***SYNTHESIS, STRUCTURAL STABILITY CALCULATION AND ANTIBACTERIAL EVALUATION OF
NOVEL 3,5 DIPHENYLCYCLOHEX-2-EN-1-ONE DERIVATIVES.***

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Supplementary Materials

Synthesis of 5,7-diphenyl-2,6,7,7a-tetrahydrobenzo[d][1,2,3]thiadiazol-1-oxide (3).

Freshly distilled thionyl chloride (8 ml) was taken to a round bottom flask (50 ml) placed in a salt-ice bath at about -10 °C , 3,5-diphenylcyclohex-2-en-1-semicarbazone (2) (3.05 g, 0.01 mol) was added in three portions maintaining temperature between 0 °C and -10 °C. After complete addition, the reaction mixture was allowed to stand at room temperature, 25 ml of dichloromethane was added and the mixture was stirred for another 2 hrs. Excess thionyl chloride was decomposed by slowly adding sodium carbonate solution (15-20 ml) to the reaction mixture, the organic layer was separated, washed thoroughly with water, sodium bicarbonate (5%) solution and dried over anhydrous sodium sulphate. The solvent was evaporated under reduced pressure, the separated was filtered off and crystallized from water as pale green fine powder; Yield: 52%; mp 133-135°C; IR (λ max, cm^{-1}): 3462(NH), 3041($\text{CH}_{\text{arom.}}$), 2945, 2896($\text{CH}_{\text{aliphatic}}$), 1659($\text{C}=\text{N}$), 1262(C-S), 1019($\text{S}=\text{O}$); $^1\text{H-NMR}$ (DMSO-d_6), δ ppm : 7.70-7.26(m, 10H, $\text{CH}_{\text{arom.}}$), 6.47(s, 1H, $\text{CH}_{\text{oliph.}}$), 3.45-3.44 (d, 2H, CH_2), 3.02(s, 2H, 2NH, disappeared by D_2O), 2.80(t, 1H, CH-Ph.); $^{13}\text{C-NMR}$ (DMSO-d_6), δ ppm: 35.74(CH_2), 44.06(CH-Ph), 124.67($\text{CH}_{\text{oliph.}}$), 126.82, 127.13, 127.54, 128.97, 129.27, 136.58, 138.51(C of Ph rings), 144.21 ($=\text{C-S}=\text{O}$), 159.10 (C-NH), 198.80 (C-S). MS: (m/z, I %): 308(4.4) (M^+), 261 (27.8), 244 (16.9), 216 (122.0), 167.9(11.2), 143.8 (18.3), 127.9(42.4), 107.6(16.3), 91.1(100.0), 77.9(24.4), 62.0(10.2), 49.8(16.6), calc. for $\text{C}_{18}\text{H}_{16}\text{N}_2\text{OS}$ (M.wt.= 308.3).

Synthesis of 3-carbaldehyde-4,6-diphenyl-3,3a,4,5-tetrahydro-2H-indazole (4)

A mixture of DMF (2.3 ml, 0.3 mole) and POCl₃ (2.8 ml, 0.3 mole) was stirred at 0 °C for 10 min, a solution of the 3,5-diphenylcyclohex-2-en-1-semicarbazone (**2**) (3.05 g, 0.1 mol) in DMF (10 ml) was added dropwise. The reaction mixture was stirred at room temperature for 1 hrs. and then heated at 70 °C for 4 hrs. After cooling at room temperature, the mixture was neutralized with cold potassium carbonate solution. The separated solid was filtered off, washed with water, dried and crystallized from ethanol as yellow crystal; Yield: 47%; mp 169-170°C; IR (λ_{max}, cm⁻¹): 3454, 3403(NH), 3041(CH_{arom}), 2927(CH_{aliphatic}), 1682(C=O), 1573(C=C), 1262(C-S), 1107(S=O); ¹H-NMR (DMSO-d₆), δ ppm: 9.35(s, 1H, CHO), 7.72-7.24(m, 10H, CH_{arom}), 6.67(s, 1H, CH_{oliph.}), 6.46, 6.34 (s, 2H, 2NH), 5.86(m, 1H, CHPh), 3.15(m, 1H, CH-CHCHO), 3.03-3.01(d, 1H, CH-CHO), 2.77-2.81(d, 1H, CH₂); MS: (m/z, I %): 304(13.5)(M⁺), 305(27)(M⁺ + 1), 287(12.9), 245 (28.4), 231(23.9), 203(14.2), 142(28.4), 128(47.1), 115(74.2), 103(38.1), 91(100), 78 (27.1), calc. for C₂₀H₂₀N₂O (M. wt. = 304.3).

Synthesis of 5,7-diphenyl-6,7-dihydro-2(3H)-imino-1,3-benzo[d]thiazol (5).

