Synthesis of 6-(*N*-azolyl)cyclohex-2-enones from *N*-acetonylazoles*

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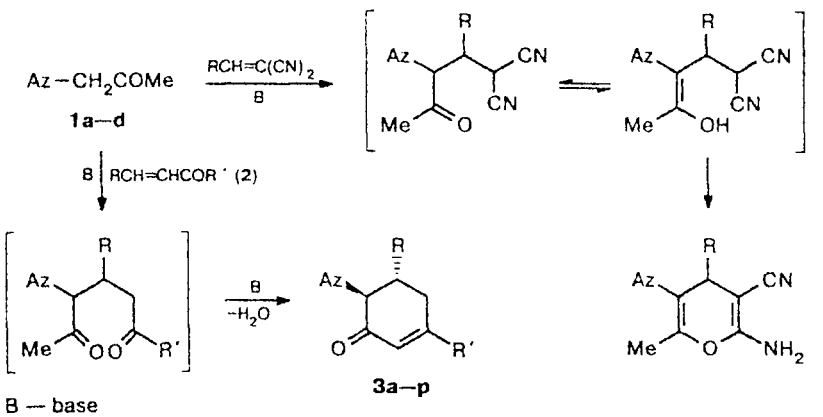
N-Acetonylazoles react with chalcones in the presence of a base to give *trans*-3,5-disubstituted 6-(*N*-azolyl)cyclohex-2-enones. Usually, the reactions are fast and high-yielding.

Key words: *N*-acetonylazoles, α,β-unsaturated ketones, Michael addition, intramolecular crotonic condensation, 6-(*N*-azolyl)cyclohex-2-enones.

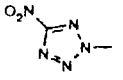
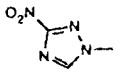
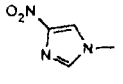
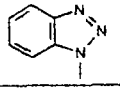
Earlier, we showed that *N*-acetonyl- and *N*-phenacylazoles can react with α,β-unsaturated nitriles of the $RCH{=}C(CN)_2$ type under very mild conditions by involving the acidic CH_2 fragment in the Michael addition. The addition products were not isolated because of further immediate cyclization into the corresponding 2-amino-4*H*-pyrans.[2,3]

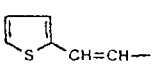
In the present work, we found that the stage of the Michael addition is also not a final one in the reactions of *N*-acetonylazoles **1** with α,β-unsaturated ketones **2**, which yield cyclization products. However, unlike the reactions with nitriles, cyclization involves here the CH_3 group of *N*-acetonylazole to give cyclohex-2-enones **3** (intramolecular crotonic condensation, Scheme 1) rather than pyrans.

Scheme 1



B — base

Compound	Az
1a, 3a—d	
1b, 3e—h	
1c, 3i—l	
1d, 3m—p	

Compound	R	R'
3a,e,i,o	Ph	Ph
3b	Ph	Me
3c,g		
3d,j	Ph	

Compound	R	R'
3h,k		Ph
3f,p		
3l	Ph	PhCH=CH—
3m	Ph	*cyclo*-C_3H_5
3n	3-$NO_2C_6H_4$	Ph

* For the preliminary report see Ref. 1.

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Table 1. Reaction conditions and yields of cyclohex-2-enones **3a—p**

Compound	NaOEt (mol.%)	*t*/min	Yield (%)
3a	10	30	80
3b	40	210	59
3c	10	5	64
3d	10	5	69
3e	10	30	75
3f	10	90	52
3g	10	5	65
3h	10	5	75
3i	25	50	57
3j	25	45	62
3k	25	45	55
3l	25	50	65
3m	40	240	48
3n	10	5	85
3o	10	5	89
3p	10	5	67

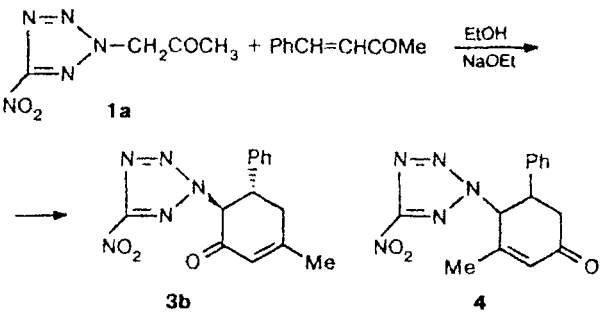
Note: t is the reaction time.

