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Regioselectivity of Nitrilimines 1,3-Dipolar Cycloaddition: Novel Synthesis of Spiro[4,4]nona-2,8-dien-6-one Derivatives

Omar A. Miqdad,[1] Nada M. Abunada,[1] and Hamdi M. Hassaneen[2]

[1]*Department of Chemistry, Faculty of Applied Sciences, Al-Aqsa University, 76888 Gaza, Palestine*

[2]*Department of Chemistry, Faculty of Science, Cairo University, Giza, Egypt*

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ABSTRACT: *Regioselective 1,3-dipolar cycloaddition of nitrilimines (generated in situ from dehydrohalogenation of the corresponding hydrazonoyl halides by the action of triethylamine) with 4-arylidene-1-aryl-2-phenyl-1H-imidazol-5(4H)-one* **3** *afforded the corresponding spiro[4,4]nona-2,8-dien-6-one* **4**. *The reaction was carried out in dry benzene under reflux temperature. Refluxing in acetic acid,* **4a** *was converted to its respective N-phenylpyrazole-5-carboxamide* **8**. *The structures of prepared compounds were established by elemental analyses and spectral data (IR, MS, ^{1}H, and ^{13}C NMR).* © 2011 Wiley Periodicals, Inc. Heteroatom Chem 22:131–136, 2011; View this article online at wileyonlinelibrary.com. DOI 10.1002/hc.20666