To a solution of 3,5-diphenylcyclohex-2-en-1-one (**1**) (0.39 g, 0.0015 mol) and thiourea (0.11 g, 0.0015 mol) in isopropanol (40 ml), iodine (0.25 g, 0.002 mol) was added. The reaction mixture was refluxed for 6-8 hrs. The excess of the solvent was removed under reduced pressure, aqueous NaHCO₃ solution was added and the obtained semisolid was extracted with ether. The ether layer was washed thoroughly with water, dried over anhydrous Na₂SO₄, evaporate and the separated solid was collected as yellow crystal; yield: 33%; mp 115-118°C; IR (λ_{max}, cm⁻¹): 3383, 3279(2NH), 3173(CH_{arom.}), 2924 (CH_{aliphatic}), 1617(C=N), 1461(C=C), 1083(C-S); ¹H-NMR (DMSO-d₆), δ ppm : 7.91-7.06(m, 10H, CH_{arom.}), 6.77(s, 1H, NH, disappeared by D₂O), 6.48(s, 1H, CH_{oliph.}), 5.45(s, 1H, NH, disappeared by D₂O), 3.04-3.03 (d, 2H, CH₂), 2.86-2.82 (t, 1H, CH-ph); MS: (m/z, I %): 303 (3.1) (M⁺-1), 243 (2.6), 226 (3.5), 165(7.0), 126(14.4), 107 (17.0), 84(100), 76(2.6), calc. for C₁₉H₁₆N₂S (M. wt. = 304.4);

Synthesis of 4,6-diphenyl-4,5-dihydro-1H-indazole (7).

To a solution of compound 6 (2.95g, 0.01 mol) in ethanol (20 ml), hydrazine hydrate (0.31 ml, 0.015mol) and a catalytic amount of triethylamine were added. The reaction mixture was refluxed for 6 hrs. The excess of solvent was evaporated under reduced pressure, the obtained solid was collected, dried and crystallized from ethanol as yellow crystal; yield: 94%; mp 154-157°C; IR ($\lambda_{\max}$, cm⁻¹): 3356, 3209(NH), 3037 (CH_{arom.}), 2904(CH_{aliphatic}), 1649(C=N), 1562 (C=C), 1442(C-N); ¹H-NMR (DMSO-d₆), δ ppm : 7.51-7.35 (m, 10H, CH_{arom.}); 6.58 (s, 1H, CH_{oliph.}); 6.35 (s, 1H, NH, disappear by D₂O); 3.08-3.06 (m, 1H, CH-ph); 2.85-2.84 (d, 1H, CH=N); 2.81-2.80 (d, 1H, =CH-CHPh); 2.73-2.72 (d, 1H, =CH-C-ph); ¹³C-NMR (DMSO-d₆), δ ppm: 29.76(CH₂), 34.82(CH-ph), 125.2(CH_{oliph.}), 125.3(CH=N), 126.87, 127.64, 128.60, 128.9, 137.5, 140.3(C of Ph ring), 145.8 (CH=C-ph), 146.1(=CH-N); MS: (m/z, I %): 269(22.7)(M⁺-3), 210(31.8), 185(31.8), 160(36.4), 145(36.4), 121(59.1), 105 (54.5), 91(50.0), 64(63.6). calc. for C₁₉H₁₆N₂ (M. wt. = 272.3).

Synthesis of compounds (8) and (9).

To a solution of compound 7 (2.95 g, 0.01 mol) in ethanol (20 ml), the proper amine (ethylenediamine or cystamine hydrochloride) (0.01 mol) and triethylamine (1.4 ml, 0.01 mol) were added. The reaction mixture was refluxed for 6 hrs. The separated product was filtered off and crystallized from suitable solvent.

6,8-Diphenyl-2,3,4,6-tetrahydro-1H-benzo[1,4-e] diazepine (8)

Pale yellow crystal from ethanol; yield: 70%; mp 197-199°C; IR ($\lambda_{\max}$, cm⁻¹): 3415(NH-), 3053 CH_{arom.}), 2934(CH_{aliphatic}), 1663(C=N), 1602 (C=C), 1474(C-N); ¹H-NMR (DMSO-d₆), δ ppm : 7.71-7.20 (m, 10H, CH_{arom.}), 6.73 (s, 1H, CH=N), 6.59 (s, 1H, CH_{oliph.}), 6.50 (t, 1H, CH-CH=N), 5.61 (s, 1H, NH), 4.67 (t, 2H, CH₂-N), 3.69 (t, 2H, CH₂-NH), 3.03 (d, 1H, CH=C-ph), 2.5 (m, 1H, CH-ph), calc. for C₂₁H₂₀N₂ (M. wt = 300.4).