Optimum reaction conditions were found from the reaction of 2-acetonyl-5-nitrotetrazole (**1a**) with chalcone PhCH=CHCOPh, because the conditions of successful reactions of *N*-acetonylazoles with the nitriles $RCH{=}C(CN)_2$ (MeCN, 20 °C, 10 mol. % Et_3N),[2,3] carried out earlier, proved to be unsuitable for this case. The use of MeCN as a solvent and Et_3N as a catalyst initiates the reaction neither at 20 °C nor on refluxing for 6 h. At the same time, refluxing in EtOH with the same amount of Et_3N for 6 h yields product **3a** (conversion of the initial compounds under these conditions is equal to 50% (^{1}H NMR)). When NaOEt (10 mol. %) is used as a catalyst, the initial compounds are completely consumed upon refluxing in EtOH for 30 min to give product **3a** in 80% yield. For this reason, all reactions were carried out in boiling ethanol in the presence of NaOEt as a catalyst. Reaction conditions and the yields of compounds **3** are given in Table 1. A typical procedure of synthesis and isolation of products is described in Experimental.

In the case of 1-acetonyl-4-nitroimidazole (**1c**), a larger amount of catalyst and a longer reaction time are required, probably, because of a lower acidity of the CH_2 group (the same is true for the reactions with ketones **2**, where R′ = Me or *cyclo*-C_3H_5). In the latter case, a portion of base is apparently consumed to abstract the R′ proton nearest the carbonyl group. It is of interest that the reaction of **1a** with benzalacetone PhCH=CHCOMe yields cyclohexenone **3b** as a sole product rather than compound **4** (Scheme 2). This seems to be accounted for by the strong $-I$ effect of the nitrotetrazole substituent and, consequently, the higher acidity of the neighboring CH_3 group.

The reaction is stereoselective, which is evidenced by the ^{1}H NMR spectroscopic data (Table 2). In the spectra of compounds **3**, a signal of the H(6) proton is a doublet with $J_{H(5)H,H(6)}$ = 13.0—13.6 Hz, which suggests a *trans*-diaxial orientation of the H(5) and H(6) protons (or, more precisely, pseudoaxial because the cyclohex-2-enone molecules exist, according to the literature data,[4] in the semichair or sofa conformation). In most cases, two H(4) protons are nonequivalent and have different coupling constants with H(5) (except for compounds **3d** and **3j**). Usually, a signal of the H(4) proton that has the higher coupling constant with H(5) also has a long-range coupling constant (^4J) with the H(2) proton ($J_{H(4),H(2)}$ = 1.8—2.3 Hz). The structures of compounds **3a—c,e,f,i,j,m** were also confirmed by ^{13}C NMR spectra (Table 3). IR spectra were recorded for compounds **3a** and **3b** (see Experimental).

Scheme 2



Other compounds of the CH_3COCH_2X type generally react with chalcones in much the same manner as illustrated in Scheme 1. Thus, *N*-acetonylpyridinium salts (X = $C_5H_5N^+$) yield phenols (*via* elimination of pyridine from intermediate type **3** cyclohexenones),[5] while ethyl acetoacetate (X = COOEt) can give, depending on the reaction conditions, either Michael adducts, or 3-hydroxycyclohexanones (products of their intramolecular aldol condensation), or type **3** cyclohexenones when the latter lose a water molecule.[6,7]

Experimental

^{1}H and ^{13}C NMR spectra of compounds **3** were recorded on a Bruker AM-300 instrument (300.13 and 75.47 Hz, respectively) in $(CD_3)_2CO$ (^{1}H) and $(CD_3)_2SO$ (^{13}C). IR spectra were recorded on a Specord M-80 instrument (KBr). 2-Acetonyl-5-nitrotetrazole (**1a**) was obtained according to the known procedure.[8]