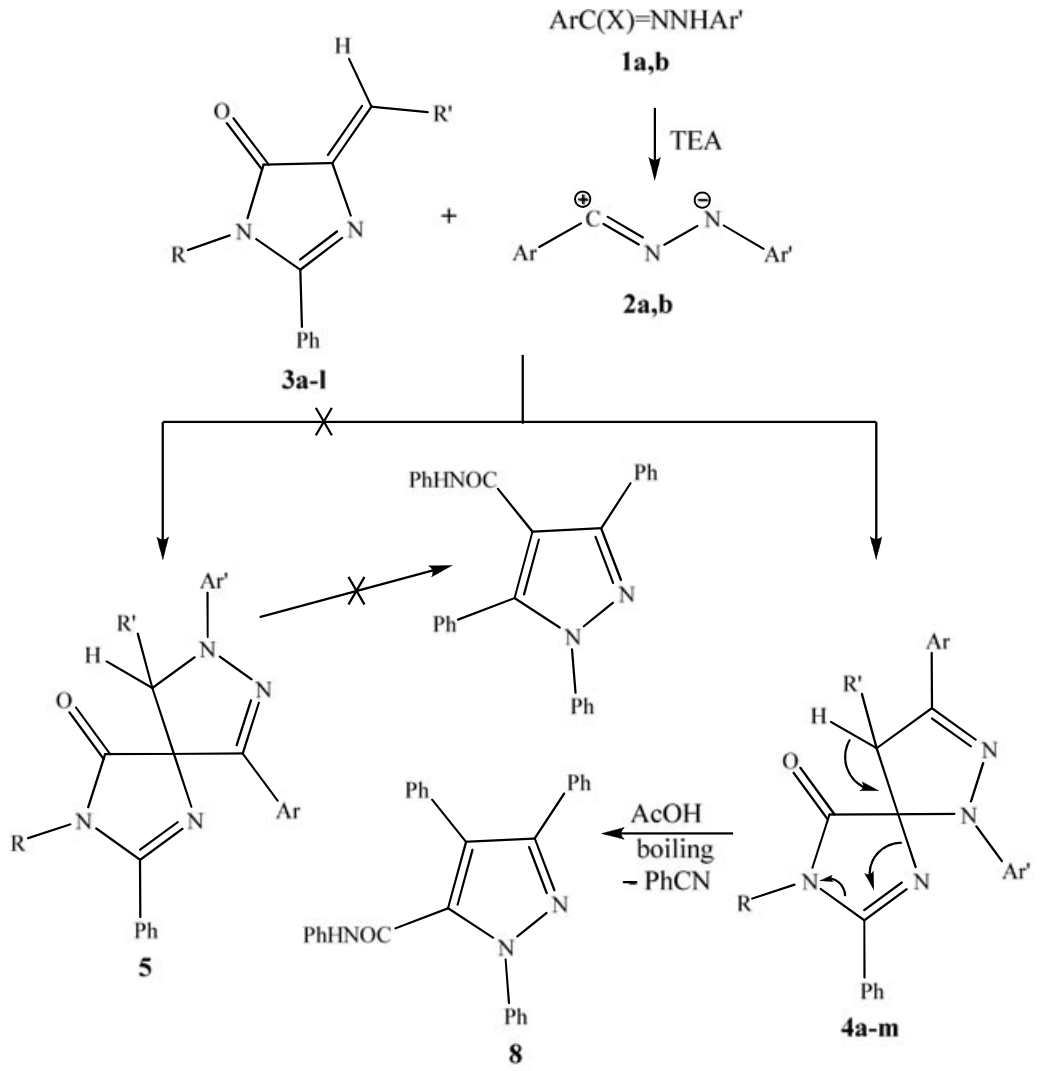
INTRODUCTION

The imidazolinone derivatives are associated with a wide range of therapeutic activities such as anticonvulsant [1] and potent CNS depressant [2,3]. Recently, some new imidazolinone derivatives have been reported as antimicrobial [4,5] and L-DOPA prodrugs in the treatment of Parkinson's disease [6]. Some workers have recognized 5-imidazolone as having anticancer activity [5]. Also, pyrazoline derivatives have been found to possess antifungal [7,8], antidepressant [9–12], anticonvulsant [11,12], anti-inflammatory [13], antibacterial [8,14], and antitumor [15] activities. Furthermore, many spirocompounds were found to possess antimicrobial [16], antitumor [17] activities and display modest inhibition of human peptidyl prolyl cis/trans isomerase Pin1 [18]. Moreover, spirocompounds were found to exhibit vasodilation activities [19]. The most developed procedure for construction of spirocompounds depends mainly on 1,3-dipolar cycloadditions to exocyclic double bonds [20–22]. On continuation of our research on the chemistry of hydrazonoyl halides, which are the major precursors for nitrilimines [8,21,23], it is intended to explore the 1,3-dipolar cycloaddition reaction of nitrilimines with exocyclic double bond of 2-phenyl-4-arylmethylene-2-imidazolin-5-one that have been unreported hitherto, on the one hand, to synthesize the corresponding novel spiro-heterocyclic ring systems with anticipated biological activities and, on the other hand, to study the regioselectivity of the reaction.

RESULTS AND DISCUSSION

The reaction of the nitrilimines **2**, generated in situ by the action of triethylamine on hydrazonoyl halides **1a,b** in dry benzene at reflux temperature, with 4-arylidene-1-aryl-2-phenyl-1H-imidazol-5(4*H*)-one **3**, gave in each case, one regioisomer whose elemental analysis and mass spectral data were compatible with the molecular formula of 1,3,4,7,8-pentaryl-1,2,7,9-tetrazaspiro[4,4]nona-2,8-dien-6-one **4** or 1,3,4,7,8-pentaryl-2,3,7,9-tetrazaspiro[4,4]nona-1,8-dien-6-one **5** (Scheme 1).

Correspondence to: Nada M. Abunada; e-mail: nadanadannrs@yahoo.com.



3a, R, R' = Ph
3b, R = 4-$CH_3C_6H_4$, R' = Ph
3c, R = 4-ClC_6H_4, R' = Ph
3d, R = 4-BrC_6H_4, R' = Ph
3e, R = Ph, R' = 4-$CH_3C_6H_4$
3f, R = 4-ClC_6H_4, R' = 4-$CH_3C_6H_4$
3g, R = Ph, R' = 4-$OCH_3C_6H_4$
3h, R = 4-ClC_6H_4, R' = 4-$OCH_3C_6H_4$
3i, R = 4-BrC_6H_4, R' = 4-$OCH_3C_6H_4$
3j, R = Ph, R' = 4-ClC_6H_4
3k, R = 4-$CH_3C_6H_4$, R' = 4-ClC_6H_4
3l, R = 4-ClC_6H_4, R'

