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Synthesis of 4'-[3-Methyl-5-thioxo-1*H*-1,2,4-triazol-4(5*H*)-yl]-2',5'-diphenyl-2',4'-dihydro Spiro[indolin-3,3'[1,2,4]triazol]-2-one Derivatives[1]

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Abstract—In this paper 1,3-dipolar cycloaddition reaction has been studied. An efficient synthesis of 4'-[3-methyl-5-thioxo-1*H*-1,2,4-triazol-4(5H)-yl]-2',5'-diphenyl-2',4'-dihydro spiro(indolin-3,3'[1,2,4]triazol)-2-one derivatives using triethylamine in MeCN at room temperature is reported. The structures of the obtained compounds were confirmed by means of elemental analysis, MS and spectral (IR, ^{1}H, and ^{13}C NMR) methods.

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INTRODUCTION

Chemistry of nitril imines has been developing for more than forty years already. Because of their chemical activity and readiness to undergo cycloaddition reaction with different substances they found important usage for preparation of substantial raw material [1–5]. Hidrazonyl halides are most usual starting materials used for preparation of nitril amines. Final cyclization step to form spiro structure is accomplished with a suitable base, such as triethyl amine. Mechanism of this reaction has been reported by Shavali [6–10]. Indol core of isatin has been long known for different biological activities, like anti-microbial anti-mold, anti-cancer [11–19]. Among these heterocycles, spiro indoles have been identified as privileged structures in medicinal chemistry and have attracted increasing interest in the recent years [20–22]. Considering the above reasons, the development of new and simple synthetic method for the efficient preparation of spiro indole heterocycles containing 1,2,4-triazol ring fragments is therefore an interesting challenge. We studied the reaction of isatin imine and (*Z*)-1-[chloro (phenyl) methylene]-2-phenylhydrazine in the presence of triethyl amine in MeCN at room temperature. The reaction was complete after 18–21 h (reaction progress was monitored by TLC) and 4'-[3-methyl-5-thioxo-1*H*-1,2,4-triazol-4(5*H*)-yl]-2',5'-diphenyl-2',4'-dihydro spiro(indolin-3,3'[1,2,4]triazol)-2-one derivatives were obtained in 75–95% yield.

[1] The text was submitted by the authors in English.

EXPERIMENTAL

All reagents used in this research were GR grade and all solvents were redistilled before use. Merk thin layer chromatography sheets Art no: 1 : 0554 were used for TLC. NMR spectra were recorded on an JEOL DELTA NMR 400 MHz (^{1}H NMR) and 100 MHz (^{13}C NMR) instruments, using $CDCl_3$ and DMSO-d_6 as solvent. IR spectra were recorded on a Perkin-Elmer model 1420 spectrophotometer.

Thiocarbonohydrazide was prepared from hydrazine hydrate and carbon disulfide in 89% yield. White crystals, mp, °C: 195–197; IR (KBr): 3461, 3272, 3204, 1300; ^{1}H NMR (400 MHz, $CDCl_3$), δ, ppm: 2 d (2H), 2.01–2.03 d (J = 2 and 8 Hz, 4H); ^{13}C NMR ($CDCl_3$), δ, ppm: 186; MS, *m/z*; 106 (M^+, 92.16); Calculated, %: C 11.31; H 5.70; N 52.78; S 30.21. CH_4N_4S. Found, %: C 11.28; H 5.32; N 52.80; S 30.20.

***N*'-Phenylbenzohydrazide** was prepared from 15 g of phenyl hydrazine and 19 g of benzoyl chloride in pyridine in 77% yield. White crystals; mp, °C: 202–204; IR (KBr): 3324, 3269, 3054, 1644; ^{1}H NMR (400 MHz, $CDCl_3$), δ, ppm: 4.01 d (1H), 6.66 m (1H), 7.18 q (2H), 7.44–7.46 m (J = 2 and 8 Hz, 2H), 7.51 t (1H), 7.95 m (2H), 8.00 m (1H); ^{13}C NMR ($CDCl_3$), δ, ppm: 113.2, 127.5, 128.9, 129.3, 132.2, 134.1, 151.5, 164.9; MS, *m/z*: 212 (M^+, 90.10); Calculated, %: C 73.56; H 5.70; N 13.20; O 7.54. $C_{13}H_{12}N_2O$. Found, %: C 73.52; H 5.72; N 13.18; O 7.50.

