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Reactions of Aliphatic Diazo Compounds: VI.* Reactions of Diazomethane and Ethyl Diazoacetate with (*E*)-2-Arylmethylene-1,2,3,4-tetrahydronaphthalen-1-ones

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Abstract—Diazomethane and ethyl diazoacetate add to (*E*)-2-arylmethylene-1,2,3,4-tetrahydronaphthalen-1-ones in a regio- and stereoselective fashion, yielding the corresponding 4'-aryl-1,2,3,4,4',5'-hexahydro-3'*H*-naphthalene-2-spiro-3'-pyrazol-1-ones. The products formed by addition of ethyl diazoacetate undergo isomerization into 4,5-dihydro-1*H*-pyrazole derivatives.

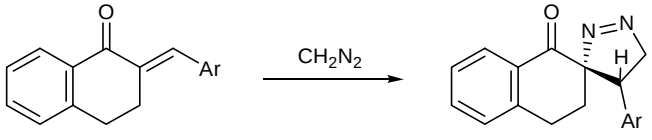
We previously showed that 1,3-dipolar cycloaddition of ethyl diazoacetate to 1,3-diaryl-2-propen-1-ones occurs with high regioselectivity to afford isomeric 4,5-dihydro-1*H*-pyrazoles [2]. Reactions of diazomethane with α,β-unsaturated cyclic ketones having an exocyclic double bond, e.g., 2-arylmethylene-3-phenyl-1-indanones, 3-arylmethyleneflavanones, etc. [3], were studied by many authors. It was found that the process is regioselective and that the products are spiro-4,5-dihydro-3*H*-pyrazoles. However, there are no published data on reactions of such compounds with diazoacetic acid esters.

We have studied reactions of diazomethane and ethyl diazoacetate with (*E*)-2-arylmethylene-1,2,3,4-tetrahydronaphthalen-1-ones **Ia**–**Ih**. Diazomethane reacted with compounds **Ia**–**Ih** in diethyl ether at 0°C to afford 37–69% of the corresponding 4'-aryl-1,2,3,4,-4',5'-hexahydro-3'*H*-naphthalene-2-spiro-3'-pyrazol-1-ones **IIa**–**IIh** (Scheme 1). The structure of products **IIa–IIh** was determined on the basis of their spectral parameters and elemental compositions. The IR spectra of **IIa–IIh** lacked NH absorption, while their ^{1}H NMR spectra contained signals from the 4'-H proton in the pyrazole ring as a doublet of doublets at δ 3.82–4.31 ppm and signals from two 5'-H protons in the δ region of 5.0 ppm. According to our previous data and those reported in [4], the phenyl group in compound **IIa** is arranged *trans* with respect to the carbonyl group.

Reactions of tetralones **Ia**–**Id**, **If**, and **Ii** with ethyl diazoacetate in toluene at 90–95°C in 120 h afforded up to 56% of the corresponding ethyl 4'-aryl-1-oxo-1,2,3,4,-4',5'-hexahydro-1'*H*-naphthalene-2-spiro-5'-pyrazole-3'-carboxylates **IIIa**–**IIIf** (Scheme 2).

The structure of compounds **IIIa**–**IIIf** was derived from their elemental analyses and spectral data. The IR spectra of **IIIa**–**IIIf** contained an absorption band at 3370 cm^{-1} due to NH group. In the ^{1}H NMR spectra we observed a singlet at δ 9.25–9.52 ppm (NH), a singlet at δ 4.57–4.92 ppm from 4'-H, and also signals from aromatic protons and protons of the ethyl and –CH_2CH_2– groups. The signal from the aromatic *ortho* protons appears as a broadened singlet at room temperature; on heating to 50°C it is converted into

Scheme 1.



Ia–Ih **IIa–IIh**

Ar = Ph (**a**), 4-MeC_6H_4 (**b**), 4-ClC_6H_4 (**c**), 4-BrC_6H_4 (**d**), 4-$MeOC_6H_4$ (**e**), 3-$NO_2C_6H_4$ (**f**), 3-$MeOC_6H_4$ (**g**), 2-ClC_6H_4 (**h**).

* For communication V, see [1].

