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Reactions of Aliphatic Diazo Compounds: IV. Reaction of Diphenyldiazomethane with Substituted Imides of Maleic and Itaconic Acids

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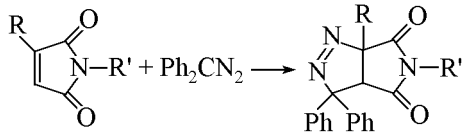
Abstract— Diphenyldiazomethane regioselectively adds to 2-R-substituted maleimides to yield 1-pyrazoline derivatives, 1-R-7-aryl-6,8-dioxo-4,4-diphenyl-2,3,7-triazabicyclo[3.3.0]oct-2-enes that on heating liberate nitrogen to afford substituted 3-azabicyclo[3.1.0]hexanes. To the *N*-arylsubstituted imides of itaconic acid the diphenyldiazomethane adds to furnish 5-aryl-4,6-dioxo-1,1-diphenyl-5-azaspiro[2.4]heptanes.

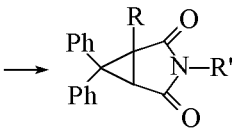
Reactions of diazoalkanes with unsaturated compounds containing an activated multiple bond result in 1- or 2-pyrazolines depending on the structure of the diazo compound. It was shown formerly that alkyl diazoacetates added to maleimides regioselectively to afford 2-pyrazolines [2, 3]. The reaction of diazomethane with *N*-phenylmaleimide gave rise to a substituted 1-pyrazoline [4]. To the reaction product of diphenyldiazomethane and *N*-phenylmaleimide was assigned a structure of 2-pyrazoline [5] and not 1-pyrazoline as had been reported before [2]. In reaction of diphenyldiazomethane with N-phenylimide of citraconic acid was isolated only a substituted cyclopropane, and the regioselectivity of the addition was not established [2]. The reaction between diazo compounds and imides of itaconic acid was not investigated.

In this study were investigated reactions of diphenyldiazomethane with N-arylsubstituted maleimides **Ia–h**, maleimides **IIa–p** containing a methyl or aryl substituent at the double C=C bond, and substituted imides of itaconic acid **VIIa–d**. The reaction of diphenyldiazomethane with imides **Ia–h** in dichloromethane at room temperature afforded 6,8-dioxo-4,4-diphenyl-2,3,7-triazabicyclo[3.3.0]oct-2-enes (**IIIa–h**). The composition and the structure of pyrazolines **IIIa–h** were established from elemental analyses (Table 1) and spectral data (Table 2).

In the IR spectrum of pyrazoline **IIIa** appears an absorption band of C=O group (1720 cm^{-1}) and lacks the absorption band of NH group. In the ^1H NMR spectra of pyrazolines **IIIa–h** are present doublet signals from hydrogen atoms attached to the bridgehead carbons: C^5H-doublet in the 4.1–4.21 ppm region (J 8 Hz) and a doublet from the proton linked to C^1 in a weaker field at 6.53–6.60 ppm (J 8 Hz). The downfield shift of the signal from the proton at C^1 evidences that this atom is located in *3* position of 1-pyrazoline. In the ^1H NMR spectra of pyrazolines **IVa–d** appears a singlet in the 4.08–4.28 ppm region; this proton is consequently attached to C^5 atom, and the compounds **IV** obtained have the structure of 1-pyrazolines.



Ia–h, IIa–p **IIIa–h, IVa–p**



Va–h, VIa–p

I, III, V, R = H, R' = Ph (**a**), 4-$CH_3C_6H_4$ (**b**), 4-ClC_6H_4 (**c**), 4-BrC_6H_4 (**d**), 4-$NO_2C_6H_4$ (**e**), 2-ClC_6H_4 (**f**), 2-$CH_3OC_6H_4$ (**g**), 2,4-$(CH_3)_2C_6H_3$ (**h**); **II, IV, VI,** R = Me, R' = Ph (**a**), 4-$CH_3C_6H_4$ (**b**), 4-ClC_6H_4 (**c**), 2-ClC_6H_4 (**d**), 3-$CF_3C_6H_4$ (**e**), 3-BrC_6H_4 (**f**); R = R' = Ph (**g**); R = Ph, R' = 4-$CH_3C_6H_4$ (**h**), 4-ClC_6H_4 (**i**); R = 4-$CH_3C_6H_4$, R' = Ph (**j**), 4-$CH_3C_6H_4$ (**k**), 4-ClC_6H_4 (**l**), 2-ClC_6H_4 (**m**); R = 4-ClC_6H_4, R' = Ph (**n**), 4-$CH_3C_6H_4$ (**o**), 4-ClC_6H_4 (**p**).