4-Amino-3-methyl-1*H*-1,2,4-triazol-5-thione (I). Thiocarbohydrazide, 2 g, and acetic acid, 16 mL, were

placed in a flask and heated on an oil bath to 120°C for 20 min to form clear solution that after 5 min suddenly started to form deposit. After heating for further 15 min the reaction mixture was cooled on an ice bath. Solid crystalline product was collected by filtration to give 1.08 g (91% yield) as white crystals; mp, °C: 198–201; IR (KBr): 3112, 2946, 1628, 1318; ^{1}H NMR (400 MHz, $CDCl_3$), δ, ppm: 0.9 s (3H), 2.00 d (2H), 7.01 d (1H); ^{13}C NMR ($CDCl_3$), δ, ppm: 22.7, 155, 186; MS, *m/z*: 130 (M^+, 85.00); Calculated, %: C 27.68; H 4.65; N 43.04; S 24.63. $C_3H_6N_4S$. Found, %: C 27.60; H 4.62; N 43.01; S 24.62.

(3*Z*)-3-[3-Methyl-5-thioxo-1*H*-1,2,4-triazol-4(5*H*)-ylimino]indolin-2-one (II). 1*H*-amino-3-methyl-1*H*-1,2,4-triazol-5-tion, 0.390 g, and isatin, 0.441g were mixed with 20 mL of acetonitrile and 20 drops of acetic acid, heated for 5 hours and kept for 1 day at room temperature. Crystalline deposit formed was washed with ethanol to give **II** in 89% yield as yellow crystals; mp, °C: 234–236; IR (KBr): 3254, 2925, 1752; ^{1}H NMR (400 MHz, $CDCl_3$), δ, ppm: 0.9 s (3H), 7.00 m (1H), 7.01 t (1H), 7.30 t (1H), 7.60–7.70 m (*J* = 2 and 8 Hz, 2H), 8.00 s (1H); ^{13}C NMR ($CDCl_3$), δ, ppm: 23.2, 117.8, 121.7, 124.5, 129.4, 131.3, 132.9, 146.8, 155.00, 167.5, 186; MS, *m/z*: 259 (M^+, 76.53); Calculated, %: C 50.95; H 3.50; N 27.01; O 6.17; S 12.37. $C_{11}H_9N_5OS$. Found, %: C 50.93; H 3.49; N 26.95; O 6.14; S 12.35.

(*Z*)-1-[Chloro(phenyl)methylene]-2-phenylhydrazine (III). Triphenyl phosphine, 1.55 g, and *N*'-phenylbenzohydrazide, 4.24 g, were mixed in a beaker with 40 mL of acetonitril at room temperature. After 30 min 1.95 mL of carbon tetrachloride were added slowly over 2 h to the suspension. The reaction continued for 20 h, then the beaker was placed on an ice bath for 30 min. Solid deposit was filtered off to give **III** as white crystals in 69% yield; mp, °C: 240–243; IR (KBr): 3304, 3051, 1598, 752; ^{1}H NMR (400 MHz, $CDCl_3$), δ, ppm: 6.46 t (2H), 6.62 t (1H), 7.00 m (1H), 7.02–7.04 m (*J* = 2 and 8 Hz, 2H), 7.30 d (3H), 7.60 q (2H); ^{13}C NMR ($CDCl_3$), δ, ppm: 116.3, 118.4, 128.9, 129.4, 131.2, 143.3, 155.00; MS, *m/z*: 230 (M^+, 65.56). Calculated, %: C 67.68; H 4.81; Cl 15.37; N 12.14. $C_{13}H_{11}ClN_2$. Found, %: C 67.62; H 4.78; Cl 15.32; N 12.20.

