IIId, 105335-91-9; IIIe, 105335-92-0; IIIf, 105335-93-1.

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Reaction of 1-Benzyl-1*H*-triazole-4,5-dicarboxaldehyde with Cyclic Ketones

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Three 4-hydroxy-1-benzyl-4,5-dihydro-5,7-polymethyleno-6(2*H*)-cycloheptatriazolones (IVa–c) and 1-benzyl-5,7-dodecano-6(2*H*)-cycloheptatriazolone (III) were synthesized by the aldol-type condensation reaction of 1-benzyl-1*H*-triazole-4,5-dicarboxaldehyde (I) with cyclododecanone (IIa), cycloundecanone (IIb), cyclododecanone (IIc), and cyclopentadecanone (IId), respectively.

Azides have been considered as one of the most important precursors for the synthesis of triazoles by their reactions with substituted acetylenes. As part of our continuous interest in the syntheses and reactions of triazole derivatives (*1*, *2*). I report here details of the condensation reaction of 1-benzyl-1*H*-triazole-4,5-dicarboxaldehyde (I) with cyclic ketones (IIa–d) to form polymethylene-bridged 1-benzylcycloheptatriazolones. Bridged cycloheptatriazolone (III) was obtained when I was condensed under aldol condensation reaction condition with cyclopentadecanone (IId). But when I was condensed with IIa–c under the same condition, the corresponding hydroxycycloheptatriazolones (IVa–c) were obtained (Scheme I). This I:I cyclic addition is closely analogous to known cycloaddition between phthalaldehyde and cyclic ketones (*3*, *4*).

Scheme I

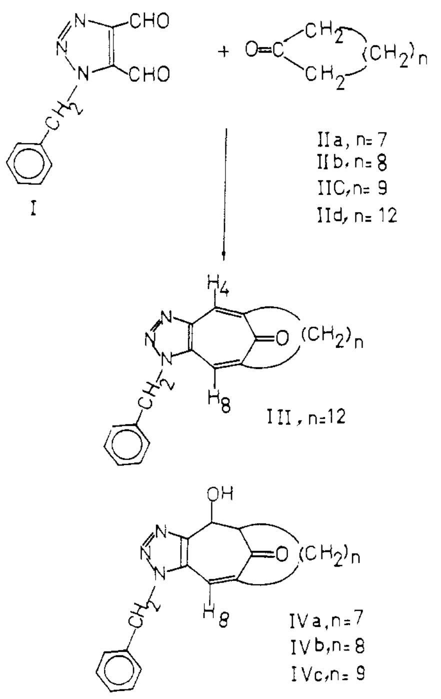

Table I. Physical Data and NMR Spectra of Prepared Compounds[a]

compd	mp, °C	reflux time, h	yield, %	^{1}H NMR[b] ppm	^{1}H NMR[b] assignments
III	205–207	8	60	7.75	(H_4, H_8, 2 H, S)
				7.22	(C_6H_k–, 5 H, S)
				5.75	(C_6H_5–CH_2–, 2 H, m)
				2.60–3.0	(C═C—CH_2, 4 H, m)
				0.85–1.65	(–CH_2–, 20 H, m)
IVa	197–199	9	54	7.28	(C_6H_5–, 5 H, S)
				6.85	(H_8, 1 H, S)
				5.70	(–*OH*, 1 H, S)
				5.55	(C_6H_5–CH_2–, 2 H, S)
				4.80	(>C(OH)H, 1 H, m)
				3.15	(H_5, 1 H, m)
				0.9–1.90	(–CH_2–, 14 H, m)
IVb	205–207	2.5	72	7.27	(C_6H_5–, 5 H, S)
				6.90	(H_8, 1 H, S)
				5.68	(–*OH*, 1 H, S)
				5.52	(C_6H_5–CH_2–, 2 H, S)
				4.60	(>C(OH)H, 1 H, m)
				3.17	(H_5, 1 H, m)
				2.36–2.63	(–CH_2–, 2 H, m)
				2.0–2.20	(–CH_2–, 2 H, m)
				1.0–1.35	(–CH_2–, 12 H, m)
IVc	217–218	2	71	7.20	(C_6H_5–, 5 H, S)
				6.90	(H_8, 1 H, S)
				5.70	(–OH, 1 H, S)
				5.57	(C_6H_5–CH_2–, 2 H, S)
				4.68	(>C(OH)H, 1 H, m)
				3.25	(H_5, 1 H, m)
				2.45–2.65	(–CH_2–, 2 H, m)
				2.11–2.20	(–CH_2–, 2 H, m)
				0.90–1.30	(–CH_2–, 14 H, m)

[a] Elemental analysis, (C, H, N) in agreement with theoretical values were obtained and submitted for review. [b] $CDCl_3$–Me_2SO-d_6 was used as the NMR solvent for all compounds except $CDCl_3$ used for compound III. 4-Hydroxy-1-benzyl-4,5-dihydro-5,7-heptano-6(2*H*)-cycloheptatriazolone (IVa). 4-Hydroxy-1-benzyl-4,5-dihydro-5,7-octano-6(2*H*)-cycloheptatriazolone (IVb). 4-Hydroxy-1-benzyl-4,5-dihydro-5,7-nonano-6(2*H*)-cycloheptatriazolone(IVc).

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Experimental Section

1-Benzyl-1*H*-triazole-4,5-dicarboxaldehyde (I) was prepared according to the procedure reported by Henkel and Weygand (*5*). The cyclic ketones were prepared according to the methods of Blomquist (*6*, *7*). Melting points were determined with a Thomas-Hoover Unimelt instrument and are uncorrected. NMR spectra were recorded on a Varian A-60 spectrometer using tetramethylsilane as an internal reference and shifts (δ) are reported in ppm.