1-Pyrazolines **IIIa–h** and **IVa–d** readily loose nitrogen already at room temperature, and the substances obtained contain a little of cyclopropane compounds impurities. On heating pyrazolines **IIIa–h**

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Table 1. Yields, melting points, and elemental analyses of compounds **IIIa, IVb, Vc–g, VIa–p, VIIIa–d**

Compd. no.	Yield, %	mp, °C	Found, %			Formula	Calculated, %		
			C	H	N		C	H	N
IIIa	93	143[a]	75.26	4.75	11.45	$C_{23}H_{17}N_3O_2$	75.19	4.66	11.44
IVb	55	135[a]	74.10	5.26	10.35	$C_{20}H_{20}N_2O_3$	73.93	5.35	10.63
Vc	95	170	74.15	4.41	3.84	$C_{23}H_{16}ClNO_2$	73.90	4.31	3.74
Vd	90	206	65.87	3.91	3.54	$C_{23}H_{16}BrNO_2$	66.04	3.86	3.35
Ve	50	209	72.03	4.45	7.05	$C_{23}H_{16}N_2O_4$	71.87	4.20	7.28
Vf	83	205	73.74	4.52	3.97	$C_{23}H_{16}ClNO_2$	73.90	4.31	3.74
Vg	70	190	75.84	4.79	3.92	$C_{24}H_{19}NO_3$	75.58	5.02	3.67
VIa	78	189	81.53	5.42	3.87	$C_{24}H_{19}NO_2$	81.56	5.42	3.96
VIb	41	191	81.57	5.86	3.63	$C_{25}H_{21}NO_2$	81.72	5.76	3.81
VIc	83	156	74.41	4.65	3.38	$C_{24}H_{18}ClNO_2$	74.32	4.68	3.61
VId	46	195	74.37	4.74	3.39	$C_{24}H_{18}ClNO_2$	74.32	4.68	3.61
VIe	68	151	71.37	4.50	3.32	$C_{25}H_{18}F_3NO_2$	71.25	4.31	3.32
VIf	80	169	66.82	4.44	3.20	$C_{24}H_{18}BrNO_2$	66.68	4.20	3.24
VIg	43	105	83.98	5.22	3.26	$C_{29}H_{21}NO_2$	83.83	5.09	3.37
VIh	85	104	83.69	5.61	3.15	$C_{30}H_{23}NO_2$	83.89	5.40	3.26
VIi	68	175	77.51	4.60	2.78	$C_{29}H_{20}ClNO_2$	77.42	4.48	3.11
VIj	87	99	83.81	5.52	3.11	$C_{30}H_{23}NO_2$	83.89	5.40	3.26
VIk	62	173	84.06	5.88	2.92	$C_{31}H_{25}NO_2$	83.95	5.68	3.16
VIl	68	201	77.76	4.89	2.73	$C_{30}H_{22}ClNO_2$	77.66	4.78	3.02
VIm	36	236	77.51	4.91	2.46	$C_{30}H_{22}ClNO_2$	77.66	4.78	3.02
VIn	88	199	77.16	4.63	2.94	$C_{29}H_{20}ClNO_2$	77.42	4.48	3.11
VIo	66	206	77.43	5.00	2.73	$C_{30}H_{22}ClNO_2$	77.66	4.78	3.02
VIp	64	207	71.79	4.08	2.55	$C_{29}H_{19}Cl_2NO_2$	71.91	3.95	2.89
VIIIa	64	192	81.51	5.45	3.84	$C_{24}H_{19}NO_2$	81.56	5.42	3.96
VIIIb	88	178	81.38	5.72	3.72	$C_{25}H_{21}NO_2$	81.72	5.72	3.72
VIIIc	66	212	74.27	4.69	3.41	$C_{24}H_{18}ClNO_2$	74.32	4.68	3.61
VIIId	75	188–195	74.42	4.75	3.53	$C_{24}H_{18}ClNO_2$	74.32	4.68	3.61

[a] With decomposition.

in toluene to 110°C for 1 h, and pyrazolines **IVa–d** in benzene to 80°C for 1.5 h a nitrogen evolution was observed, and 3-azabicyclo[3.1.0]hexanes **Va–h** and **VIa–d** were obtained in good yield. In reactions of diphenyldiazomethane with maleimides containing aromatic substituents at the double bond we in most cases failed to detect 1-pyrazolines: when the reaction was carried out at room temperature only the corresponding cyclopropane derivatives **VIg–p** were isolated. From reaction products of diphenyldiazomethane and imide **IIh** we isolated a mixture of cyclopropane **Vh** with a compound that was assigned a structure of 1-pyrasoline **IVh** basing on the presence in its ^{1}H NMR spectrum of a singlet at 4.67 ppm. The composition and structure of compounds **Va–h** and **VIa–p** were established from the elemental analyses (Table 1) and spectral data (Table 2).