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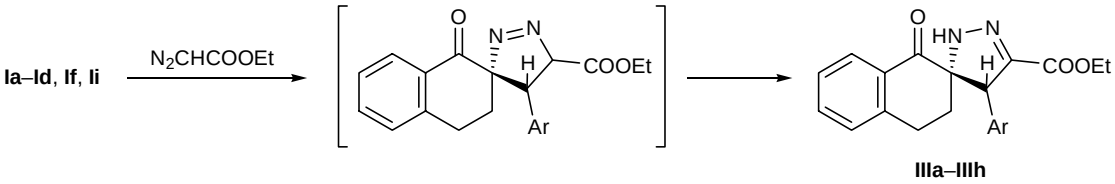
Scheme 2.



Ii, Ar = 4-$NO_2C_6H_4$; **III**, Ar= Ph (**a**), 4-MeC_6H_4 (**b**), 4-ClC_6H_4 (**c**), 4-BrC_6H_4 (**d**), 4-$NO_2C_6H_4$ (**e**), 3-$NO_2C_6H_4$ (**f**).

a doublet with J = 8.0 Hz. This patten may be interpreted in terms of restrictied rotation of the aryl group in molecules **IIIa**–**IIIf** due to spatial interaction with the $-C^3H_2C^4H_2-$ group, which is possible only when the aromatic and carbonyl groups are arranged *trans*. The $^1H-^1H$ NOESY spectrum of compound **IIIb** displayed interaction between the C^3H_2 protons, protons in the *ortho* position of the aromatic ring, and NH proton, whereas no interaction between NH and 4'-H was observed. The ^{13}C NMR spectrum of **IIIa** contained signals at δ_C 14.9 (CH_3), 26.1 (CH_2), 29.2 (CH_2), 54.0 (CH), 60.8 (CH_2), 74.1 (C), 162.5 (CO), and 194.1 ppm (CO), as well as signals from the aromatic carbon atoms. The signal from $C^{3'}$ (where the ester group is attached) is located at δ 130–145 ppm.

Thus the reactions of diazomethane and ethyl diazoacetate with 2-arylmethylenetetrahydronaphthalen-1-ones **I** are regio- and stereoselective, and the nucleophilic carbon atom in the diazo compound adds at the unsaturated carbon atom in the β-position with respect to the carbonyl group, i.e., in keeping with the Auvers rule.

EXPERIMENTAL

The IR spectra were recorded on a UR-20 spectrophotometer from 2% solutions in $CHCl_3$. The ^1H and ^{13}C NMR spectra were recorded on a Bruker DPX-300 instrument at 300.13 and 75.47 MHz, respectively, from solutions in DMSO-d_6. The reaction mixtures were analyzed, and the purity of the products was checked, by TLC on Silufol UV-254 plates. Initial 2-arylmethylene-1,2,3,4-tetrahydronaphthalen-1-ones **Ia**–**Ii** were synthesized as described in [5].

4'-Aryl-1,2,3,4,4',5'-hexahydro-3'*H*-naphthalene-2-spiro-3'-pyrazol-1-ones IIa–IIh (*general procedure***).** A solution of diazomethane in diethyl ether, prepared from 5 g (49 mmol) of *N*-nitrosomethylurea, was added to a cold solution of 4 mmol of the corresponding 2-arylmethylene-1,2,3,4-tetrahydronaphthalen-1-one **Ia–Ih** in 10 ml of benzene, and the mixture was left overnight. The solvent was evaporated, and the residue was recrystallized from ethanol.

4'-Phenyl-1,2,3,4,4',5'-hexahydro-3'*H*-naphthalene-2-spiro-3'-pyrazol-1-one (IIa). Yield 54%, mp 70–72°C. IR spectrum, ν, cm^{-1}: 920, 1300, 1455, 1600 s, 1680 v.s, 2940, 3030. ^1H NMR spectrum, δ, ppm (*J*, Hz): 1.73 m (1H), 2.10 d.t (1H, *J* = 14, 5), 2.77 d.t (1H, *J* = 17, 5), 3.23 m (1H), 3.90 d.d (1H, *J* = 7, 6), 4.98 d. d (1H, *J* = 18, 7), 5.00 d. d (1H, *J* = 18, 6), 7.05 d (1H, *J* = 7), 7.19–7.33 (3H), 7.36–7.48 (2H), 7.65 t (1H, *J* = 8), 7.95 d (1H, *J* = 8). Found %: C 78.04; H 5.87; N 9.87. $C_{18}H_{16}N_2O$. Calculated, %: C 78.24; H 5.84; N 10.14.