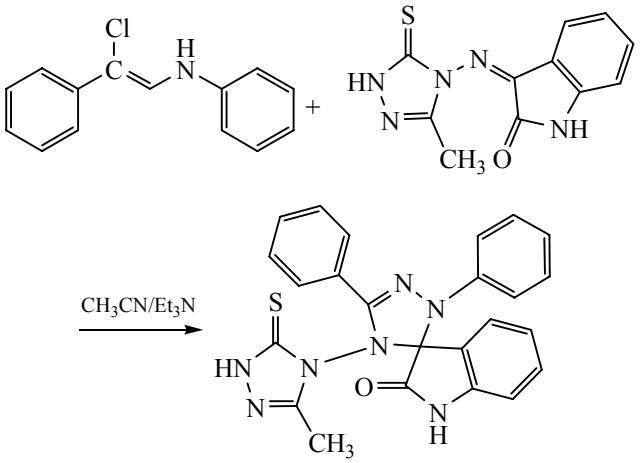
4-Chloro-*N*'-phenylbenzohydrazide (IV). Prepared similarly to *N*'-phenylbenzohydrazide in 70% yield. white crystals; mp, °C: 242–246; IR (KBr): 3347, 3237, 3054, 1648, 1095; ^{1}H NMR (400 MHz, $CDCl_3$), δ, ppm: 4.00 d (1H), 6.66 m (2H), 6.71 m (1H), 7.18 d (*J* = 2 and 8 Hz, 2H), 7.43–7.45 d (2H), 7.89 d (2H), 8.01 s (1H); ^{13}C NMR ($CDCl_3$), δ, ppm: 113.9, 119.2, 128.9, 129.00, 129.3, 132.3, 137.7, 151.1, 164.9; MS, *m/z*: 246 (M^+, 85.13); Anal. Calculated, %: C 63.29; H 4.49; Cl 14.37; N 11.36; O 6.49. $C_{13}H_{11}ClN_2O$. Found, %: C 63.30; H 4.46; Cl 14.33; N 11.35; O 6.51.

(*Z*)-1-[Chloro(4-chlorophenyl)methylene]-2-phenylhydrazine (V). Prepared similarly to phenylhydrazine **III** from 4-chloro-*N*'-phenylbenzohydrazide **IV** in 64% yield. white crystals; mp, °C: 251–253; IR (KBr): 3308, 3053, 1649, 753; ^{1}H NMR (400 MHz, $CDCl_3$), δ, ppm: 6.46 q (2H), 6.62 q (1H), 7.00 m (1H), 7.01–7.03 t (*J* = 2 and 8 Hz, 2H), 7.3 q (2H), 7.6 t (2H); ^{13}C NMR ($CDCl_3$), δ, ppm: 116.3, 118.8, 127.7, 129.00, 129.6, 130.6, 136.6, 143.1, 155.1; MS, *m/z*: 264 (M^+, 85.20); Calculated, %: C 58.89; H 3.80; Cl 26.74; N 10.57. $C_{13}H_{10}Cl_2N_2$. Found, %: C 58.85; H 3.79; Cl 26.75; N 10.52.

3,5-Dinitro-*N*'-phenylbenzohydrazide (VI). Prepared similarly to *N*'-phenylbenzohydrazide in 72% yield. silver crystals; mp, °C: 243–245; IR (KBr): 3281, 3094, 1648, 1541, 1345; ^{1}H NMR (400 MHz, $CDCl_3$), δ, ppm: 4.01 s (1H), 6.66 m (2H), 6.71 m (1H), 7.17–7.19 t (*J* = 2 and 8 Hz, 2H), 8.00–8.02 s (1H), 9.27 d (2H), 9.37 d (1H); ^{13}C NMR ($CDCl_3$), δ, ppm: 113.2, 119.2, 122.1, 128.5, 129.3, 136.00, 149.4, 151.00, 164.9; MS, *m/z*: 302 (M^+, 95.16); Calculated, %: C 51.66; H 3.33; N 18.54; O 26.47. $C_{13}H_{10}N_4O_5$. Found, %: C 51.64; H 3.30; N 18.53; O 26.45.