1a, **2a**; Ar, Ar' = Ph
1b, **2b**; Ar = 2,4-$Cl_2C_6H_3$, Ar' = 4-$NO_2C_6H_4$

4a, R, R', Ar, Ar' = Ph
4b, R = 4-$CH_3C_6H_4$, R', Ar, Ar' = Ph
4c, R = 4-ClC_6H_4, R', Ar, Ar' = Ph
4d, R = 4-BrC_6H_4, R', Ar, Ar' = Ph
4e, R, Ar, Ar' = Ph, R' = 4-$CH_3C_6H_4$
4f, R = 4-ClC_6H_4, R' = 4-$CH_3C_6H_4$, Ar, Ar' = Ph
4g, R, Ar, Ar' = Ph, R' = 4-$OCH_3C_6H_4$
4h, R = 4-ClC_6H_4, R' = 4-$OCH_3C_6H_4$, Ar, Ar' = Ph
4i, R = 4-BrC_6H_4, R' = 4-$OCH_3C_6H_4$, Ar, Ar' = Ph
4j, R, Ar, Ar' = Ph, R' = 4-ClC_6H_4
4k, R = 4-$CH_3C_6H_4$, R' = 4-ClC_6H_4, Ar, Ar' = Ph
4l, R = 4-ClC_6H_4, R' = 4-ClC_6H_4, Ar, Ar' = Ph
4m, R, R' = Ph, Ar = 2,4-$Cl_2C_6H_3$, Ar' = 4-$NO_2C_6H_5$

SCHEME 1

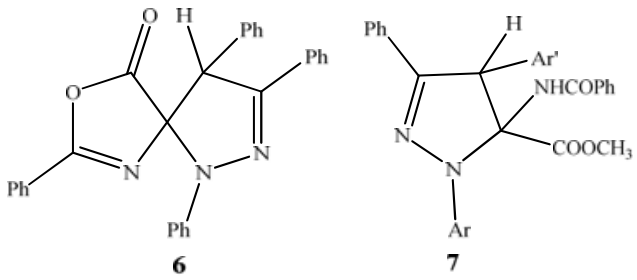


FIGURE 1

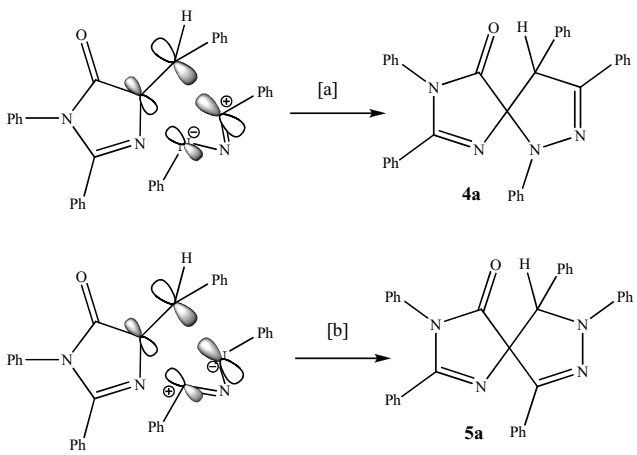


FIGURE 2 (a) Magnitude of the coefficients is estimated from the calculation based on ab initio molecular orbital method [29,30]. (b) Magnitude of the coefficients is estimated from the calculation based on CNDO/2 molecular orbital method [28].

The structural assignment of the isolated spiropyrazoline was achieved on the basis of their spectral data and chemical transformation. The IR spectra of the products isolated showed carbonyl absorption bands in the region 1751.2–1759.0 cm^{-1}. The ^1H NMR spectra showed the pyrazoline ring proton resonates at δ 5.46–5.60 ppm. Comparison of this chemical shift of the 4-CH proton of the pyrazoline ring residue of **4** with those of the related compounds **6** and **7** showed similarity between the proton chemical shifts for the 4-CH group of **4** with those reported for compounds **6** (δ 5.20–5.30) and **7** (δ 5.46–5.77) (Fig. 1) [24,25], such similarity of the chemical shifts give a good evidence that the isolated regioisomer for the cycloadducts studied is **4**.