In the IR spectra of compounds **V** and **VI** appears the absorption band of C=O group (1710 cm^{-1}). In the ^{1}H NMR spectra of products **Va–h** and **VIa–f** the singlet signal of cyclopropane protons is observed in 3.35–3.65 ppm region, and of compounds **VIg–p** with aryl substituents in position *1* this signal is shifted downfield to 4.00 ppm. Physical constants of compounds **Va, b, h** are in agreement with the published data [2].

The reaction of diphenyldiazomethane with the *ortho*-substituted maleimides **If, g, IId** due to the hindered rotation of the *ortho*-substituted aryl group around the bond N–C_{arom} gives rise to two conformational forms of pyrasolines **IIIf, g** and **IVd** in the ratios 1.6:1, 1.2:1, and 1.2:1 for compounds **IIIf, IIIg**, and **IVd** respectively. The signals from hydrogen atoms at the bridgehead carbons appear downfield

Table 2. IR and ^{1}H NMR spectra of compounds **IIIa–g, IVa–d, Va–h, VIa–p, VIIIa–d**

Compd. no.	IR spectrum, cm^{-1}	^{1}H NMR spectrum, δ, ppm (*J*)
IIIa	1185, 1235, 1380 s, 1450, 1500, 1600, 1720 v.s, 1800, 3070	4.18 d (1H, 8), 6.55 d (1H, 8), 6.73–6.80 (4H), 7.25–7.58 (9H), 7.72–7.78 (2H)
IIIb		2.28 s (3H), 4.16 d (1H, 8), 6.53 d (1H, 8), 6.65–6.73 (4H), 7.15–7.58 (8H), 7.73 d (2H, 8)
IIIc		4.17 d (1H, 8), 6.54 d (1H, 8), 6.73 d (2H, 7), 6.83 d (2H, 8), 7.31–7.58 (8H), 7.74 d (2H, 8)
IIId		4.17 d (1H, 8), 6.54 d (1H, 8), 6.71–6.78 (4H), 7.27–7.93 (10H)
IIIe		4.21 d (1H, 8), 6.60 d (1H, 8), 6.74 · (2H), 7.10 d (2H, 9), 7.28–7.60 (6H), 7.76 d (2H, 7), 8.28 d (2H, 9)
IIIf		4.35 d, 4.53 d (1H, 8), 6.48 d, 6.74 d (1H, 8), 6.55 d, 6.84 d (1H, 8), 6.98 m (1H), 7.27–7.65 (10H), 7.77 d (2H, 8)
IIIg		3.72 s, 3.74 s (3H), 4.27 d, 4.44 d (1H, 8), 6.29 d, 6.77 d (1H, 8), 6.38 d, 6.67 d (1H, 8), 6.93–7.60 (11H), 7.76 d (2H, 8)
IVa		1.80 s (3H), 4.10 s (1H), 6.78 m (2H), 6.89 m (2H), 7.28–7.75 (11H)
IVb	1130, 1240 s, 1370 s, 1385 s, 1450, 1525, 1600, 1710 v.s, 3055	1.97 s (3H), 2.33 s (3H), 3.68 s (1H), 6.58 d (2H, 8), 6.90 m (2H), 7.13 d (2H, 8), 7.30–7.60 (6H), 7.85 d (2H, 8)