Synthesis of *N*-acetonylazoles 1b—d (general procedure). A solution of bromoacetone (1.5 g, 10% excess) in 6 mL of dichloroethane was added to a solution of the corresponding azole (0.02 mol), KOH (1.2 g), and Bu_4NBr (0.3 g) in 8 mL of water. The resulting emulsion was vigorously stirred at ~20 °C for 1.5 h. Then the precipitate that formed was filtered off, washed with ether and water, and dried. 1-Acetonyl-4-nitroimidazole (**1c**) was obtained in 86% yield. In the case of 1-acetonyl-3-nitro-1,2,4-triazole (**1b**) and 1-acetonylbenzotriazole (**1d**), additional amounts of the products can be isolated from the mother emulsion on keeping it in a refrigera-

Table 2. ^{1}H NMR spectra of compounds **3a—p** (δ, *J*/Hz)

Compound	H(2)	H(4)		H(5)	H(6)	Other protons*
		H_a(4)	H_b(4)			
3a	6.72 (d, *J* = 2.4)	3.47 (dd, *J* = 18.2, *J* = 5.0)	3.64 (ddd, *J* = 18.2, *J* = 11.1, *J* = 2.4)	4.43 (ddd, *J* = 13.5, *J* = 11.1, *J* = 5.0)	6.74 (d, *J* = 13.5)	7.2—7.35 (m, 3 H); 7.51 (m, 5 H); 7.82 (m, 2 H)
3b	6.20 (br.s)	2.91 (dd, *J* = 18.5, *J* = 5.0)	3.20 (ddm, *J* = 18.5, *J* = 11.5)	4.25 (ddd, *J* = 13.5, *J* = 11.5, *J* = 5.0)	6.63 (d, *J* = 13.5)	2.20 (s, 3 H, Me); 7.2—7.4 (m, 3 H); 7.45 (d, 2 H)
3c	6.40 (d, *J* = 1.9)	3.32 (ddd, *J* = 17.7, *J* = 12.2, *J* = 1.9)	3.55 (dd, *J* = 17.7, *J* = 4.7)	4.62 (ddd, *J* = 13.0, *J* = 12.2, *J* = 4.7)	6.51 (d, *J* = 13.0)	6.90 (dd, 1 H); 7.02 (d, 1 H, =C—H, *J* = 16.0); 7.07 (d, 1 H); 7.13 (dd, 1 H); 7.32 (d, 1 H); 7.41 (d, 1 H); 7.59 (d, 1 H); 7.65 (d, 1 H, =C—H, *J* = 16.0)
3d	6.66 (br.s)	3.58 (d, 2 H, *J* = 7.8)		4.44 (dt, *J* = 13.6, *J* = 7.8)	6.70 (d, *J* = 13.6)	7.20—7.35 (m, 4 H); 7.50 (d, 2 H); 7.81 (m, 2 H)
3e	6.60 (d, *J* = 2.3)	3.40 (dd, *J* = 18.3, *J* = 5.0)	3.56 (ddd, *J* = 18.3, *J* = 11.1, *J* = 2.3)	4.32 (ddd, *J* = 13.2, *J* = 11.1, *J* = 5.0)	6.07 (d, *J* = 13.2)	7.2—7.4 (m, 3 H); 7.50 (m, 5 H); 7.80 (m, 2 H); 8.56 (s, 1 H, H_{Az})
3f	6.61 (d, *J* = 2.2)	3.54 (ddd, *J* = 17.8, *J* = 11.2, *J* = 2.2)	3.66 (dd, *J* = 17.8, *J* = 5.1)	4.61 (ddd, *J* = 13.2, *J* = 11.2, *J* = 5.1)	5.85 (d, *J* = 13.2)	6.95 (dd, 1 H); 7.10 (d, 1 H); 7.27 (t, 1 H); 7.34 (d, 1 H); 7.80 (m, 2 H); 8.59 (s, 1 H, H_{Az})
3g	6.35 (d, *J* = 2.0)	3.23 (ddd, *J* = 17.7, *J* = 11.7, *J* = 2.0)	3.49 (dd, *J* = 17.7, *J* = 4.7)	4.48 (ddd, *J* = 13.0, *J* = 11.7, *J* = 4.7)	5.80 (d, *J* = 13.0)	6.93 (dd, 1 H); 6.98 (d, 1 H, =C—H, *J* = 16.0); 7.08 (d, 1 H); 7.12 (dd, 1 H); 7.31 (d, 1 H); 7.40 (d, 1 H); 7.60 (m, 2 H); 8.56 (s, 1 H, H_{Az})
