ClN_3OS_2 + H]^+$ 361.01, found 362.0.

Anal. Calcd For $C_{16}H_{12}ClN_3OS_2$: C, 53.11; H, 3.34; N, 11.61. Found: C, 53.17; H, 3.29; N, 11.65%.

N-Phenyl-2-((4-(p-tolyl)-1,2,3-thiadiazol-5-yl)thio)acetamide (12d). White solid; yield: 68%; mp = 136–137°C; IR (KBr) 3257, 3035, 1662, 1597, 1527 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$) 2.46 (s, 3H), 3.83 (s, 2H), 7.15 (t, *J* = 7.2 Hz, 1H), 7.29−7.37 (m, 6H), 7.84 (d, *J* = 8.1 Hz, 2H), 7.95 (bs, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) 21.3, 41.9, 120.0, 125.1, 127.2, 128.6, 129.0, 129.5, 136.6, 139.6, 144.2, 159.1, 163.8; ESI-mass (*m/z*) calcd $[C_{17}H_{15}N_3OS_2 + H]^+$ 341.07, found 342.1.

Anal. Calcd for $C_{17}H_{15}N_3OS_2$: C, 59.80; H, 4.43; N, 12.31. Found: C, 59.79; H, 4.37; N, 12.38%; Calcd for $C_{17}H_{15}N_3OS_2$: C, 59.80; H, 4.43; N, 12.31. Found: C, 59.79; H, 4.37; N, 12.38%.

2-(4-(4-Fluorophenyl)-1,2,3-thiadiazol-5-yl)thio)-N-phenylacetamide (12e). White solid; yield: 63%; mp = 140–141°C; IR (KBr) 3251, 3058, 1658, 1597, 1535 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$) 3.75 (s, 2H), 7.16 (t, *J* = 7.2 Hz, 1H), 7.21–7.26 (m, 2H), 7.33 (t, *J* = 7.2 Hz, 2H), 7.38 (d, *J* = 7.2 Hz, 2H), 7.86 (bs, 1H), 7.93–7.96 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) 41.9, 116.0, 120.1, 125.3, 126.3, 129.1, 130.7, 130.8, 136.6, 145.0, 160.0, 164.5; ESI-mass (*m/z*) calcd $[C_{16}H_{12}FN_3OS_2 + H]^+$ 345.04, found 346.1.

Anal. Calcd for $C_{16}H_{12}FN_3OS_2$: C, 55.63; H, 3.50; N, 12.17. Found: C, 55.57; H, 3.48; N, 12.22%.

N-Phenyl-2-((4-aryl-1,2,3-selenadiazol-5-yl)thio)acetamide (13). To a solution of 0.001 mol of the appropriate semicarbazone dissolved in tetrahydrofuran, 0.01 mol of powdered selenium dioxide was added portion wise with stirring. The reaction mixture was heated to reflux at 95°C for 2 h. The deposited selenium was removed by filtration, and the filtrate was poured into crushed ice and the solid filtered. Compounds **13** were prepared by this method.

N-Phenyl-2-((4-phenyl-1,2,3-selenadiazol-5-yl)thio)acetamide (13a). Viscous liquid; yield: 68%; ^1H NMR (400 MHz, DMSO-d_6) 3.87 (s, 2H), 7.16 (t, *J* = 7.2 Hz, 1H), 7.33 (t, *J* = 7.2 Hz, 2H), 7.40 (d, *J* = 8.5 Hz, 2H), 7.50–7.65 (m, 3H), 7.89 (d, *J* = 8.5 Hz, 2H), 8.00 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 44.5, 119.3, 120.1, 123.7, 128.6, 128.7, 130.3, 131.5, 138.3, 155.9, 156.6, 165.2; ESI-mass (*m/z*) calcd $[C_{16}H_{13}N_3OSSe + H]^+$ 374.99, found 375.0.

Anal. Calcd for $C_{16}H_{13}N_3OSSe$: C, 51.34; H, 3.50; N, 11.23. Found: C, 51.26; H, 3.43; N, 11.20%.

2-((4-(4-Chlorophenyl)-1,2,3-selenadiazol-5-yl)thio)-N-phenylacetamide (13b). Viscous liquid; yield: 63%; ^1H NMR (400 MHz, DMSO-d_6) 4.19 (s, 2H), 7.08 (t, *J* = 7.2 Hz, 1H), 7.32 (t, *J* = 7.2 Hz, 2H), 7.55 (d, *J* = 8.4 Hz, 2H), 7.64 (d, *J* = 8.4 Hz, 2H), 7.86 (d, *J* = 7.2 Hz, 2H), 10.42 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 45.1, 119.7, 124.3, 129.3,* 130.9, 131.0, 133.7, 138.8, 155.2, 157.8, 165.7; ESI-mass (*m/z*) calcd $[C_{16}H_{12}ClN_3OSSe + H]^+$ 408.96, found 410.0. *Two carbon signals merged here.