IVc		1.79 s (3H), 4.08 s (1H), 6.76 d (2H, 8), 6.95 d (2H, 8), 7.35–7.60 (8H), 7.70 d (2H, 8)
IVd		1.61 s, 1.84 s (3H), 4.28 s, 4.50 s (1H), 6.51 d (0.5H, 7), 6.80 d (1H, 8), 6.95 d (1H, 8), 7.20–7.70 (11.5H), 7.78 d (1H, 8)
Va	1190, 1240, 1390 s, 1450, 1505, 1600, 1710 v.s, 1785, 3035, 3070	3.35 s (2H), 6.38 m (2H), 7.20–7.43 (11H), 7.55 m (2H)
Vb	1979, 1190, 1240, 1390 s, 1450, 1520, 1600, 1710 v.s, 1785, 3060	2.28 s (3H), 3.33 s (2H), 6.22 d (2H, 8), 7.05 d (2H, 8), 7.25–7.42 (8H), 7.57 m (2H)
Vc	1020, 1070, 1100, 1185, 1230, (2H) 1385 s, 1450, 1495 s, 1600, 1710 v.s, 1785, 3070	3.35 s (2H), 6.31 d (2H, 8), 7.21 d (2H, 8), 7.25–7.43 (10H), 7.55 m
Vd	1020, 1075, 1190, 1230, 1385 s, 1450, 1490 s, 1600, 1710 v.s, 1785,	3.34 s (2H), 6.26 d (2H, 8), 7.23–7.37 (10H), 7.55 m (2H) 3070
Ve	1070, 1180, 1230, 1335 s, 1385 s, 1450, 1500, 1530, 1600, 1710 v.s, 1785, 2960, 3030	3.40 s (2H), 6.67 d (2H, 8), 7.27–7.42 (8H), 7.55 m (2H), 8.10 d (2H, 8)
Vf	1075, 1190, 1230, 1385 s, 1450, 1490 s, 1600, 1715 v.s, 1785, 3070	3.40 s (2H), 5.18 d (1H, 8), 7.00 m (1H), 7.25–7.47 (10H), 7.60 m (2H)
Vg	1035, 1075, 1125, 1190, 1260, 1285, 1390 s, 1465, 1505, 1600, 1710 v.s, 1785, 1785, 3060	3.35 s (2H), 3.79 s (3H), 5.18 d (1H, 8), 6.65 m (1H), 6.92 d (1H, 8), 7.25–7.45 (9H), 7.58 m (2H)
Vh	1075, 1190, 1230, 1385 s, 1450, 1505, 1600, 1710 v.s, 1785, 3050	2.13 s (3H), 2.27 s (3H), 3.35 s (2H), 4.90 d (1H, 8), 6.71 d (1H, 8), 7.03 m (1H), 7.26–7.48 (8H), 7.60 m (2H)
VIa	1140, 1240, 1390 s, 1450, 1500, 1600, 1710 v.s, 1785, 3070	1.50 s (3H), 3.28 s (1H), 6.73 m (2H), 6.88 m (2H), 7.30–7.55 (9H), 7.87 m (2H)
VIb	1140, 1235, 1390 s, 1450, 1520, 1600, 1710 v.s, 1785, 3040	1.49 s (3H), 2.28 s (3H), 3.25 s (1H), 6.20 d (2H, 8), 7.03 d (2H, 8), 7.25–7.55 (10H)
VIc	1020, 1100 s, 1140 s, 1240, 1390 s, 1450, 1500 s, 1600, 1710 v.s, 1780, 3065	1.49 s (3H), 3.28 s (1H), 6.30 d (2H, 8), 7.18–7.39 (12H)

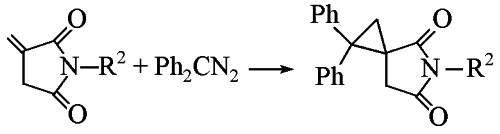
Table 2. (Contd.).