3h	6.66 (d, *J* = 1.8)	3.50—3.60 (m, 2 H)		4.58 (ddd, *J* = 13.1, *J* = 9.9, *J* = 6.3)	5.87 (d, *J* = 13.1)	6.91 (dd, 1 H); 7.07 (d, 1 H); 7.30 (d, 1 H); 7.50 (m, 3 H); 7.80 (m, 2 H); 8.59 (s, 1 H, H_{Az})
3i	6.70 (d, *J* = 2.3)	3.38 (dd, *J* = 18.3, *J* = 5.1)	3.56 (ddd, *J* = 18.3, *J* = 10.8, *J* = 2.3)	4.26 (ddd, *J* = 13.6, *J* = 10.8, *J* = 5.1)	5.85 (d, *J* = 13.6)	7.20—7.40 (m, 3 H); 7.50—7.60 (m, 6 H); 7.80 (m, 2 H); 8.12 (d, 1 H, H_{Az}, *J* = 1.3)
3j	6.60 (br.s)	3.47 (d, 2 H, *J* = 8.0)		4.22 (dt, *J* = 13.4, *J* = 8.0)	5.80 (d, *J* = 13.4)	7.20—7.40 (m, 4 H); 7.50—7.60 (m, 3 H); 7.75 (d, 1 H); 7.80 (d, 1 H); 8.10 (d, 1 H, H_{Az}, *J* = 1.5)
3k	6.70 (d, *J* = 1.8)	3.50—3.60 (m, 2 H)		4.56 (ddd, *J* = 13.3, *J* = 9.4, *J* = 6.6)	5.65 (d, *J* = 13.3)	6.95 (dd, 1 H); 7.11 (d, 1 H); 7.36 (d, 1 H); 7.50—7.60 (m, 4 H); 7.82 (m, 2 H); 8.15 (d, 1 H, H_{Az}, *J* = 1.6)
3l	6.37 (d, *J* = 1.9)	3.18 (ddd, *J* = 18.0, *J* = 11.5, *J* = 1.9)	3.35 (dd, *J* = 18.0, *J* = 4.9)	4.13 (m)	5.78 (d, *J* = 13.4)	7.20—7.55 (m, 11 H); 7.68 (d, 2 H); 8.08 (br.s, 1 H, H_{Az})
3m	6.14 (d, *J* = 2.4)	2.63 (dd, *J* = 18.0, *J* = 4.5)	3.00 (ddd, *J* = 18.0, *J* = 11.7, *J* = 2.4)	4.33 (ddd, *J* = 13.5, *J* = 11.7, *J* = 4.5)	6.25 (d, *J* = 13.5)	0.95—1.05 (m, 4 H); 1.80—1.90 (m, 1 H) (all *cyclo*-C_3H_5); 7.00—7.15 (m, 3 H); 7.25—7.35 (m, 3 H); 7.41 (t, 1 H, H_{Az}); 7.66 (d, 1 H, H_{Az}); 7.85 (d, 1 H, H_{Az})
3n	6.73 (br.s)	3.52 (dd, *J* = 18.0, *J* = 5.2)	3.72 (br.dd, *J* = 18.0, *J* = 11.2)	4.82 (m)	6.61 (d, *J* = 13.3)	7.31 (t, 1 H, H_{Az}); 7.40—7.60 (m, 5 H); 7.73 (d, 1 H, H_{Az}); 7.80—8.00 (m, 5 H); 8.40 (br.s, 1 H)
3o	6.70 (d, *J* = 1.9)	3.39 (dd, *J* = 18.1, *J* = 5.0)	3.59 (ddd, *J* = 18.1, *J* = 11.3, *J* = 1.9)	4.69 (ddd, *J* = 13.6, *J* = 11.3, *J* = 5.0)	6.45 (d, *J* = 13.6)	7.00—7.20 (m, 3 H); 7.30 (t, 1 H, H_{Az}); 7.40—7.60 (m, 6 H); 7.72 (d, 1 H, H_{Az}); 7.80—7.90 (m, 4 H)
3p	6.63 (br.s)	3.50—3.70 (m, 2 H)		4.87 (ddd, *J* = 13.2, *J* = 11.6, *J* = 5.1)	6.33 (d, *J* = 13.2)	6.74 (dd, 1 H); 6.90 (d, 1 H); 7.12 (d, 1 H); 7.26 (t, 1 H); 7.33 (t, 1 H, H_{Az}); 7.47 (t, 1 H, H_{Az}); 7.69 (d, 1 H, H_{Az}); 7.80 (d, 2 H); 7.93 (d, 1 H, H_{Az})

* Assignments and coupling constants for the protons of the benzene and thiophene rings are not given.