6,8-Diphenyl-2,3,4,6-tetrahydro-1H-benzo[1,4-f] thiazepine (9)

Yellow crystal for ethanol; yield: 48%; mp>300°C; IR (λ_{max} , cm^{-1}): 3437(NH), 3059($\text{CH}_{\text{arom.}}$), 2928($\text{CH}_{\text{aliphatic}}$), 1660 (C=N), 1452(C=C), 1122(C-S); ^1H -NMR (DMSO- d_6), δ ppm : 8.18(s,1H,NH); 7.86-7.33(m, 10H, $\text{CH}_{\text{arom.}}$); 6.92 (s,1H, $\text{CH}_{\text{oliph.}}$), 6.79 (s,1H, CH-NH), 3.11 (t,2H, $\text{CH}_2\text{-S}$), 3.02(d, 1H, $\text{CH}=\text{C-Ph}$), 2.88(d, 1H, CH-Ph), 2.74 (t, 2H, $\text{CH}_2\text{-NH}$), calc. for $\text{C}_{21}\text{H}_{19}\text{NS}$ (M. wt.= 317.44).

Synthesis of ethyl 4,6-diphenyl-4,5-dihydro-1-benzo[b]thiophen-2-yl-2-carboxylate (10).

A mixture of compound **7** (2.95 g, 0.01 mol), ethyl mercaptoacetate (1.096 ml, 0.01 mol) and sodium ethoxide solution (0.23 g of Na dissolved in 20 ml of ethanol) was refluxed for 6 hrs. The mixture was concentrated, the formed precipitate was filtered off and crystallized from ethanol as yellow crystal; yield: 47%; mp169-170 °C; IR (λ_{max} , cm^{-1}): 3052($\text{CH}_{\text{arom.}}$),2923($\text{CH}_{\text{aliphatic}}$),1661(C=O_{ester}),1450(C=C); ^1H -NMR(DMSO- d_6), δ ppm: 7.80-7.10(m, 10H, $\text{CH}_{\text{arom.}}$); 6.92(s, 1H, $\text{CH}_{\text{oliph.}}$), 6.87(d, 1H, $\text{CH}=\text{C-COOEt}$) , 4.10-4.03(q,2H, $\text{CH}_2\text{-CH}_3$), 3.61(t,1H, CH-CH ph), 3.02(d,1H, $\text{CH}=\text{C-ph}$), 2.31-2.29 (t,1H,CH-Ph), 1.22(t, 3H, CH_3),calc. for $\text{C}_{23}\text{H}_{20}\text{O}_2\text{S}$ (M. wt.= 360.4).

Synthesis of 2E-(3,5-diphenylcyclohex-2-en-1-ylidene)hydrazene-*N'*[(2R,3R,4R,5R)-2,3,4,5,6-penta- hydroxyhexylidene(12).

To a solution of compound **11** (2.62g, 0.01 mol) in dimethylformamide (20 ml), *D*(+)glucose (1.80 g,0.01 mol) and (0.1 ml) acetic acid were added. The mixture was heated under reflux on a water-bath for 2 hrs. The separated solid on cooling was filtered off and crystallized from ethanol as pale brown crystal; yield: 85%; mp 220-222°C; IR (λ_{max} , cm^{-1}): 3433(OH and NH), 3045($\text{CH}_{\text{arom.}}$), 2924($\text{CH}_{\text{aliphatic}}$),1637(C=N), 1605 (C=C), 1438(C-N); ^1H -NMR (CDCl_3), δ ppm : 7.89-7.16(m, 10H, $\text{CH}_{\text{arom.}}$),7.45(s, 5H,5 OH, disappeared by D_2O), 6.56(s, 1H, $\text{CH}_{\text{oliph.}}$), 3.08-3.07(d, 1H, $\text{CH}=\text{N}$), 2.99-2.97(d, 2H, $\text{CH}_2\text{-CHPh}$), 2.90-2.92(t, 1H, CH-OH), 2.84 -2.80 (d, 2H, $\text{CH}_2\text{-OH}$) ,3.01 -2.79 (d ,2H, CH_2), 2.79-2.71(q, 3H, CH-OH), 2.49(m, 1H, CH-Ph); ^{13}C -NMR (CDCl_3 , δ ppm : 32.49, 32.53,35.82($\text{CH}_2\text{-OH} + \text{CH}_2\text{-C}=\text{N} + \text{CH}_2\text{-C-Ph}$); 40.62, 40.25, 40.11,36.01,

(4CH-OH); 116.21(CH-Ph); 124.65(CH_{oliph.}), 125.56,126.09,126.78,127.06,128.57,128.67 (C of Ph ring); 139.76(=C-Ph); 144.63,147.37 (2 C=N), calc. for C₂₄H₂₈N₂O₅ (M. wt.= 424.4).

Synthesis of (3,5-diphenylcyclohex-2-en-1-ylidene)hydrazene-N'-(4-nitrobenzylidene) (13).