If the cycloaddition took place in the opposite direction to give **5**, the chemical shift of 5-CH proton would shift downfield due to the deshielding effect of the more electronegative nitrogen atom attached directly to the carbon atom bearing the hydrogen one. It has been shown that the chemical shift of the 5-CH proton of pyrazoline is about δ 5.9–6.0 ppm [21,26]. The ^{13}C NMR spectra of **4** add further evidence for the proposed structure, revealing the presence of 4-CH and spiro-C atoms at δ 62.747–64.082 and 90.961–91.140 ppm, respectively. The observed regioselectivity can be explained on the basis of FMO theory. It is fully in agreement with the prediction [27,28] that dipole–HOMO/dipolarophile–LUMO interaction would be favored since the dipolarophilic exocyclic double bond of **3** is electron deficient, as it conjugated with two electron-withdrawing groups, the C=O and C=N groups of the imidazolinone ring moiety, which cause its LUMO to be relatively low in energy. Since the coefficient of nitrogen atom in HOMO of dipole is much larger than that of carbon atom, the **5** isomer will be favored (Fig. 2). But on the basis of ab initio molecular orbital calculations, it is argued that in the nonplanar most stable conformation of H—C ≡ N^+—N^-—H the biggest HOMO coefficient is to be found on the carbon atom [29] and this is also the case in diphenylnitrilimine [30]. Therefore, the formation of **4** can be explained on the basis of dipole–HOMO control by assuming that the coefficient of carbon atom in HOMO of diphenylnitrilimine is much larger than that of nitrogen atom (Fig. 2).

An unequivocal evidence for the production of **4** was achieved on the basis of their chemical transformations. Thus, when the spiropyrazole **4a**, taken as a typical example of the series studied, was refluxed in acetic acid it gave the respective *N*,1,3,4-tetraphenyl-1*H*-pyrazole-5-carboxamide **8a** with elimination of benzonitrile (Scheme 1). The structure of compound **8a** was established on the basis of elemental and spectral data. Its IR spectrum reveals absorption bands at ν 3313.5 and 1651.0 cm^{-1} corresponding to NH and C=O of the amide moiety. ^1H NMR spectrum exhibits broad characteristic signal at δ 10.76 ppm for NH and was D_2O-exchangeable. Fortunately, compound **8a** was previously synthesized through a different route [31] and is found to be identical in all respects with our compound.

It is worthy to mention that all attempts to isolate products from the reaction of *N*-phenyl-2-oxopropanehydrazonoyl chloride **9** [32], ethyl *N*-(phenylhydrazono)chloroacetate **10** [33], *N*-phenyl-C-phenylaminocarbonylmethanohydrazonoyl chloride **11** [34], and *N*,*N*′-diphenylethane(bis-hydrazonoyl chloride) **12** [35] (Fig. 3) with **3** failed. This finding can be attributed to the presence of electron-withdrawing group in the nitrilimines of **9**–**12**, which will lower both dipole orbitals in energy,

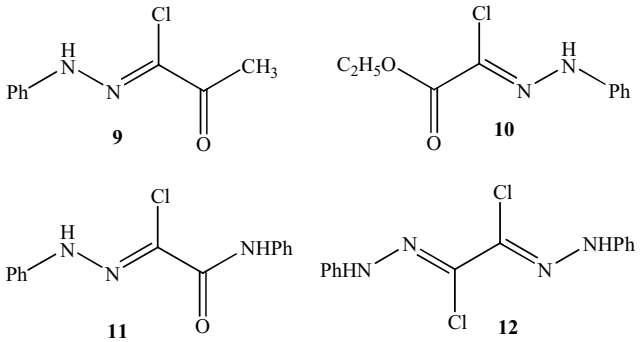


FIGURE 3 Unreacted hydrazonoyl halides.

thought to cause higher energy barrier between HOMO of nitrilimine and LUMO of dienophile to the extent of preventing their interaction to give product.