Compd. no.	IR spectrum, cm^{-1}	^1H NMR spectrum, δ, ppm (*J*)
VId	1140, 1270, 1390 s, 1450, 1490 s, 1600, 1710 v.s, 1785, 3070 3070	1.52 s (3H), 3.32 s (1H), 5.15 d.d (1H, 8, 1), 7.00 m (1H), 7.20–7.50 (10H), 7.58 m (2H)
VIe	1080, 1140, 1180, 1230, 1330 s, 1390, 1445, 1495, 1710 v.s, 1785,	1.36 s (3H), 3.68 s (1H), 6.33 s (1H), 6.81 d (1H, 8), 7.37–7.70 (12H)
VIf	3070, 1080, 1140 s, 1230, 1385 s, 1460,1480, 1590, 1710 v.s, 1785, 3070	1.35 s (3H), 3.65 s (1H), 6.28 s (1H), 6.42 d (1H, 8), 7.22–7.58 (12H)
VIg	1140, 1180, 1240, 1385 s, 1450, 1500, 1600 s, 1710 v.s, 1785, 3070	4.01 s (1H), 6.37 m (2H), 7.03–7.75 (18H)
VIh	1140, 1185, 1235, 1390 s, 1450, 1510, 1600, 1710 v.s, 1785, 3040	2.29 s (3H), 4.00 s (1H), 6.23 d (2H, 8), 7.00–7.72 (17H)
VIi	1020, 1100, 1140, 1180, 1240, 1385 s, 1450, 1500 s, 1600, 1710 v.s, 1785, 3070	4.01 s (1H), 6.33 d (2H, 9), 7.00–7.72 (17H)
VIj	1140, 1185, 1240, 1390 s, 1450, 1520, 1600, 1710 v.s, 1785, 3065	2.29 s (3H), 3.97 s (1H), 6.37 m (2H), 7.03–7.48 (15H), 7.70 m (2H)
VIk	1140, 1185, 1240, 1390 s, 1450, 1520, 1600, 1710 v.s, 1785, 3050	2.28 s (3H), 2.29 s (3H), 3.95 s (1H), 6.23 d (2H, 8), 7.00–7.18 (9H), 7.37–7.47 (5H), 7.68 d (2H, 8)
VIl	1020, 1100, 1140, 1180, 1240, 1385 s, 1450, 1495 s, 1600, 1710 v.s, 1785, 3060	2.29 s (3H), 3.96 s (1H), 6.32 d (2H, 8), 7.02–7.26 (9H), 7.32–7.45 (5H), 7.74 m (2H)
VIm	1140, 1180, 1390 s, 1450, 1485, 1600, 1710 v.s, 1790, 3050	2.29 s (3H), 4.00 s (1H), 5.22 d (1H, 7), 7.00–7.53 (15H), 7.70 m (2H)
VIn	1020, 1140, 1180, 1240, 1390 s, 1450, 1500 s, 1600, 1710 v.s, 1785, 3070	3.99 s (1H), 6736 m (2H), 7.03–7.52 (15H), 7.67 m (2H)
VIo	1020, 1100, 1140, 1185, 1240, 1385 s, 1450, 1500, 1600, 1710 v.s, 1785, 3050	2.29 s (3H), 3.97 s (1H), 6.23 d (2H, 9), 7.00–7.50 (14H), 7.68 m (2H)
VIp	1020, 1100 s, 1140, 1180, 1240, 1385 s, 1450, 1495 s, 1600, 1710 v.s, 1785, 3070	3.99 s (1H), 6.32 d (2H, 8), 7.03–7.50 (14H), 7.67 m (2H)
VIIIa	1140 s, 1190, 1240, 1385, 1460, 1505, 1600, 1710 v.s, 1785, 3070	2.15 d (1H, 5), 2.49 d (1H, 5), 2.71 d (1H, 19), 2.89 d (1H, 19), 7.18–7.53 (15H)
VIIIb	1140, 1190, 1240, 1385 s, 1460, 1520, 1600, 1710 v.s, 1785, 3050	2.14 d (1H, 5), 2.38 s (3H), 2.48 d (5), 2.69 d (1H, 19), 2.88 d (1H, 19), 7.17–7.45 (14H)
VIIIc	1020, 1100, 1140 s, 1190, 1240, 1380 s, 1460, 1500 s, 1600, 1710 v.s, 1790, 3060	2.16 d (1H, 5), 2.49 d (1H, 5), 2.69 d (1H, 19), 2.88 d (1H, 19), 7.22–7.47 (14H)
VIIId	1070, 1145, 1190, 1235, 1385 s, 1450, 1490 s, 1600, 1720 v.s, 1790, 3070	2.15 d, 2.16 d (1H, 5), 2.49 d, 2.53 d (1H, 5), 2.71 d, 2.80 d (1H, 19), 2.91 d, 2.98 d (1H, 19), 7.14–7.60 (14H)

for the prevailing isomer, as follows: for compound **IIIf** at 4.35 d (*J* 8 Hz) and 4.53 d (*J* 8 Hz), for compound **IIIg** at 4.27 d (*J* 8 Hz) and 4.44 d (*J* 8 Hz), for compound **IVd** at 4.28 s and 4.50 s. At the same time cyclopropane compounds **Vf, g, h,** and **VId, m** with *ortho*-substituents in the aromatic ring of the imide moiety form a single isomer, presumably with *anti*-configuration due to strong sterical interactions between the *ortho*-substituted aromatic ring and the aromatic substituent located at the three-membered cycle in the *syn*-configuration. Due to the strong screening effect of phenyl group exerted on the *ortho*-

protons of the benzene ring attached to the imide moiety the signals of these protons in the ^{1}H NMR spectra appear in 4.90–5.22 ppm region. A similar upfield shift of the signal from ortho-protons of benzene ring due to the screening effect of a phenyl group was observed in imides prepared by reaction of anthracene with *ortho*-substituted maleimides [6].