A mixture of compound 5 (2.62 g, 0.01 mol) and p-nitrobenzaldehyde (1.51 g, 0.01 mol) in glacial acetic acid (20 ml) was refluxed with stirring for 5 hrs. After cooling, the formed solid was filtered off and recrystallized from ethanol as pale yellow crystal; Yield: 62%; mp 156-158°C; IR (λ_{max} , cm⁻¹): 3443(NH), 3047(CH_{arom.}), 2927(CH_{aliphatic.}), 1610(C=N), 1597(C=C), 1518-1339(NO₂); ¹H-NMR(DMSO-d₆), δ ppm : 8.85(s,1H,NH), 8.58-8.06(m,4H,Ar-NO₂),7.70-7.26(m,10H,CH_{arom.}), 7.50(s,1H, CH=N), 6.86(s,1H,CH_{oliph.}), 3.51,3.45(d,1H,CH), 2.96-2.93(d, 2H, CH₂), 2.74-2.68(m, 1H, CH-Ph), calc. for C₂₅H₂₁N₃O₂ (M. wt.= 395).

Synthesis of 3-(3,5-diphenylcyclohexa-1,5-dienylamino)-2-(4-nitrophenyl)-1,3-thiazolidin-4-one (14).

Thioglycolic acid (0.69 ml, 0.01mol) was added drop by drop to a solution of compound 13 (3.95g, 0.01 mol) in dry benzene (30 ml). The reaction mixture was refluxed for 10 hrs and the solvent evaporated under reduced pressure. The solid was triturated with petroleum ether (60-80 °C), filtered off, dried and crystallized from CHCl₃/pet.ether (40-60 °C) as deep yellow crystal; yield: 52%; mp 216-218°C; IR (λ_{max} , cm⁻¹): 3403 (NH),3084 (CH_{arom.}),2929(CH_{aliph.}), 1718 (C=O),1595(C-N), 1517-1337(NO₂), 1111(C-S); ¹H-NMR (DMSO-d₆), δ ppm: 8.84-8.15(m,4H,CH of ph-NO₂), 7.64-7.24(m,10H,CH_{arom.}), 6.87(s, 1H, CH-Ph-NO₂),6.80(s,1H,CH_{oliph.}),4.11 (s, 2H, CH₂-C=O), 3.65,3.64(d, 1H, CH=C), 2.89(d, 2H, CH₂), 2.73(m, 1H, CH-ph), 2.50(s,1H, NH), calc. for C₂₇H₂₃N₃O₃S (M. wt.= 469.55).

Synthesis of 2-(3,5-diphenylcyclohex-2-enylidene)-N-phenylhydrazinecarbothioamide (15).

A solution of compound 11 (2.62 g, 0.01 mol) and phenylisothiocyanate (0.135 g, 0.01mol) in dry dioxane (30 ml) was refluxed for 5 hrs. The reaction mixture was concentrated under reduced pressure, the obtained solid was filtered off and recrystallized from ethanol. Yellow crystal; yield: 47%; (mp. Literature²⁴); IR (λ max, cm^{-1}): 3424 (2NH), 3036 (CH_{arom}), 2927(CH_{aliph}), 1606($\text{C}=\text{N}$); ^1H NMR (400 MHz, DMSO-d_6) δ 7.26 (s, 1H, NH), 7.24(s, 1H, NH); 7.64-7.37(m, 15H, CH_{arom}), 6.87(s, 1H, SH), 6.79(s, 1H, CH_{oliph}), 3.56(d, 1H, $\text{CH}=\text{C}-\text{NH}$), 3.18(m, 1H, CH-ph); 2.87 (d, 2H, CH_2); ^{13}C NMR(400 MHz, DMSO-d_6) δ 161.51($\text{C}-\text{SH}$), 147.20($\text{N}=\underline{\text{C}}-\text{NPh}$), 144.67($\text{C}-\text{NH}$), 125.55,126.77, 127.06,128.57, 128.67, 139.80 (C of Ph), 124.74(CH_{oliph}), 67.10($\underline{\text{CH}}=\text{C}-\text{NH}$), 40.11(CH_2), 35.81(CH-Ph), calc. for $\text{C}_{25}\text{H}_{23}\text{N}_3\text{S}$ (M. wt. = 397.5).

Synthesis of compounds (16) and (17).

A mixture of compound 11 (3.97 g, 0.01 mol) and a proper dihalocompounds (chloroacetylchloride or malonyldichloride) (0.01 mol) was refluxed in dry DMF (20 ml) for 4 hrs. After cooling, the formed solid was filtered off, and recrystallized from the proper solvent.

3-(3,5-Diphenylcyclohexa-1,5-dienylamino)-2-phenyl iminothiazolidin-4-one (16).