EXPERIMENTAL

Melting points were measured on electrothermal melting point apparatus and are uncorrected. The infrared spectra were recorded in potassium bromide disks on a Pye Unicam SP 3-300 and Shimadzu FT-IR 8101 PC infrared spectrophotometer. The ^{1}H NMR (200 MHz) and ^{13}C NMR (50 MHz) spectra were recorded in DMSO-d_6 on a Varian Mercury VX 200 NMR using TMS as the internal reference. Mass spectra were measured on a GCMS-QP 1000 EX spectrometer at 70 eV. Elemental analyses were carried out at the Microanalytical Centre of Cairo University, Giza, Egypt. Hydrazonoyl halides **1a** [36], **1b** [37], and the imidazolinone **3** [38,39] were prepared according to the method reported in the literature.

Reaction of **3a-l** *with Hydrazonoyl Halides* **1a,b***(General Procedure)*

To a solution of the appropriate imidazolinone **3a-l** (5 mmol) and hydrazonoyl chloride **1a** and hydrazonoyl bromide **1b** (5 mmol) in dry benzene (50 mL), triethylamine (0.7 mL, 5 mmol) was added and the mixture was refluxed for 6 h. The reaction mixture was filtered while hot to remove triethylamine hydrochloride and evaporated under reduced pressure till dryness, and the remaining residue was triturated with boiled methanol (10 mL). The separated solid material was filtered and crystallized from a suitable solvent.

1,3,4,7,8-Pentaphenyl-1,2,7,9-tetrazaspiro[4,4]nona-2,8-dien-6-one **4a**. Pale yellow crystals, yield 84%; mp 196–198°C (dioxane); IR (KBr) ν_{max}/cm^{-1} 1751.2 (C=O), 1596.9 (C=N); ^{1}H NMR (DMSO-d_6) δ 5.556 (s, 1H, 4-CH), 6.840–7.681 (m, 25H, ArHs); MS *m/z* (%): 519 (M^+ + 1, 97.8), 491 (55%), 399 (100), 193 (30), 180 (60), 104 (24), 90 (70), 77 (60). Anal. Calcd for $C_{35}H_{26}N_4O$: C, 81.06; H, 5.05; N, 10.80. Found: C, 81.10; H, 5.08; N, 10.81%.

7-(4-Methylphenyl)-1,3,4,8-tetraphenyl-1,2,7,9-tetrazaspiro[4,4]nona-2,8-dien-6-one **4b**. Pale yellow crystals, yield 81%; mp 239–241°C (tetrahydrofuran); IR (KBr) ν_{max}/cm^{-1} 1755.1 (C=O), 1596.9 (C=N); ^{1}H NMR (DMSO-d_6) δ 2.295 (s, 3H, CH_3), 5.557 (s, 1H, 4-CH), 6.841–7.626 (m, 24H, ArHs); ^{13}C NMR (DMSO-d_6) δ 179.818, 160.657, 149.479, 143.410, 138.367, 132.534, 131.657, 131.582, 131.355, 130.871, 130.816, 130.231, 129.948, 129.246, 128.937, 128.372, 128.105, 127.964, 127.747, 126.923, 126.774, 121.334, 114.807, 91.140, 64.082, 20.602; MS *m/z* (%): 533 (M^+ + 1, 46), 505 (32), 399 (100), 194 (70), 104 (30), 91 (82), 77 (22); Anal. Calcd for $C_{36}H_{28}N_4O$: C, 81.18; H, 5.30; N, 10.52. Found: C, 81.18; H, 5.29; N, 10.52%.