Table 3. ^{13}C NMR spectra of compounds **3a—c,e,f,i,j,m** (δ)

Compound	C(1)	C(2)	C(3)	C(4)	C(5)	C(6)	Other C*
3a	189.4	122.7	161.5	36.7	46.6	73.7	127.2, 128.1, 128.5, 129.4, 131.5, 137.8, 138.9, 166.6 (C_{Az})
3b	189.0	124.3	165.9	39.5	46.5	73.6	23.9 (Me), 127.9, 128.4, 129.4, 138.9, 166.3 (C_{Az})
3c	188.6	123.7	158.5	33.4	40.4	74.1	125.4, 126.1, 126.4, 127.0, 128.5, 128.9, 130.6, 131.5, 141.0, 141.1, 165.7 (C_{Az})
3e	190.8	123.2	160.8	36.7	46.6	69.7	127.1, 128.1, 128.2, 129.4, 129.5, 131.3, 138.1, 139.8, 148.1 (C_{Az}), 163.1 (C_{Az})
3f	189.5	120.3	153.3	36.6	41.3	70.8	125.5, 126.4, 127.6, 129.5, 130.4, 131.4, 141.5, 142.3, 148.3 (C_{Az}), 163.2 (C_{Az})
3i	192.3	123.1	159.8	37.0	47.0	67.0	121.0 (C_{Az}), 126.9, 128.0, 129.2, 129.3, 131.1, 137.7, 137.9, 139.7, 147.8 (C_{Az})
3j	192.1	120.1	153.0	36.8	45.8	66.6	121.9 (C_{Az}), 128.1, 129.3, 129.7, 130.7, 131.8, 138.3 (C_{Az}), 139.7, 141.2, 148.1 (C_{Az})
3m	191.2	122.0	169.6	35.7	46.5	67.3	8.2, 9.1, 18.2 (all — *cyclo*-C_3H_5); 110.9, 119.6, 123.8 (all — C_{Az}); 127.3, 127.5, 128.0, 128.7, 134.6 (C_{Az}), 140.7, 145.8 (C_{Az})

* Assignments for the carbon atoms of the benzene and thiophene rings are not given.

Table 4. Elemental analysis data and melting points for compounds **3a—p**

Compound	M.p./°C (solvent)	Found / Calculated (%)				Molecular formula
		C	H	N	S	
3a	143—145 (EtOH)	62.96 63.15	4.09 4.18	19.66 19.38		$C_{19}H_{15}N_5O_3$
3b	165—166 (EtOH)	56.11 56.18	4.10 4.38	23.44 23.40		$C_{14}H_{13}N_5O_3$
3c	183—186 (MeCN)	50.88 51.12	3.44 3.28	17.80 17.53	15.71 16.05	$C_{17}H_{13}N_5O_3S_2$
3d	163—165 (MeCN)	56.01 55.58	3.50 3.57	18.74 19.06	9.12 8.73	$C_{17}H_{13}N_5O_3S$
3e	167—168 (EtOH)	67.00 66.66	4.08 4.48	15.53 15.55		$C_{20}H_{16}N_4O_3$
3f	181—183 (EtOH)	51.79 51.60	3.11 3.25	15.00 15.04	17.32 17.22	$C_{16}H_{12}N_4O_3S_2$
3g	187—189 (MeCN)	53.95 54.26	3.55 3.54	14.22 14.06	15.90 16.09	$C_{18}H_{14}N_4O_3S_2$
3h	175—177 (MeCN)	58.77 59.01	4.01 3.85	15.02 15.29	8.95 8.75	$C_{18}H_{14}N_4O_3S$
3i	212—213 (MeCN)	70.30 70.18	4.60 4.77	11.43 11.69		$C_{21}H_{17}N_3O_3$
3j	209—211 (MeCN)	62.59 62.45	3.88 4.14	11.65 11.50	9.08 8.77	$C_{19}H_{15}N_3O_3S$
3k	218—221* (MeCN)	62.42 62.45	4.03 4.14	11.44 11.50	9.02 8.77	$C_{19}H_{15}N_3O_3S$
3l	245—248* (MeCN)	77.39 71.68	5.10 4.97	11.13 10.90		$C_{23}H_{19}N_3O_3$
3m	186—187 (EtOH)	76.94 76.57	5.91 5.81	12.76 12.76		$C_{21}H_{19}N_3O$
3n	234—236* (MeCN)	70.04 70.23	4.49 4.42	13.90 13.65		$C_{24}H_{18}N_4O_3$
3o	223—224 (MeCN)	79.11 78.88	5.19 5.24	11.78 11.50		$C_{24}H_{19}N_3O$
3p	230—231* (MeCN)	63.60 63.64	4.15 4.01	10.81 11.13	17.18 16.99	$C_{20}H_{15}N_3OS_2$