Orange crystal from dioxane; yield: 55%; mp 244-246°C;IR (λ max, cm^{-1}): 3359(OH), 3155(NH), 3075(CH_{arom}), 2929($\text{CH}_{\text{aliphatic}}$), 1612($\text{C}=\text{N}$), 1460($\text{C}=\text{C}$), 1407 ($\text{C}-\text{N}$); ^1H -NMR (DMSO-d_6), δ ppm : 9.03-8.95(s, 2H, OH and NH),7.95-7.27 (m,14H, CH_{arom}), 7.11(s, 1H, CH_{oliph}), 4.23(s, 1H, CH-S), 3.76(d, 1H, $\text{CH}=\text{C}$), 3.06(m,1H, CH-ph), 2.88-2.72 (d, 2H, CH_2); ^{13}C -NMR (DMSO-d_6), δ ppm: 36.55(CH_2), 41.05(CH-Ph), 43.97(CH-S), 67.09 ($\text{CH}=\text{C}$),125.15(CH_{oliph}), 125.56,126.22,126.83,127.05,127.14,127.60,128.69,128.87,129.87, 130.23 (C of Ph ring), 138.38($\underline{\text{C}}-\text{Ph}$), 143.22($\underline{\text{C}}-\text{NH}$), 158.93($\text{S}-\underline{\text{C}}=\text{N}$), 162.68($\underline{\text{C}}-\text{OH}$); MS: (m/z, %): 437(5.1)(M^+), 246(6.1), 218(2.1), 143(8.2), 105(100), 93(5.1), 77(48.0), 55(10.2), calc for $\text{C}_{27}\text{H}_{23}\text{N}_3\text{OS}$ (M. wt.= 437.5).

3-(3,5-Diphenyl-cyclohexa-1,5-dienylamino)-6-hydroxy-2-phenylimino-2,3-dihydro-[1,3] thiazine-4-one (17).

Brown powder from DMF; yield: 43%; mp 126-128°C; IR (λ_{max} , cm^{-1}): 3407 (OH), 3042 ($\text{CH}_{\text{arom.}}$), 2938 ($\text{CH}_{\text{aliphatic.}}$), 1733 (C=O), 1657 (C=N), 1493 (C=C), 1435 (C-N); $^1\text{H-NMR}$ ($\text{DMSO}-d_6$), δ ppm: 7.72-7.28 (m, 14H, $\text{CH}_{\text{arom.}}$), 6.48 (s, 1H, CH_{oliph}), 4.25 (s, 1H, CH=C-OH); 4.10 (d, 1H, CH=C-NH), 3.58 (s, 1H, NH, disappeared by D_2O), 3.45 (m, 1H, CH-Ph), 3.03 (d, 2H, CH_2), 2.82 (s, 1H, OH, disappeared by D_2O). $^{13}\text{C-NMR}$ ($\text{DMSO}-d_6$), δ ppm: 37.23 (CH_2), 42.18 (CH-ph), 59.17 (CH=C), 64.05 (CH=C-OH), 124.67 ($\text{CH}_{\text{oliph.}}$), 92.01, 126.83, 127.13, 127.54, 128.97, 129, 129.27 (C of Ph ring), 130.26 (=C-Ph), 138.51 (C=N), 144.21 (C-NH), 159.10 (C-OH), 198.80 (C=O), calc. for $\text{C}_{28}\text{H}_{23}\text{N}_3\text{O}_2\text{S}$ (M. wt.= 465.3).

Synthesis of ethyl [(2E)-2-(3,5-diphenylcyclohex-2-en-1-ylidene)hydrazine]acetate (18)