7-(4-Chlorophenyl)-1,3,4,8-tetraphenyl-1,2,7,9-tetraazaspiro[4,4]nona-2,8-dien-6-one **4c**. Pale yellow crystals, yield 84%; mp 221–223 °C (tetrahydrofuran); IR (KBr) ν_{max}/cm^{-1} 1759.0 (C=O), 1600.8 (C=N); ^{1}H NMR (DMSO-d_6) δ 5.560 (s, 1H, 4-CH), 6.842–7.625 (m, 24H, ArHs); MS *m/z* (%): 554 (M^+ + 1, 100), 525 (34), 399 (95), 214 (35), 193 (19), 104 (29), 90 (45), 77 (18); Anal. Calcd for $C_{35}H_{25}ClN_4O$: C, 76.01; H, 4.56; N, 10.13; Cl, 6.41. Found: C, 76.01; H, 4.59; N, 10.11; Cl, 6.39%.

7-(4-Bromophenyl)-1,3,4,8-tetraphenyl-1,2,7,9-tetraazaspiro[4,4]nona-2,8-dien-6-one **4d**. Yellow crystals, yield 81%; mp 226–228°C (benzene); IR (KBr) ν_{max}/cm^{-1} 1755.1 (C=O), 1596.9 (C=N); ^{1}H NMR (DMSO-d_6) δ 5.492 (s, 1H, 4-CH), 6.839–7.633 (m, 24H, ArHs); MS *m/z* (%): 598 (M^+ + 1, 100), 571 (26), 399 (81), 260 (22), 193 (16), 104 (16), 90 (40), 76 (20); Anal. Calcd. for $C_{35}H_{25}BrN_4O$: C, 70.36; H, 4.22; N, 9.38; Br, 13.37. Found: C, 70.39; H, 4.21; N, 9.38; Br, 13.38%.

4-(4-Methylphenyl)-1,3,7,8-tetraphenyl-1,2,7,9-tetraazaspiro[4,4]nona-2,8-dien-6-one **4e**. Pale yellow crystals, yield 67%; mp 234–236°C (benzene); IR (KBr) ν_{max}/cm^{-1} 1755.1 (C=O), 1596.9 (C=N); ^{1}H NMR (DMSO-d_6) δ 2.291 (s, 3H, CH_3), 5.558 (s, 1H, 4-CH), 6.839–7.624 (m, 24H, ArHs); MS *m/z* (%): 533 (M^+ + 1, 100), 505 (61), 413 (99), 192 (15), 180 (75), 104 (14), 91 (42), 77 (45); Anal. Calcd for $C_{36}H_{28}N_4O$:

C, 81.18; H, 5.30; N, 10.52. Found: C, 81.20; H, 5.28; N, 10.52%.

7-(4-Chlorophenyl)-4-(4-methylphenyl)-1,3,8-triphenyl-1,2,7,9-tetraazaspiro[4,4]nona-2,8-dien-6-one **4f**. Pale yellow crystals, yield 85%; mp 240–242°C (benzene); IR (KBr) ν_{max}/cm^{-1} 1755.1 (C=O), 1600.8 (C=N); ^1H NMR (DMSO-d_6) δ 3.254 (s, 3H, CH_3), 5.493 (s, 1H, 4-CH), 6.853–7.558 (m, 23H, ArHs); ^{13}C NMR (DMSO-d_6) δ 179.467, 160.038, 149.640, 143.387, 137.264, 133.244, 132.523, 131.523, 130.856, 129.841, 129.536, 129.253, 128.914, 128.693, 128.662, 128.357, 128.235, 128.193, 128.056, 127.461, 126.766, 121.342, 114.841, 91.274, 63.838, 20.659; MS *m/z* (%): 567 (M^+, 39), 539 (22), 413 (100), 214 (78), 193 (28), 104 (63), 91 (91), 77 (58); Anal. Calcd for $C_{36}H_{27}ClN_4O$: C, 76.25; H, 4.80; N, 9.88; Cl, 6.25. Found: C, 76.19; H, 4.80; N, 9.89; Cl, 6.26%.