4'-(4-Methylphenyl)-1,2,3,4,4',5'-hexahydro-3'*H*-naphthalene-2-spiro-3'-pyrazol-1-one (IIb). Yield 60%, mp 119–121°C. IR spectrum, ν, cm^{-1}: 920, 1030, 1120, 1160, 1245, 1300, 1455, 1600 s, 1690 v.s, 2930, 3040. ^1H NMR spectrum ($CDCl_3$), δ, ppm (*J*, Hz): 1.93 m (1H), 2.20 d. t (1H, *J* = 14, 5), 2.33 s (3H), 2.88 d.t (1H, *J* = 17, 5), 3.52 m (1H), 3.88 d.d (1H, *J* = 7, 5), 5.03 d.d (1H, *J* = 18, 7), 5.05 d. d (1H, *J* = 18, 5), 6.85 d (2H, *J* = 8), 7.10 d (2H, *J* = 8), 7.30 d (1H, *J* = 8), 7.38 d.d (1H, *J* = 8, 7), 7.56 d.d (1H, *J* = 8, 7), 8.08 d (1H, *J* = 8). Found %: C 78.39; H 6.27; N 9.26. $C_{19}H_{18}N_2O$. Calculated, %: C 78.59; H 6.25; N 9.65.

4'-(4-Chlorophenyl)-1,2,3,4,4',5'-hexahydro-3'*H*-naphthalene-2-spiro-3'-pyrazol-1-one (IIc). Yield 60%, mp 87–89°C. IR spectrum, ν, cm^{-1}: 920 s, 1020, 1100 s, 1160, 1290, 1300, 1355, 1450, 1490 s, 1600 s, 1680 v.s, 2860, 2940, 3030. ^1H NMR spectrum, δ, ppm (*J*, Hz): 1.73 m (1H), 2.08 d. t (1H, *J* = 14, 5), 2.81 d.t (1H, *J* = 17, 5), 3.26 m (1H), 3.94 d.d (1H, *J* = 7, 6), 4.97 d.d (1H, *J* = 18, 7), 5.01 d.d (1H, *J* = 18, 6), 7.09 d (2H, *J* = 8), 7.35 d (2H, *J* = 8), 7.40 d (1H, *J* = 7), 7.43 d.d (1H, *J* = 8, 7), 7.65 d.d (1H, *J* = 8, 7), 7.96 d (1H, *J* = 8). Found, %: C 69.66; H 4.86; N 8.84. $C_{18}H_{15}ClN_2O$. Calculated, %: C 69.57; H 4.86; N 9.01.

4'-(4-Bromophenyl)-1,2,3,4,4',5'-hexahydro-3'*H*-naphthalene-2-spiro-3'-pyrazol-1-one (IId). Yield

47%, mp 124°C. IR spectrum, ν, cm^{-1}: 920, 1020, 1080, 1120, 1160, 1245, 1290, 1300, 1450, 1490 s, 1600 s, 1690 v.s, 2940, 3040. ^{1}H NMR spectrum ($CDCl_3$), δ, ppm (*J*, Hz): 1.89 m (1H), 2.12 d. t (1H, *J* = 14, 5), 2.90 d. t (1H, *J* = 17, 5), 3.59 m (1H), 3.93 t (1H, *J* = 7), 5.04 d (2H, *J* = 7), 6.85 d (2H, *J* = 8), 7.31 d (1H, *J* = 8), 7.38 d.d (1H, *J* = 8, 7), 7.58 d.d (1H, *J* = 8, 7), 8.08 d (1H, *J* = 8). Found, %: C 61.09; H 4.37; N 7.72. $C_{18}H_{15}BrN_2O$. Calculated, %: C 60.86; H 4.26; N 7.89.