To a solution of compound **11** (2.62 g, 0.01 mol) in dioxane (20 ml), ethyl bromoacetate (1.10 ml, 0.01 mol) and triethylamine (1.4 ml, 0.01 mol) were added. The reaction mixture was refluxed for 6 hrs. After cooling, the separated solid was collected and crystallized from ethanol as yellow crystal; yield: 64%; mp 250-252°C; IR (λ_{max} , cm^{-1}): 3361, 3151 (2NH), 3037 ($\text{CH}_{\text{arom.}}$), 2927 ($\text{CH}_{\text{aliphatic.}}$), 1740 (C=O), 1598 (C=C); $^1\text{H-NMR}$ (CDCl_3), δ ppm: 7.61-6.90 (m, 10H, $\text{CH}_{\text{arom.}}$), 6.79 (s, 1H, CH_{oliph}), 4.15, 3.63 (s, 2H, 2NH, disappeared by D_2O), 3.39-3.35 (m, 1H, CH-NH), 3.15-3.09 (q, 2H, CH_2CH_3), 2.91-2.87 (d, 2H, =C- CH_2), 2.79-2.72 (m, 2H, NHCH CH_2), 2.42-2.32 (m, 1H, CH-Ph), 1.61 (s, 2H, $\text{CH}_2\text{-C=O}$), 1.38-1.35 (t, 3H, CH_3); $^{13}\text{C-NMR}$ (CDCl_3), δ ppm: 31.50 (CH_3), 32.5 (CH_2), 36.36 ($\text{CH}_2\text{-CHPh}$), 41.05 ($\text{CH}_2\text{C=O}$), 43.97 (O- CH_2), 67.09 (CH-Ph), 125.15 (=CH), 126.22 (CH-NH), 126.83, 127.14, 128.69, 128.87, 130.23, 138.38, 158.93, 143.22 (C of Ph rings), 162.68 (=C-Ph), 199.39 (C=O); MS: (m/z, %): 351 (23.8) (M^+), 351.7 (28) (M^++1), 288 (28.6), 247 (71.4), 221 (28.6), 206 (33.3), 141 (52.4), 128 (28.6), 102 (47), 91 (81.0), 63 (81.0), 57 (100), calc. for $\text{C}_{22}\text{H}_{26}\text{N}_2\text{O}_2$ (M. wt. = 350.45).

Synthesis of 2-[(2E)-2-(3,5-diphenylcyclohex-2-en-1-ylidene)hydrazine]acetohydrazide (19).

A mixture of compound **18** (3.48 g, 0.01 mol), hydrazine hydrate (0.31 ml, 0.01 mol) in ethanol (20 ml), was refluxed for 5 hrs and allowed to cool. The solid product was collected and crystallized from ethanol as yellow crystal; yield: 62%; mp 230-232°C; IR (λ_{max} , cm^{-1}): 3419-3357(NH_2), 3209(NH), 3033($\text{CH}_{\text{arom.}}$), 2898, 2910($\text{CH}_{\text{aliphatic.}}$), 1645($\text{C=O}_{\text{amidic}}$), 1568(C=C), 1492 (C-N); $^1\text{H-NMR}$ (DMSO-d_6) δ ppm: 7.65-7.26(m, 10H, $\text{CH}_{\text{arom.}}$), 6.81(s, 1H, $\text{CH}_{\text{oliph.}}$), 5.06(s, 2H, NH_2), 3.68(s, 3H, 3NH), 3.57(s, 2H, $\text{CH}_2\text{C=O}$), 3.35(m, 1H, $\underline{\text{CH-NH}}$), 3.15(m, 2H, $\underline{\text{CH}_2-\text{CH-NH}}$), 2.90-2.89(m, 1H, CH-Ph), 2.60(d, 2H, $=\text{C-CH}_2$); $^{13}\text{C-NMR}$ (DMSO-d_6), δ ppm: 31.47(CH_2), 34.97(CH_2), 39.21($\underline{\text{CH}_2-\text{C=O}}$), 113.63, 124.18, 124.53($\underline{\text{CH-cyclohexen ring}}$), 125.75, 126, 127.54, 127.63, 138.71(C of Ph ring), 143.58 ($=\underline{\text{C-Ph}}$), 181.99 (C=O), calc. for $\text{C}_{20}\text{H}_{24}\text{N}_4\text{O}$ (M. wt.= 336.4).

Synthesis of 1-(3,5-dimethyl-1H-2,3-dihydropyrazole-1-yl)-2-[(2E)-(3,5-diphenyl-cyclohexa-1-ene)hydrazinyl] ethanone (20).

A mixture of compound 19 (3.33 g, 0.01 mol), acetyl acetone (0.31 ml, 0.01 mol) and a catalytic amount of triethylamine in ethanol (20 ml), was refluxed for 7 hrs. After cooling the solid product was collected and crystallized from ethanol as deep green powder; yield: 57%; mp 262-264°C; IR (λ_{max} , cm^{-1}): 3393, 3185 (3NH), 3064($\text{CH}_{\text{arom.}}$), 2929($\text{CH}_{\text{aliphatic.}}$), 1641 ($\text{C=O}_{\text{amidic}}$), 1599(C=N), 1493(C=C), 1439(C-N); $^1\text{H-NMR}$ (CDCl_3 , δ ppm): 7.63-7.18(m, 10H, $\text{CH}_{\text{arom.}}$); 6.86(s, 1H, $\text{CH}_{\text{pyrazol ring}}$), 6.77(s, 1H, CH_{oliph}); 3.63(s, 2H, $\text{CH}_2-\text{C=O}$); 3.38-3.34(m, 1H, $\underline{\text{CH-NH}}$); 3.12(s, 2H, NH, disappeared by D_2O), 2.90-2.86(m, 2H, CH_2); 2.78-2.75(d, 1H, $=\text{C-CH}_2$); 2.43-2.31(m, 1H, $\underline{\text{CH-Ph}}$), 1.18(s, 6H, 2 CH_3), calc. for $\text{C}_{25}\text{H}_{28}\text{N}_4\text{O}$ (M. wt.= 400.5).