4-(4-Methoxylphenyl)-1,3,7,8-tetraphenyl-1,2,7,9-tetraazaspiro[4, 4]nona - 2, 8 - dien - 6 - one **4g**. Pale yellow crystals, yield 52%; mp 236–238°C (benzene); IR (KBr) ν_{max}/cm^{-1} 1755.1(C=O), 1596.9 (C=N); ^1H NMR (DMSO-d_6) δ 3.717 (s, 3H, OCH_3), 5.466 (s, 1H, 4-CH), 6.863–7.679 (m, 24H, ArHs); MS m/z (%): 549 ($M^+ + 1$, 88), 521 (48), 429 (62), 180 (96), 104 (32), 91 (95), 77 (100); Anal. Calcd for $C_{36}H_{28}N_4O_2$: C, 78.81; H, 5.14; N, 10.21. Found: C, 78.82; H, 5.12; N, 10.20%.

7-(4-Chlorophenyl)-4-(4-methoxylphenyl)-1,3,8-triphenyl-1,2,7,9-tetraazaspiro[4,4]nona-2,8-dien-6-one **4h**. Pale yellow crystals, yield 78%; mp 254–256°C (dioxane); IR (KBr) ν_{max}/cm^{-1} 1759.0 (C=O), 1600.8 (C=N); ^1H NMR (DMSO-d_6) δ 3.714 (s, 3H, OCH_3), 5.465 (s, 1H, 4-CH), 6.868–7.688 (m, 23H, ArHs); MS *m/z* (%): 583 (M^+, 99), 555 (51), 429 (100), 214 (84), 193 (22), 104 (29), 91 (80), 77 (32); Anal. Calcd for $C_{36}H_{27}ClN_4O_2$: C, 74.16; H, 4.67; N, 9.61; Cl, 6.08. Found: C, 74.18; H, 4.69; N, 9.61; Cl, 6.09%.

7-(4-Bromophenyl)-4-(4-methoxylphenyl)-1,3,8-triphenyl-1,2,7,9-tetraazaspiro[4,4]nona-2,8-dien-6-one **4i**. Pale yellow crystals, yield 65%; mp 241–243°C (benzene); IR (KBr) ν_{max}/cm^{-1} 1751.2 (C=O), 1600.8 (C=N); ^1H NMR (DMSO-d_6) δ 3.715 (s, 3H, OCH_3), 5.464 (s, 1H, 4-CH), 6.868–7.697 (m, 23H, ArHs); MS *m/z* (%): 628 ($M^+ + 1$, 47), 600 (29), 429 (54), 260 (41), 104 (50), 91 (100), 76 (83); Anal. Calcd for $C_{36}H_{27}BrN_4O_2$: C, 68.90; H, 4.34; N, 8.93; Br, 12.73. Found: C, 68.87; H, 4.33; N, 8.91; Br, 12.71%.

4-(4-Chlorophenyl)-1,3,7,8-tetraphenyl-1,2,7,9-tetraazaspiro[4, 4]nona - 2, 8 - dien - 6 - one **4j**. Pale yellow crystals, yield 72%; mp 186–188 °C (benzene); IR (KBr) ν_{max}/cm^{-1} 1755.1 (C=O), 1596.9 (C=N); ^1H NMR (DMSO-d_6) δ 5.532 (s, 1H, 4-CH), 6.867–7.689 (m, 24H, ArHs); MS *m/z* (%): 553 (M^+, 100), 525 (56), 433 (88), 397 (39), 180 (93), 104 (35), 91 (88), 77 (97); Anal. Calcd for $C_{35}H_{25}ClN_4O$: C, 76.01; H, 4.56; N, 10.13; Cl, 6.41. Found: C, 76.02; H, 4.58; N, 10.12; Cl, 6.39%.