Anal. Calcd for $C_{16}H_{12}ClN_3OSSe$: C, 47.01; H, 2.96; N, 10.28. Found: C, 46.97; H, 3.01; N, 10.32%.

2-((4-(4-Cyanophenyl)-1,2,3-selenadiazol-5-yl)thio)-N-phenylacetamide (13c). White solid; yield: 62%; mp = 161–162°C; IR (KBr) 3254, 2226, 1652, 1599 cm^{-1}; ^1H NMR (400 MHz, Acetone-d_6) 4.23 (s, 2H), 7.12 (t, *J* = 7.2 Hz, 1H), 7.34 (t, *J* = 7.2 Hz, 2H), 7.65 (d, *J* = 7.2 Hz, 2H), 8.00 (d, *J* = 8.2 Hz, 2H), 8.16 (d, *J* = 8.2 Hz, 2H), 9.65 (bs, 1H); ^{13}C NMR (100 MHz, Acetone-d_6) 44.9, 111.8, 118.2. 119.4, 124.1, 128.8, 129.7, 132.5, 136.4, 138.5, 154.7, 158.5, 165.0; ESI-mass (*m/z*) calcd $[C_{17}H_{12}N_4OSSe + H]^+$ 399.99, found 401.0.

Anal. Calcd for $C_{17}H_{12}N_4OSSe$: C, 51.13; H, 3.03; N, 14.03. Found: C, 51.17; H, 3.05; N, 13.97%.

N-Phenyl-2-((4-(p-tolyl)-1,2,3-selenadiazol-5-yl)thio)acetamide (13d). Viscous liquid; yield: 71%; ^1H NMR (400 MHz, DMSO-d_6) 2.31 (s, 3H), 4.15 (s, 2H), 7.08 (t, *J* = 7.8 Hz, 1H), 7.32 (t, *J* = 8.0 Hz, 2H), 7.37 (d, *J* = 8.0 Hz, 2H), 7.56 (d, *J* = 8.0 Hz, 2H), 7.71 (d, *J* = 8.0 Hz, 2H); ^{13}C NMR (100 MHz, DMSO-d_6) 21.3, 45.0, 119.7, 124.3, 129.2, 129.3, 129.8,* 138.7, 138.9, 156.4, 156.5, 165.8; ESI-mass (*m/z*) calcd $[C_{17}H_{15}N_3OSSe + H]^+$ 389.01, found 390.0. *Two carbon signals merged here.

Anal. Calcd for $C_{17}H_{15}N_3OSSe$: C, 52.58; H, 3.89; N, 10.82. Found: C, 52.47; H, 3.92; N, 10.81%.

2-((4-(4-Fluorophenyl)-1,2,3-selenadiazol-5-yl)thio)-N-phenylacetamide (13e). Viscous liquid; yield: 66%; ^1H NMR (400 MHz, DMSO-d_6) 4.17 (s, 2H), 7.08 (t, *J* = 7.4 Hz, 1H), 7.32 (t, *J* = 7.4 Hz, 2H), 7.39–7.45 (m, 2H), 7.55 (d, *J* = 8.7 Hz, 2H), 7.84–7.88 (m, 2H), 10.40 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 44.9, 116.8, 119.6, 124.3, 128.5, 131.5, 134.1, 138.8, 155.5, 161.2, 163.6, 165.8; ESI-mass (*m/z*) calcd $[C_{16}H_{12}FN_3OSSe - H]^+$ 392.99, found 391.2.

Anal. Calcd for $C_{16}H_{12}FN_3OSSe$: C, 48.98; H, 3.08; N, 10.71. Found: C, 48.92; H, 3.04; N, 10.78%.

2-((4-(3-Nitrophenyl)-1,2,3-selenadiazol-5-yl)thio)-N-phenylacetamide (13f). Yellow solid; yield: 32%; mp = 147–148°C; IR (KBr) 3282, 2956, 1653, 1520 cm^{-1}; ^1H NMR (400 MHz, DMSO-d_6) 4.25 (s, 2H), 7.10 (t, J = 7.6 Hz, 1H), 7.34 (t, J = 7.6 Hz, 2H), 7.57 (d, J = 7.6 Hz, 2H), 7.89 (t, J = 8.0 Hz, 1H), 8.32–8.36 (m, 2H), 8.68 (s, 1H), 10.44 (bs, 1H); ^{13}C NMR (100 MHz, DMSO-d_6) 49.7, 124.4, 128.1, 128.4, 129.0, 134.0, 135.7, 138.2, 140.1, 143.5, 153.1, 158.9, 164.0, 170.3; ESI-mass (*m*/*z*) calcd $[C_{16}H_{12}N_4O_3SSe + H]^+$ 419.98, found 420.9.

Anal. Calcd for $C_{16}H_{12}N_4O_3SSe$: C, 45.83; H, 2.88; N, 13.36. Found: C, 45.86; H, 2.80; N, 13.29%.

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