(*Z*)-1-[Chloro(3,5-dinitrophenyl)methylene]-2-phenylhydrazine (VII). Prepared similarly to phenylhydrazine **III** from 3,5-dinitro-*N*'-phenylbenzohydrazide **VI** in 66% yield. silver crystals; mp, °C: 250–252; IR (KBr): 3296, 3096, 1539, 1345, 725; ^{1}H NMR (400 MHz, $CDCl_3$), δ, ppm: 6.46 d (2H), 6.62 m (1H), 7.00 m (1H), 7.01 m (*J* = 2 and 8 Hz, 2H), 8.89–8.91 d (2H), 9.1 d (1H); ^{13}C NMR ($CDCl_3$), δ, ppm: 116.3, 118.7, 121.00, 129.6, 130.2, 131.4, 143.1, 149.4, 155; MS, *m/z*: 320 (M^+, 100.00); Calculated, %: C 48.69; H 2.83; Cl 11.06; N 17.47; O 19.96. $C_{13}H_9ClN_4O_4$. Found, %: C 48.67; H 2.82; Cl 11.01; N 17.42; O 19.93.

4'-[3-Methyl-5-thioxo-1*H*-1,2,4-triazol-4(5*H*)-yl]-2',5'-diphenyl-2',4'-dihydro spiro(indolin-3,3'[1,2,4]-triazol)-2-one (VIII). 1-(Chloro(phenyl)methylene)-2-phenyl hydrazine **III,** 0.07 g, and 3-[3-methyl-5-thioxo-1*H*-1,2,4-triazol-4(5*H*)-ylimino]indolin-2-one **II,** 0.08 g, were added to a mixture of 1 mL of triethyl amine and 4 mL of acetonitrile in a beaker at room



Synthetic route for the 4'-[3-methyl-5-thioxo-1*H*-1,2,4-triazol-4(5*H*)-yl]-2',5'-diphenyl-2',4'-dihydro spiro(indolin-3,3'-[1,2,4]triazol)-2-one derivatives.

temperature. Yield 89%; orange crystals; mp, °C: 301–303; IR (KBr): 3285, 3000, 1747, 1608; ^{1}H NMR (400 MHz, $CDCl_3$), δ, ppm: 0.9 s (3H), 6.43 m (2H), 6.58 m (1H), 6.88 m (*J* = 2 and 8 Hz, 1H), 7.00 m (1H), 7.04–7.05 m (4H), 7.30 d (3H), 7.52 t (1H), 7.60 d (2H), 8.01 s (1H); ^{13}C NMR ($CDCl_3$), δ, ppm: 23.00, 87.1, 113.5, 117.2, 122.1, 124.9, 125.1, 127.9, 128.7, 129.9, 130.2, 131.1, 141.2, 143.8, 155.00, 158.2, 185.01; MS, *m/z*: 453 (M^+, 91.10); Calculated, %: C 63.56; H 4.22; N 21.62; O 3.53; S 7.07. $C_{24}H_{19}N_7OS$. Found, %: C 63.52; H 4.20; N 21.61; O 3.52; S 7.01

5'-(4-Aminophenyl)-4'-[3-methyl-5-thioxo-1*H*-1,2,4-triazol-4(5*H*)-yl]-2',5'-diphenyl-2',4'-dihydro spiro(indolin-3,3'[1,2,4]triazol)-2-one (IX). Prepared similarly to product **VIII** from corresponding 4-aminophenyl derivative in 76% yield. Brown crystals; mp, °C: 293–296; IR (KBr): 3292, 3150, 1752, 1610; ^{1}H NMR (400 MHz, $CDCl_3$), δ, ppm: 0.9 s (3H), 4.5 d (2H), 6.43 m (2H), 6.50 m (*J* = 2 and 8 Hz, 2H), 6.58 m (1H), 7.00 m (1H), 7.04–7.05 m (4H), 7.42 d (2H), 7.52 t (1H), 8.00 s (1H); ^{13}C NMR ($CDCl_3$), δ, ppm: 23.01, 87.3, 113.5, 115.4, 117.5, 118.7, 122.1, 122.9, 125.9, 127.7, 129.7, 130.7, 141.2, 143.8, 155.00, 158.2, 185.01; MS, *m/z*: 468 (M^+, 98.58); Calculated, %: C 61.52; H 4.30; N 23.92; O 3.41; S 6.84. $C_{24}H_{20}N_8OS$. Found, %: C 61.50; H 4.29; N 23.91; O 3.42; S 6.81.