1-Benzyl-5,7-dodecano-6(2H)-cycloheptatriazolone (III). A solution of 2.15 g (0.01 mol) of 1-benzyl-1*H*-triazole-4,5-dicarboxaldehyde and 2.24 g (0.01 mol) of cyclopentadecanone in 0.3 g of KOH and 50 mL of methanol was heated under reflux. After the mixture was cooled, the product was collected and then recrystallized from ethanol.

4-Hydroxy-1-benzyl-4,5-dihydro-5,7-polymethyleno-6-(2H)-cycloheptatriazolones (IVa–c). In the same manner 2.15 g (0.01 mol) of 1-benzyl-1*H*-triazole-4,5-dicarboxaldehyde and 0.01 mol of the corresponding cyclic ketones in 0.4 g of KOH and 60 mL of methanol was heated under reflux. The solvent was removed under pressure and the resulting product was recrystallized from chloroform–petroleum ether.

The melting points, reflux time, yields, and NMR data for compounds III and IVa–c are listed in Table I.

Registry No. I, 103532-75-8; IIa, 1502-06-3; IIb, 878-13-7; IIc, 830-13-7; IId, 502-72-7; III, 105309-39-5; IVa, 105309-40-8; IVb, 105309-41-9; IVc, 105309-42-0.

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Synthesis of Some New 2,4,6-Triarylpyridines Using Phenacylidenedimethylsulfuranes

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Phenacylidenedimethylsulfurane and *p*-chlorophenacylidenedimethylsulfurane were reacted with α,β-unsaturated ketones in the presence of ammonium acetate in glacial acetic acid to give 2,4,6-triarylpyridines in 45–70% yields. The structures of the pyridines were confirmed by IR and NMR spectra.

In continuation of our earlier researches (*1–6*) on the synthetic potentialities of ylides, we now report herein the aza ring closure reaction of two phenacylidenedimethylsulfuranes with a wide variety of α,β-unsaturated ketones with a view to test the domain of applicability of these ylides.

Experimental Section

Phenacyldimethylsulfonium bromide (**1a**) and *p*-chlorophen-

Table I. Physical and Spectral Data of Compounds[a]

compd	R_1	R_2	R_3	yield, %	recryst solvent	mp, °C	IR (KBr),[b] cm^{-1} ν(Ar–H)	ν(C=C)	ν(C=N)	ϕ(C–H)	NMR[c] data ($CDCl_3$) δ, ppm
5a	H	H	4-FC_6H_4	45	a	150–52	3150	1605	1500	995	
5b	H	3,4-O_2CH_2	4-FC_6H_4	60	b	160–62	3010	1610	1508	995	5.95 (s, 2H, –O_2CH_2); 6.40–8.00 (m, 14 H, Ar–H)
5c	H	3,4-di-OCH_3	4-FC_6H_4	60	a	120–22	3000	1608	1500	992	3.70–7.75 (d (J = 5 Hz), 6 H, diOCH_3) 6.58–7.95 (m, 14 H, ArH)
5d	4-Cl	H	4-FC_6H_4	55	b	168–70	3060	1600	1505	1005	
5e	4-ClC_6H_4	4-$CH_3OC_6H_4$	C_6H_5	50	c	95–96	3030	1510	1520	986	3.70 (s, 3 H, OCH_3); 6.40–8.20 (m, 15 H, ArH)
5f	4-ClC_6H_4	3-NO_2–C_6H_4	C_6H_5	65	d	90–92	3060	1595	1005	1005	
5g	4-ClC_6H_4	4-CH_3O–C_6H_4	4-$NO_2C_6H_4$	60	a	82–85	3040	1598	1510	988	3.75 (s, 3 H, OCH_3); 6.50–8.30 (m, 14 H, ArH)
5h	4-ClC_6H_4	3-$NO_2C_6H_4$	4-$CH_3OC_6H_4$	65	c	140–42	3030	1585	1510	985	
5i	4-ClC_6H_4	2-C_4H_3S	3-$CH_3OC_6H_4$	70	f	70–72	3045	1590	1520	975	
5j	4-ClC_6H_4	2-C_4H_3S	4-ClC_6H_4	60	e	128–30	3015	1588	1510	1010	
5k	4-ClC_6H_4	2-C_4H_3O	4-ClC_6H_4	60	a	126–28	3050	1615	1500	980	
51	4-ClC_6H_4	3-$NO_2C_6H_4$	4-ClC_6H_4	65	c	84–85	3080	1600	1520	985	
5m	4-ClC_6H_4	4-$CH_3OC_6H_4$	2-C_4H_3O	60	c	96–98	3025	1618	1516	990	3.85 (s, 3 H, OCH_3); 7.15–8.25 (m, 13 H, ArH)
5n	4-ClC_6H_4	4-FC_6H_4	2-C_4H_3S	60	a	50–55	3050	1590	1500	980	

[a] All compounds gave satisfactory elemental analysis for C, H, N. [b] IR spectra were recorded on Perkin Elmer Infracord spectrometer using KBr phase. [c] NMR spectra were run on Varian A-60 spectrometer using Me_4Si as internal standard. ν = stretching vibratioins, ϕ = bending vibrations (out-of-plane vibrations), s = singlet, m = multiplet. Recrystallization solvent: a = C_5H_5N–MeOH; b = EtOH; C = C_5H_5N–EtOH; d = C_6H_6–AcOEt; e = C_5H_5N–$CHCl_3$, f = $CHCl_3$–C_6H_6.

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