4'-(4-Bromophenyl)-1,2,3,4,4',5'-hexahydro-3'*H*-naphthalene-2-spiro-3'-pyrazol-1-one (IIe). Yield 67%, mp 138°C. IR spectrum, ν, cm^{-1}: 910, 1040, 1260, 1300, 1455, 1520, 1600 s, 1690 s, 2830, 2940, 3040. ^{1}H NMR spectrum, δ, ppm (*J*, Hz): 1.75 m (1H), 2.14 d.t (1H, *J* = 14, 5), 2.79 d.t (1H, *J* = 17, 5), 3.19 m (1H), 3.71 s (3H), 3.82 t (1H, *J* = 7), 4.92 d (1H, *J* = 7), 6.84 d (2H, *J* = 8), 6.95 d (2H, *J* = 8), 7.39 d (1H, *J* = 8), 7.42 t (1H, *J* = 8), 7.64 t (1H, *J* = 8), 7.96 d (1H, *J* = 8). Found, %: C 74.49; H 5.88; N 8.88. $C_{19}H_{18}N_2O_2$. Calculated, %: C 74.49; H 5.92; N 9.14.

4'-(4-Nitrophenyl)-1,2,3,4,4',5'-hexahydro-3'*H*-naphthalene-2-spiro-3'-pyrazol-1-one (IIf). Yield 69%, mp 80–82°C. IR spectrum, ν, cm^{-1}: 915, 1020, 1090, 1160, 1300, 1350 s, 1450, 1540 s, 1600, 1690 s, 2950, 3040. ^{1}H NMR spectrum, δ, ppm (*J*, Hz): 1.75 m (1H), 2.05 d.t (1H, *J* = 14, 5). 2.85 d.t (1H, *J* = 17, 5), 3.30 m (1H), 4.16 d.d (1H, *J* = 8, 6), 5.05 d.d (1H, *J* = 18, 8), 5.12 d.d (1H, *J* = 18, 6), 7.40 d (1H, *J* = 8), 7.44 t (1H, *J* = 8), 7.53 d (1H, *J* = 8), 7.60 t (1H, *J* = 8), 7.66 t (1H, *J* = 8), 7.95 s (1H), 7.98 d (1H, *J* = 8), 8.12 d (1H, *J* = 8). Found, %: C 67.45; H 4.69; N 12.99. $C_{18}H_{15}N_3O_3$. Calculated, %: C 67.28; H 4.71; N 13.08.

4'-(4-Methoxyphenyl)-1,2,3,4,4',5'-hexahydro-3'*H*-naphthalene-2-spiro-3'-pyrazol-1-one (IIg). Yield 38%, mp 107°C. IR spectrum, ν, cm^{-1}: 905, 1060, 1160, 1300, 1455, 1490, 1600 s, 1690 s, 2840, 2940, 3040. ^{1}H NMR spectrum, δ, ppm (*J*, Hz): 1.79 m (1H), 2.12 d.t (1H, *J* = 14, 5), 2.81 d.t (1H, *J* = 17, 5), 3.24 m (1H), 3.70 s (3H), 3.88 d.d (1H, *J* = 8, 6), 4.97 d.d (1H, *J* = 18, 8), 5.01 d.d (1H, *J* = 18, 6), 6.58 d (1H, *J* = 8), 6.64 d (1H, *J* = 3), 6.82 d. d (1H, *J* = 8, 3), 7.21 t (1H, *J* = 8), 7.39 d (1H, *J* = 8), 7.42 t (1H, *J* = 8), 7.65 t (1H, *J* = 8), 7.98 d (1H, *J* = 8). Found, %: C 74.52; H 5.91; N 9.26. $C_{19}H_{18}N_2O_2$. Calculated, %: C 74.49; H 5.92; N 9.14.

4'-(2-Chlorophenyl)-1,2,3,4,4',5'-hexahydro-3'*H*-naphthalene-2-spiro-3'-pyrazol-1-one (IIh). Yield 43%, mp 103°C. IR spectrum, ν, cm^{-1}: 920, 1045, 1160, 1300 s, 1455, 1480, 1600 s, 1690 s, 2940, 3035. ^{1}H NMR spectrum, δ, ppm (*J*, Hz): 1.78 m (1H), 1.91 d.t (1H, *J* = 14, 5), 2.82 d.t (1H, *J* = 17, 5), 3.40 m (1H), 4.31 d.d (1H, *J* = 9, 3), 4.98 d.d (1H, *J* = 18, 9), 5.24 d.d (1H, *J* = 18, 3), 7.05 d.d (1H, *J* = 8, 2), 7.32 t.d (1H, *J* = 8, 2), 7.37– 7.48 (3H), 7.66 t (1H, *J* = 8), 7.96 d (1H, *J* = 8). Found, %: C 69.42; H 5.04; N 8.77. $C_{18}H_{15}ClN_2O$. Calculated, %: C 69.57; H 4.86; N 9.01.