* Decomposes when melted.

tor for 1 h. The overall yields of **1b** and **1d** were 71% and 55%, respectively. Until recently, data on compound **1d** have not been published.*

1-Acetonylbenzotriazole (1d), m.p. 123—124 °C (recrystallized from ethanol or toluene) (*cf.* Ref. 9: m.p. 56—57 °C (recrystallized from toluene)). Found (%): C, 61.98; H, 5.05; N, 24.07. $C_9H_9N_3O$. Calculated (%): C, 61.70; H, 5.18; N, 23.99. ^{1}H NMR, δ: 2.42 (s, 3 H, CH_3); 5.76 (s, 2 H, CH_2); 7.40 (t, 1 H, J = 8.35 Hz); 7.51 (t, 1 H, J = 8.35 Hz); 7.66 (d, 1 H, J = 8.3 Hz); 8.01 (d, 1 H, J = 8.4 Hz) (all H_{arom}). Prior to recrystallization, **1d** contains a minor admixture (5%), and its ^{1}H NMR spectrum exhibits additional signals: 2.23 (s, 3 H); 5.70 (s, 2 H); 7.43 (m, partially overlapped with a signal of **1d** at δ 7.40); 7.78 (m, 2 H) (integral intensities of signals for the protons of the impurity are indicated with respect to each other). The presence of the admixture lowers the melting point of nonrecrystallized compound **1d** to 118—119 °C. The ^{1}H NMR spectrum suggests that the admixture is 2-acetonylbenzotriazole (*cf.* Ref. 10).

Synthesis of cyclohexenones 3 (general procedure). NaOEt was added to a boiling solution of *N*-acetonylazole **1** (5 mmol) and chalcone **2** (5 mmol) in 5 mL of EtOH (15 mL of EtOH in the case of **3i—l** and **3n** because of the poor solubility of **1c** and (3-nitrobenzylidene)acetophenone, respectively), and the reaction mixture was refluxed with stirring. To isolate compounds **3a,b,e,f,m**, the reaction mixture was then cooled to 20 °C, and the precipitate that formed was filtered off and recrystallized from EtOH. In all the other cases, the reaction mixture was filtered hot, and the precipitate was washed with 5 mL of hot EtOH and dried. The required amount of NaOEt, the reaction time, and the yields of compounds **3a—p** are given in Table 1. Melting points and elemental analysis data are given in Table 4.

* When this work was already performed, this compound was described in the paper[9] (the procedure of synthesis is somewhat different from ours). However, the melting point cited in that paper is much lower than that found by us.

6-(5-Nitrotetrazol-2-yl)-3,5-diphenylcyclohex-2-enone (3a). IR (ν/cm^{-1}): 1675 (C=O), 1600 (C=C), 1560, 1320 (NO_2).

3-Methyl-6-(5-nitrotetrazol-2-yl)-5-phenylcyclohex-2-enone (3b). IR (ν/cm^{-1}): 1680 (C=O), 1630 (C=C), 1565, 1320 (NO_2).

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