AS-14
proton_su DMSO {C:\nmr-data} Student 20

Current Data Parameters
NAME Aug22-2013
EXPNO 70
PROCNO 1

F2 - Acquisition Parameters
Date_ 20130822
Time_ 13.46
INSTRUM spect
PROBHD 5 mm PABBO BB/
PULPROG zg30
TD 65536
SOLVENT DMSO
NS 8
DS 2
SWH 8012.820 Hz
FIDRES 0.122266 Hz
AQ 4.0894465 sec
RG 199.04
DW 62.400 usec
DE 6.50 usec
TE 298.2 K
D1 1.00000000 sec
TD0 1

----- CHANNEL f1 -----
SF01 400.1324710 MHz
NUC1 1H
P1 12.00 usec
PLW1 22.00000000 W

F2 - Processing parameters
SI 65536
SF 400.1300032 MHz
WDW EM
SSB 0
LB 0.30 Hz
GB 0
PC 1.00

Figure S1. ^1H NMR (400 MHz, DMSO-d_6) of compound 2.

Figure S2. Mass spectra of compound 2.

Figure S3. ^1H NMR (400 MHz, DMSO-d_6) of compound 3.

Figure S4. ^{13}C NMR (400 MHz, DMSO- d_6) of compound 3.

Figure S5. Mass spectra of compound 3.

AS-18
proton_su DMSO (C:\nmr-data) Student 7

7.91
7.71
7.62
7.46
7.37
7.28
7.28
7.17
7.06
6.77
6.48
— 5.45

3.47
3.04
3.03
2.86
2.85
2.82
2.79
2.52

Current Data Parameters
NAME May20-2013
EXPR0 30
PROCNO 1

F2 - Acquisition Parameters
Date 20130525
Time 18.58
INSTRUM spect
PROBHD 5 mm JAGG0 BB/
PULPROG zg30
TD 4096
SOLVENT DMSO
NS 2
DS 2
SWH 4012.820 Hz
FIDRES 0.122264 Hz
AQ 4.0854465 sec
RG 104.74
SW 62.400 MHz
SB 4.50 MHz
TR 298.1 s
D1 1.00000000 sec
TD0 1

----- CHANNEL f1 -----
NUC1 400.1324710 MHz
P1 12.00 usec
PLM1 22.00000000 W

F2 - Processing parameters
SI 45536
SF 400.1300000 MHz
WDW EM
SSB 0
LB 0.30 Hz
GB 0
PC 1.60

Figure S6. ^1H NMR (400 MHz, DMSO- d_6) of compound **5**.

Comment: No 18

SOHAG_SC.X02 Date 04/08/11 Time 11:29:00

TIC

S=70 B=4 Bp=84 Bi=2290. RT=3.46 CT=264

S List > S=70 B=4 Pos=4 Tot=4

Figure S7. Mass spectra of compound **5**.

As-44
 PHOTON_1H DMSO (F:\nmr_data) Student 7

7.714
 7.705
 7.696
 7.658
 7.452
 7.441
 7.418
 7.373
 7.355
 7.336
 7.276
 7.258
 7.240
 6.464

3.442
 3.029
 3.011
 2.842
 2.809
 2.802
 2.769
 2.564
 2.554
 2.511

Current Data Parameters
 NAME Jul09-2012
 EXNO 31
 PROCNO 1

F2 - Acquisition Parameters
 Date 20120704
 Time 14.25
 INSTRUM spect
 PROBHD 5 mm CPMG BB/
 PULPROG zgpg
 TD 65536
 SOLVENT DMSO
 NS 16
 DS 2
 SWE 8917.870 Hz
 FIDRES 0.122266 Hz
 AQ 4.989468 sec
 RG 120
 DW 82.400 usec
 DE 6.50 umm
 TE 298.6 K
 D1 1.00000000 sec
 TDS 1

===== CHANNEL f1 =====
 SFO1 400.1374710 MHz
 NUC1 1H
 P1 12.00 usec
 PSW1 22.00000000 W

F0 - Processing parameters
 SI 65536
 SF 400.1300010 MHz
 MDW 32768
 SSB 0
 LB 0.30 Hz
 GB 0
 PC 1.00

Figure S8. ^1H NMR (400 MHz, DMSO- d_6) of compound **6**.