5'-(4-Methylphenyl)-4'-[3-methyl-5-thioxo-1*H*-1,2,4-triazol-4(5*H*)-yl]-2',5'-diphenyl-2',4'-dihydro spiro(indolin-3,3'[1,2,4]triazol)-2-one (X). Prepared similarly to product **VIII** from corresponding 4-methylphenyl derivative in 85% yield. White crystals; mp, °C: 285–288; IR (KBr): 3250, 3205, 1782, 1630; ^{1}H NMR (400 MHz, $CDCl_3$), δ, ppm: 0.9 s (3H), 2.35 s (3H), 6.43 m (2H), 6.57–6.59 (*J* = 2 and 8 Hz, m, 1H), 6.88 t (1H), 7.00 m (1H), 7.04–7.05 m (4H), 7.12 m (2H), 7.50–7.52 d (2H), 8.00 s (1H); ^{13}C NMR ($CDCl_3$), δ, ppm: 23.00, 24.3, 113.5, 117.2, 124.9, 125.1, 125.7, 127.3, 129.9, 130.6, 139.8, 141.5, 143.6, 155.05, 158.3, 185.00; MS, *m/z*: 467 (M^+, 91.85); Calculated, %: C 64.22; H 4.53; N 20.97; O 3.42; S 6.86. $C_{25}H_{21}N_7OS$. Found, %: C 64.21; H 4.50 N 20.96; O 3.43; S 6.84.

5'-(4-Methoxyphenyl)-4'-[3-methyl-5-thioxo-1*H*-1,2,4-triazol-4(5*H*)-yl]-2',5'-diphenyl-2',4'-dihydro spiro(indolin-3,3'[1,2,4]triazol)-2-one (XI). Prepared similarly to product **VIII** from corresponding 4-methoxyphenyl derivative in 92% yield. White crystals; mp, °C: 288–291; IR (KBr): 3310, 2992, 1783, 1640; ^{1}H NMR (400 MHz, $CDCl_3$), δ, ppm: 0.9 s (3H), 3.73 s (3H), 6.45 t (2H), 6.55–6.58 m (*J* = 2 and 8 Hz, 1H), 6.80–6.82 m (2H), 6.88 m (1H), 7.02 m (1H), 7.05–7.07m (4H), 7.50–7.52 t (3H), 8.00 s (1H); ^{13}C NMR ($CDCl_3$), δ, ppm: 22.95, 55.9, 87.00, 113.8, 114.2, 117.5, 121.3, 122.4, 124.9, 127.5, 129.7, 131.00, 141.2, 143.8, 155.1, 168.2, 186.00; MS, *m/z*: 191 (M^+, 92.16); Calculated, %: C 62.10; H 4.38; N 20.28; O 6.62; S 6.63. $C_{25}H_{21}N_2O_2S$. Found, %: C 62.12; H 4.36 N 20.20; O 6.61; S 6.60.

5'-(4-Hydroxyphenyl)-4'-[3-methyl-5-thioxo-1*H*-1,2,4-triazol-4(5*H*)-yl]-2',5'-diphenyl-2',4'-dihydro spiro(indolin-3,3'[1,2,4]triazol)-2-one (XII). Prepared similarly to product **VIII** from corresponding 4-hydroxyphenyl derivative in 75% yield. Black crystals; mp, °C: 298–301; IR (KBr): 3295, 3195, 1742, 1650; ^{1}H NMR (400 MHz, $CDCl_3$), δ, ppm: 0.9 s (3H), 5.00 s (1H), 6.43 t (2H), 6.56–6.58 m (*J* = 2 and 8 Hz, 1H), 6.79–6.81 t (2H), 6.88 t (1H), 7.00 m (1H), 7.04–7.05 m (4H), 7.40 d (2H), 7.50–7.52 d (1H), 8.01 s (1H); ^{13}C NMR ($CDCl_3$), δ, ppm: 23.00, 87.03, 113.5, 116.2, 117.2, 121.3, 122.1, 124.7, 127.6, 129.5, 130.7, 141.5, 143.8, 155.00, 159.9, 168.2, 186.1; MS, *m/z*: 469 (M^+, 98.10); Calculated, %: C 61.39; H 4.08; N 20.88; O 6.82; S 6.83. $C_{24}H_{19}N_7O_2S$. Found, %: C 61.37; H 4.02 N 20.84; O 6.80; S 6.81.