4-(4-Chlorophenyl)-7-(4-methylphenyl)-1,3,8-triphenyl-1,2,7,9-tetraazaspiro[4,4]nona-2,8-dien-6-one **4k**. Yellow crystals, yield 74%; mp 228–230°C (dioxane); IR (KBr) ν_{max}/cm^{-1} 1755.1 (C=O), 1596.9 (C=N); ^1H NMR (DMSO-d_6) δ 2.321 (s, 3H, CH_3), 5.601 (s, 1H, 4-CH), 6.856–7.599 (m, 23H, ArHs); ^{13}C NMR (DMSO-d_6) δ 179.635, 161.057, 149.365, 143.364, 138.504, 132.645, 131.864, 131.790, 131.561, 130.806, 130.631, 130.032, 129.284, 129.089, 128.510, 128.250, 128.159, 127.976, 127.682, 126.907, 126.785, 121.559, 115.028, 90.961, 62.747, 20.640; MS m/z (%): 567 (M^+, 36), 539 (27), 433 (62), 194 (70), 104 (32), 91 (100), 77 (22); Anal. Calcd. for $C_{36}H_{27}ClN_4O$: C, 76.25; H, 4.80; N, 9.88; Cl, 6.25. Found: C, 76.23; H, 4.81; N, 9.90; Cl, 6.26%.

4,7-Di-(4-chlorophenyl)-1,3,8-triphenyl-1,2,7,9-tetraazaspiro[4, 4]nona - 2, 8 - dien - 6 - one **4l**. Yellow crystals, yield 76%; mp 224–226°C (dioxane); IR (KBr) ν_{max}/cm^{-1} 1759.0 (C=O), 1596.9 (C=N); ^1H NMR (DMSO-d_6) δ 5.594 (s, 1H, 4-CH), 6.865–7.627 (m, 23H, ArHs); MS *m/z* (%): 587 (M^+, 65), 559 (28), 434 (61), 193 (68), 104 (35), 91 (100), 77 (31); Anal. Calcd for $C_{35}H_{24}Cl_2N_4O$: C, 71.55; H, 4.12; N, 9.54; Cl, 12.07. Found: C, 71.58; H, 4.11; N, 9.52; Cl, 12.09%.

3-(2,4-Dichlorophenyl)-1-(4-nitrophenyl)-4,7,8-triphenyl-1,2,7,9-tetraazaspiro[4,4]nona-2,8-dien-6-one **4m**. Yellow crystals, yield 66%; mp 214–216°C (dioxane); IR (KBr) ν_{max}/cm^{-1} 1751.2 (C=O), 1581.5 (C=N); ^1H NMR (DMSO-d_6) δ 5.600 (s, 1H, 4-CH), 6.866–7.821 (m, 22H, ArHs); MS *m/z* (%): 633 ($M^+ + 1$, 37), 605 (56), 513 (30), 191 (15), 180 (81), 105 (24), 89 (37), 77 (100); Anal. Calcd for $C_{35}H_{23}Cl_2N_5O_3$: C, 66.46; H, 3.67; N, 11.07; Cl, 11.21. Found: C, 66.45; H, 3.69; N, 11.09; Cl, 11.23%.

Synthesis of N,1,3,4-Tetraphenyl-1H-pyrazole-5-carboxamide **8a**

A mixture of compound **4a** (0.5 g) and acetic acid (20 mL) was refluxed for 2 h. The mixture was left to cool, and then the precipitated solid was filtered off and recrystallized from ethanol.

N,1,3,4-tetraphenyl-1H-pyrazole-5-carboxamide **8a**. White, yield 83%; mp 243–244°C (lit. mp 240–242°C) [31]; IR (KBr) ν_{max}/cm^{-1} 3313.5 (NH), 1651.0 (C=O amide), 1596.9 (C=N); ^1H NMR (DMSO-d_6) δ 7.06–7.72 (m, 21H, ArH), 10.76 (br, s, 1H, NH, D_2O-exchangeable); ^{13}C NMR (DMSO-d_6) δ 119.1, 119.8, 122.8, 124.4, 124.5, 127.7, 127.9, 128.0, 128.3, 128.6, 128.7, 128.9, 129.2, 130.1, 131.2, 131.9, 132.5, 137.8, 138.0, 139.1, 149.0, 158.9, 187.1; MS *m/z* (%): 416 (M + 1^+, 56), 323 (100), 190 (14), 165 (16), 89 (69), 77 (31); Anal. Calcd. for $C_{28}H_{21}N_3O$: C, 80.94; H, 5.09; N, 10.11. Found: C, 80.92; H, 5.10; N, 10.12%.

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