As-44
 C13CPD_13C (MSO (E:\nmr_data) Student 7

Current Data Parameters
 NAME Jul08-2012
 SATNO 01
 PRINNO 1

F2 - Acquisition Parameters
 Date_ 20101008
 Time 20.53
 INSTRUM spect
 PULPROG zgpg30
 TO 455.00
 SOLVENT DMSO
 NS 1024
 DS 4
 SWH 24038.441 Hz
 FIDRES 0.386788 Hz
 AQ 1.3421488 sec
 SFO 100.13
 DM 20.000 usec
 DE 0.50 usec
 TE 300.2 K
 VL 2.00000000 sec
 U1 0.00000000 sec
 FOS 1

===== CHANNEL F1 =====
 SFO1 100.6130793 MHz
 NUQ1 130
 SI 0.50 usec
 PL1 00.00000000 W

===== CHANNEL F2 =====
 SFO2 400.1314000 MHz
 NUQ2 1H
 CPDPRG2 waltz16
 PULP2 80.00 usec
 PLW2 22.00000000 W
 PLW12 0.41001001 W
 PLW13 0.33284000 W

F2 - Processing parameters
 SI 32768
 SF 100.6131800 MHz
 NS 0
 DS 0
 LA 1.00 Hz
 GB 0
 PC 1.40

Figure S9. ^{13}C NMR (400 MHz, DMSO- d_6) of compound **6**.

Comment: No 44

SOHAG_SC.X04 Date 01/01/85 Time 11:51:40

TIC

S=54 Bp=60 Bi=2.00 RT=2.66 CT=181

S List > S=54 B 0 Pos=1 Tot=1

Figure S10. Mass spectra of compound **6**

Figure S11. ^1H NMR (400 MHz, DMSO-d_6) of compound **7**.

Figure S12. ^{13}C NMR (400 MHz, DMSO-d_6) of compound 7.

Figure S13. Mass spectra of compound **7**.

Figure S14. ^1H NMR (400 MHz, CDCl_3) of compound **12**.

Figure S15. ^{13}C NMR (400 MHz, CCL_3) of compound **12**.

Figure S16. ^1H NMR (400 MHz, CCl_3) of compound **15**.

AS-87
c13_su CDC13 {C:\nmr-data} Student 16

Current Data Parameters
NAME Jul27-2013
EXPNO 21
PROCNO 1

F2 - Acquisition Parameters
Date_ 20130727
Time 8.58
INSTRUM spect
PROBHD 5 mm PABBO BB/
PULPROG zgpg30
TD 65536
SOLVENT CDC13
NS 512
DS 4
SWH 24038.461 Hz
FIDRES 0.366798 Hz
AQ 1.3631488 sec
RG 100.43
DW 20.800 usec
DE 6.50 usec
TE 298.1 K
D1 2.00000000 sec
D11 0.03000000 sec
TD0 1

===== CHANNEL f1 =====
SFO1 100.6238364 MHz
NUC1 13C
P1 9.50 usec
PLW1 56.00000000 W

===== CHANNEL f2 =====
SFO2 400.1316005 MHz
NUC2 1H
CPDPRG[2] waltz16
PCPD2 90.00 usec
PLW2 22.00000000 W
PLW12 0.41091001 W
PLW13 0.33284000 W

F2 - Processing parameters
SI 32768
SF 100.6127690 MHz
WDW EM
SSB 0
LB 6.00 Hz
GB 0
PC 1.40

Figure S17. ^{13}C NMR (400 MHz, CCl_3) of compound 15.

Figure S18. ^1H NMR (400 MHz, CCl_3) of compound 18.

AS-118
 c13_nu CDCl3 (C:\nmr-data) Student 3

Current Data Parameters
 NAME Jun13-2013
 EXPRD 33
 PROCNO 1

F2 - Acquisition Parameters
 Date_ 20130613
 Time 14.38
 INSTRUM spect
 PROBRD 5 mm TARGO DR/
 PULPROG zgpg30
 VD 455.94
 SOLVENT CDCl3
 NS 512
 DS 4
 SWH 24028.401 MHz
 FIDRES 0.348798 Hz
 AQ 1.34031488 sec
 RG 100.43
 CW 20.800 usec
 DE 8.50 usec
 TE 298.2 K
 O1 2.00000000 sec
 D11 0.03000000 sec
 TDO 1

===== CHANNEL F1 =====
 NUC1 100.6258364 MHz
 MUX1 130
 P1 8.50 usec
 PLW1 34.00000000 W

===== CHANNEL F2 =====
 NUC2 400.1314000 MHz
 MUX2 18
 CPDPRG2 waltz16
 PCDP2 90.00 usec
 PLW2 22.00000000 W
 PLW12 0.41091001 W
 PLW13 0.33284000 W

F2 - Processing parameters
 SI 32768
 SF 100.6127400 MHz
 WHW 88
 SSB 0
 LB 0.00 Hz
 GB 0
 PC 1.40

Figure S19. ^{13}C NMR (400 MHz, CCL_3) of compound 18.

Figure S20. Mass spectra of compound **18**.