Ethyl 4'-aryl-1-oxo-1,2,3,4,4',5'-hexahydro-1'*H*-naphthalene-2-spiro-5'-pyrazole-3'-carboxylates IIIa–IIIf (*general procedure*). Ethyl diazoacetate, 2 g (18 mmol), was added to a solution of 3.7 mmol of the corresponding 2-arylmethylene-1,2,3,4-tetrahydronaphthalen-1-one in 30 ml of anhydrous toluene, and the mixture was heated for 120 h at 90–95°C (the progress of the reaction was monitored by TLC). The solvent was distilled off under reduced pressure, and the residue was recrystallized from a 1:8 ethyl acetate–hexane mixture.

Ethyl 1-oxo-4'-phenyl-1,2,3,4,4',5'-hexahydro-1'*H*-naphthalene-2-spiro-5'-pyrazole-3'-carboxylate (IIIa). Yield 34%, mp 148– 149°C. IR spectrum, ν, cm^{-1}: 1070, 1115, 1135, 1300, 1420, 1480, 1500, 1690 s, 2940, 3040, 3370. ^{1}H NMR spectrum, δ, ppm (*J*, Hz): 1.15 t (3H, *J* = 7), 1.69 d.t (1H, *J* = 14, 5), 1.82 m (1H), 2.74 d.t (1H, *J* = 18, 5), 2.91 m (1H), 4.01 m (2H), 4.65 s (1H), 7.08 br.s (2H), 7.26–7.38 m (4H), 7.41 t (1H, *J* = 8), 7.60 t.d (1H, *J* = 8, 1), 7.96 d (1H, *J* = 8), 9.25 s (1H). ^{13}C NMR spectrum (DMSO-d_6), δ, ppm: 14.9 (CH_3), 26.1 (CH_2), 29.2 (CH_2), 54.0 (CH), 60.8 (CH_2), 74.1 (C), 127.8 (CH), 128.3 (CH), 128.6 (CH), 129.4 (CH), 129.8 (CH), 130.7 (C), 134.9 (CH), 136.9 (C), 143.6 (C), 144.5 (C), 162.5 (CO), 194.1 (CO). Found, %: C 72.31; H 5.76%; N 7.95. $C_{21}H_{20}N_2O_3$. Calculated, %: C 72.40; H 5.79; N 8.04.

Ethyl 4'-(4-methylphenyl)-1-oxo-1,2,3,4,4',5'-hexahydro-1'*H*-naphthalene-2-spiro-5'-pyrazole-3'-carboxylate (IIIb). Yield 40%, mp 185–186°C. IR spectrum, ν, cm^{-1}: 1070, 1115, 1250, 1300, 1420, 1600, 1700 s, 2930, 3040, 3370. ^{1}H NMR spectrum, δ, ppm (*J*, Hz): 1.10 t (3H, *J* = 7), 1.75 m (2H), 2.29 s (3H), 2.76 m (1H), 2.88 m (1H), 4.02 m (2H), 4.57 s (1H), 6.97 br.s (2H), 7.15 d (2H, *J* = 8), 7.30 d (1H, *J* = 8), 7.41 t (1H, *J* = 8), 7.58 t (1H, *J* = 8), 7.95 d (1H, *J* = 8), 9.30 s (1H). Found, %: C 72.64; H 6.05; N 7.29. $C_{22}H_{22}N_2O_3$. Calculated, %: C 72.91; H 6.12; N 7.73.