5'-(4-Iodophenyl)-4'-[3-methyl-5-thioxo-1*H*-1,2,4-triazol-4(5*H*)-yl]-2',5'-diphenyl-2',4'-dihydro spiro-(indolin-3,3'[1,2,4]triazol)-2-one (XIII). Prepared similarly to product **VIII** from corresponding 4-iodophenyl derivative in 83% yield. Yellow crystals; mp, °C: 284–286; IR (KBr): 3155, 3100, 1723, 1670;

Yields and reaction conditions of the synthesized compounds

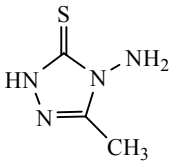
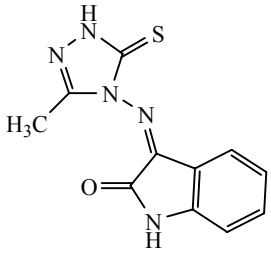
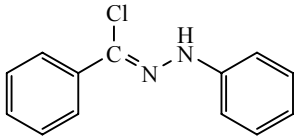
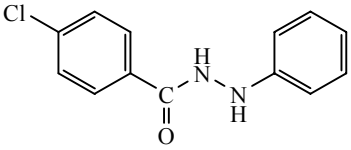
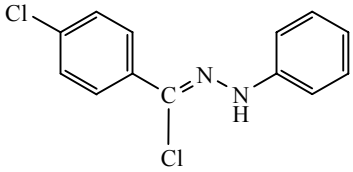
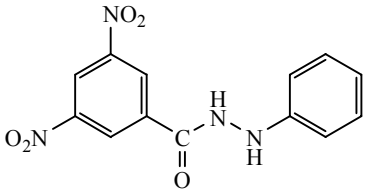
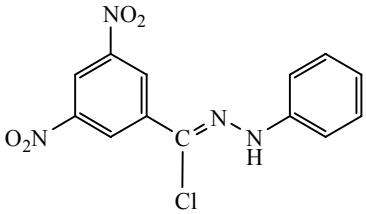
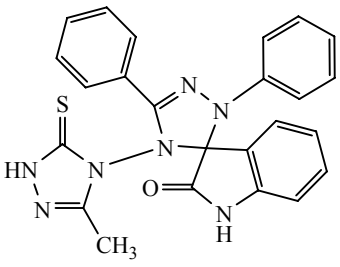
Comp. no.	Product	Color	Reaction time, h	Yield, %[a]	mp, °C
I		White	40	91	201
II		Yellow	20	89	235
III		White	4	69	240
IV		White	20	70	245
V		White	20	64	252
VI		Silver	20	72	243
VII		Silver	21	66	252
VIII		Orange	18	89	303

Table (Contd.)

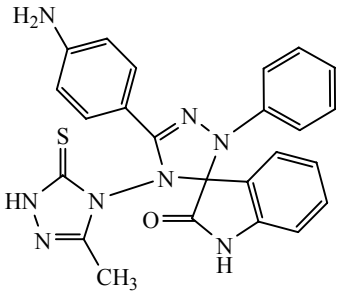
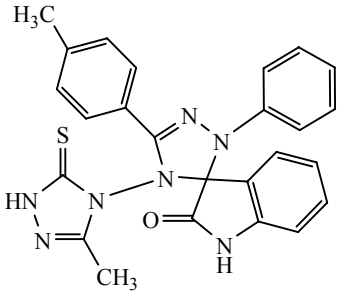
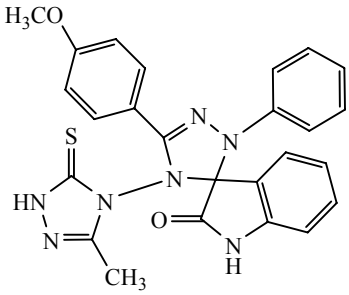
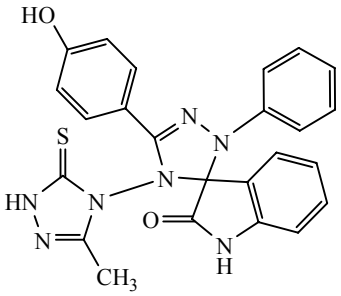
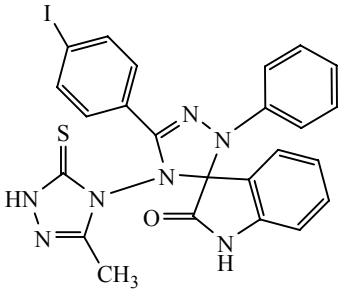
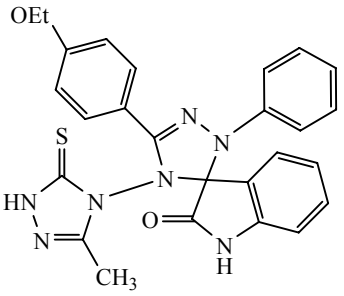
Comp. no.	Product	Color	Reaction time, h	Yield, %[a]	mp, °C
IX		Brown	20	76	295
X		White	19	85	285
XI		White	17	92	290
XII		Black	18	75	300
XIII		Yellow	20	83	285