At treating N-arylsubstituted imides of itaconic acid **VIIa–d** with diphenyldiazomethane in dichloromethane at 18–20°C we failed to isolate the intermediately arising pyrazolines. In the course of reaction nitrogen is liberated, and substituted 5-azaspiro[2.4]heptanes (**VIII–d**) form in good yield.



VIIa–d **VIIIa–d**

VII, VIII, R = H (**a**), 4-Me (**b**), 4-Cl (**c**), 2-Cl (**d**).

In the IR spectra of compounds obtained is present an absorption band of C=O group (1710 cm^{-1}), in the ^{1}H NMR spectra appear doublet signals of the protons in the cyclopropane ring in the 2.15 and 2.50 ppm region (*J* 5 Hz), and doublets of protons attached to C^7 atom at 2.70 and 2.90 ppm (*J* 19 Hz). Unlike *ortho*-substituted bicyclohexanes **Vf, g, h** and **VId, m** that were obtained as single stereoisomers, spiroheptane **VIIId** arose as two stereoisomers.

Thus it is presumable that the reaction between diphenyldiazomethane and substituted maleimides occurs regioselectively to afford 1-pyrazolines which at heating liberate nitrogen to form 3-azabicyclo[3.1.0]hexanes; the substituents at the multiple bond facilitate the process. In reaction of diphenyldiazomethane with imides of itaconic acid the intermediate 1-pyrazolines decompose with nitrogen evolution already at room temperature forming substituted spiroheptanes.

EXPERIMENTAL

IR spectra were recorded on spectrophotometer UR-20 from 2% solutions of compounds in chloroform. ^{1}H NMR spectra were registered on spectrometer Bruker DPX-300 (300 MHz) from 2% solutions in $CDCl_3$ or DMSO-d_6.

7-Aryl-6,8-dioxo-4,4-diphenyl-2,3,7-triazabicyclo[3.3.0]oct-2-enes (IIIa–h), and 7-aryl-1-methyl-6,8-dioxo-4,4-diphenyl-2,3,7-triazabicyclo[3.3.0]-oct-2-enes (IVa–f). To a solution of 5 mmol of *N*-arylmaleimide (**Ia–h**) or N-arylimide of citraconic acid (**IIa–d**) in 10 ml of dichloromethane was added a solution of 1 g (5 mmol) of diphenyldiazomethane in 10 ml of dichloromethane, The mixture was kept for 2–4 days till disappearance of the diphenyldiazomethane color. The separated precipitate was filtered off and washed with ethanol.

3-Aryl-2,4-dioxo-6,6-diphenyl-3-azabicyclo-[3.1.0]hexanes (Va–h). A mixture of 4 mmol of pyrazoline **IIIa–h** and 10 ml of toluene was heated to 100°C for 1 h. The pyrazoline gradually dissolved. The toluene was distilled off in a vacuum, the separated precipitate was recrystallized from ethanol.

3-Aryl-1-methyl-6,6-diphenyl-3-azabicyclo[3.1.0]-hexanes (VIa–f). A mixture of 0.7 mmol of pyrazoline **IVa–f** with 5 ml of benzene was heated to 80°C for 1.5 h. The pyrazoline gradually dissolved in the course of heating. Then benzene was vacuum-evaporated, and the residue was crystallized from ethanol.

1,3-Diaryl-6,6-diphenyl-3-azabicyclo[3.1.0]-hexanes (VIg–p). To a solution of 0.5 mmol of imide **IIg–p** in 5 ml of dichloromethane was added 0.1 g (0.5 mmol) of diphenyldiazomethane in 5 ml of dichloromethane. The mixture was kept for 3–4 days at room temperature till disappeared the color of diphenyldiazomethane, then the solvent was vacuum-evaporated, and the residue was recrystallized from ethanol.

5-Aryl-4,6-dioxo-1,1-diphenyl-5-azaspiro[2.4]-heptanes (VIIIa–d). To a solution of 0.5 mmol of imide **VIIa–d** in 5 ml of dichloromethane was added a solution of 0.1 g (0.5 mmol) of diphenyldiazomethane. The reaction mixture was kept at room temperature for 48 h, the solvent was evaporated, and the residue was crystallized from benzene–ethanol mixture.

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