Ethyl 4'-(4-chlorophenyl)-1-oxo-1,2,3,4,4',5'-hexahydro-1'*H*-naphthalene-2-spiro-5'-pyrazole-3'-carboxylate (IIIc). Yield 54%, mp 209–210°C. IR spectrum, ν, cm^{-1}: 1020, 1070, 1095, 1115, 1130, 1250, 1300, 1430, 1490, 1600, 1690 s, 3040, 3370. ^{1}H NMR spectrum, δ, ppm (*J*, Hz): 1.12 t (3H, *J* = 7), 1.68 d.t (1H, *J* = 14, 5), 1.81 m (1H), 2.75 d.t (1H, *J* = 17, 5), 2.92 m (1H), 4.01 m (2H), 4.68 s (1H), 7.11 br.s (2H), 7.31 d (1H, *J* = 8), 7.37–7.45 (3H), 7.59 t (1H, *J* = 8), 7.95 d (1H, *J* = 8), 9.37 s (1H). Found, %: C 65.84; H 5.10; N 7.22. $C_{21}H_{19}ClN_2O_3$. Calculated, %: C 65.88; H 5.00; N 7.32.

Ethyl 4'-(4-bromophenyl)-1-oxo-1,2,3,4,4',5'-hexahydro-1'*H*-naphthalene-2-spiro-5'-pyrazole-3'-carboxylate (IIId). Yield 44%, mp 204°C. IR spectrum, ν, cm^{-1}: 1015, 1075, 1110, 1255, 1300, 1420, 1600, 1700 s, 3365. ^{1}H NMR spectrum, δ, ppm (*J*, Hz): 1.11 t (3H, *J* = 7), 1.66 d.t (1H, *J* = 13, 5), 1.82 m (1H), 2.74 d.t (1H, *J* = 17, 5), 2.94 m (1H), 4.05 m (2H), 4.67 s (1H), 7.05 br.s (2H), 7.31 d (1H, *J* = 8), 7.42 t (1H, *J* = 8), 7.54 d (2H, *J* = 8), 7.61 t (1H, *J* = 8), 7.96 d (1H, *J* = 8), 9.36 s (1H). Found, %: C 58.91; H 4.51; N 6.31. $C_{21}H_{19}BrN_2O_3$. Calculated, %: C 59.03; H 4.48, N 6.56.

Ethyl 4'-(4-nitrophenyl)-1-oxo-1,2,3,4,4',5'-hexahydro-1'*H*-naphthalene-2-spiro-5'-pyrazole-3'-carboxylate (IIIe). Yield 56%, mp 203°C. ^{1}H NMR spectrum, δ, ppm (*J*, Hz): 1.10 t (3H, *J* = 7), 1.64 d.t (1H, *J* = 13, 5), 1.84 m (1H), 2.75 d.t (1H, *J* = 17, 5), 2.94 m (1H), 4.08 m (2H), 4.90 s (1H), 7.32 d (1H, *J* = 8), 7.40 br.s (2H), 7.42 t (1H, *J* = 8), 7.60 t.d (1H, *J* = 8, 1), 7.97 d (1H, *J* = 8), 8.23 d (2H, *J* = 9), 9.52 s (1H). Found, %: C 64.41; H 4.84; N 10.70. $C_{21}H_{19}N_3O_5$. Calculated, %: C 64.12; H 4.87; N 10.68.

Ethyl 4'-(3-nitrophenyl)-1-oxo-1,2,3,4,4',5'-hexahydro-1'*H*-naphthalene-2-spiro-5'-pyrazole-3'-carboxylate (IIIf). Yield 27%, mp 143°C. IR spectrum, ν, cm^{-1}: 1070, 1110, 1240 s, 1300, 1350 s, 1470, 1535, 1600, 1700 s, 3365. ^{1}H NMR spectrum, δ, ppm (*J*, Hz): 1.12 t (3H, *J* = 7), 1.64 m (1H), 1.88 m (1H), 2.76 m (1H), 2.94 m (1H), 4.06 m (2H), 4.92 s (1H), 7.31 d (1H, *J* = 8), 7.41 t (1H, *J* = 8), 7.59 t (1H, *J* = 7), 7.62 br.s (1H), 7.67 t (1H, *J* = 8), 7.97 d (1H, *J* = 8), 8.17 d (1H, *J* = 7), 9.52 s (1H). Found, %: C 64.06; H 5.07; N 10.52. $C_{21}H_{19}N_3O_5$. Calculated, %: C 64.12; H 4.87; N 10.68.

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