Table (Contd.)

Comp. no.	Product	Color	Reaction time, h	Yield, %[a]	mp, °C
XIV		White	21	76	302

[a] Yields refer to isolated and purified products.

^{1}H NMR (400 MHz, $CDCl_3$), δ, ppm: 0.9 s (3H), 6.43 t (2H), 6.58 m (1H), 6.85–6.88 t (*J* = 2 and 8 Hz, 1H), 7.00–7.02 m (1H), 7.04–7.06 m (4H), 7.42 d (2H), 7.54 d (1H), 7.70 t (2H), 8.00 s (1H); ^{13}C NMR ($CDCl_3$), δ, ppm: 23.03, 87.00, 95.8, 113.4, 117.2, 122.2, 124.9, 127.6, 129.9, 130.6, 137.5, 141.2, 143.8, 155.00, 168.25, 186.00; MS, *m/z*: 579 (M^+, 92.89); Calculated, %: C 49.75; H 3.13;I, 21.90 N 16.92; O 2.76; S 5.53. $C_{24}H_{18}IN_7O_2S$. Found, %: C 49.73; H 3.09; I, 21.87 N 16.90; O 2.80; S 5.50.

5'-(4-Ethoxyphenyl)-4'-[3-methyl-5-thioxo-1*H*-1,2,4-triazol-4(5*H*)-yl]-2',5'-diphenyl-2',4'-dihydro spiro-(indolin-3,3'[1,2,4]triazol)-2-one (XIV). Prepared similarly to product **VIII** from corresponding 4-ethoxyphenyl derivative in 76% yield. White crystals; mp, °C: 300–302; IR (KBr): 3320, 3152, 1743, 1650; ^{1}H NMR (400 MHz, $CDCl_3$), δ, ppm: 0.9 s (3H), 1.33 t (3H), 3.98 m (2H), 6.43 t (2H), 6.56–6.58 m (*J* = 2 and 8 Hz, 1H), 6.80 d (2H), 6.88 t (1H), 7.01 m (1H), 7.04–7.05 m (4H), 7.50–7.52 d (3H), 8.00 s (1H); ^{13}C NMR ($CDCl_3$), δ, ppm: 14.8, 23.01, 64.7, 87.02, 113.5, 114.5, 117.2, 120.3, 122.1, 124.9, 126.7, 127.9, 129.9, 130.7, 141.5, 143.8, 155.02, 158.2, 168.2, 186.00; MS, *m/z*: 497 (M^+, 99.54); Calculated, %: C 62.76; H 4.66; N 19.71; O 6.43; S 6.44. $C_{26}H_{23}N_7O_2S$. Found, %: C 62.75; H 4.68 N 19.70; O 6.41; S 6.42.

RESULTS AND DISCUSSION

Structures of all synthesized compounds in table were confirmed on the basis of their ^{1}H NMR, ^{13}C NMR, and IR spectral data as well as MS and elemental analyses. In ^{13}C NMR spectrum, specific tertiary carbon pick appears at about 90 ppm. Total process and obtained results are shown in the table and the figure.

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