9e were formed (Scheme I). 1e gave 8e and 9e on heating the reaction mixture and 8e, 9e, and 3e at room temperature. On the other hand, 9f, 6g, 8f, and 3f were obtained from 1g under hot condition; 2g and 3f were formed when the same reaction was performed at room temperature. 1h gave 9h, whereas, at room temperature the reaction products were 2h, 3l, in addition to 6h and 9h. In the case of 1l, 8l was the only product. 1j gave 3k, 8k, 8l, 9l, and 6j when the reaction mixture was heated for 3 h. Authentic samples of 2a, 8b, 6a, 3e, 8e, 3f, 6f, 3l, 8l, and 6h were prepared by the methods reported earlier (3).

Experimental Section

Melting points are uncorrected. Reaction mixtures were separated by column chromatography using silica gel and the purity was checked by thin-layer chromatography (TLC).

Reaction of 2'-Hydroxychalcone Dibromides (1a) with Me_2SO. (*a*) The chalcone dibromide (1a) in Me_2SO (20 mL) was heated on a water bath for 3 h and the reaction mixture was allowed to cool to room temperature. It was diluted with water and the precipitate was filtered off, washed with water, and dried. TLC showed six spots. By column chromatography 8b, 9c, 3b, 6a, and 2a were separated using petroleum ether, benzene, and other as eluotropic solvents.

(*b*) A mixture of the chalcone dibromide (1a) and Me_2SO (20 mL) was kept at room temperature for 5 days. It was then worked up as described above to give 3b, 2a, and 6a.

Similarly, the reactions of other chalcone dibromides (1b–1) with Me_2SO were carried out and data of the reaction products are given in Table I. Satisfactory C, H, and halogen analyses were obtained for all the compounds.

Registry No. 1a, 39729-11-8; 1b, 35820-37-2; 1c, 10372-55-1; 1d, 22219-26-7; 1e, 29976-68-9; 1f, 102260-70-8; 1g, 75227-44-0; 1h, 22129-40-4; 1l, 29976-70-3; 1j, 43016-14-4; 1k, 102260-69-5; 1l, 75767-98-5; 2a, 1214-47-7; 2g, 16635-10-2; 2h, 16635-13-5; 3b, 1218-22-0; 3e, 29976-64-5; 3f, 102260-59-3; 3l, 29976-66-7; 3k, 102260-60-6; 6a, 487-26-3; 6g, 14166-16-6; 6h, 102260-61-7; 8e, 29976-76-9; 8f, 102260-62-8; 8k, 102260-63-9; 9b, 102260-64-0; 9c, 102260-65-1; 9e, 102260-66-2; 9f, 102260-67-3; 9h, 72149-92-9; 9l, 102260-68-4; Me_2SO, 67-68-5.

Literature Cited

(1) Epstein, W. W.; Sweat, F. W. *Chem. Rev.* **1967**, *67*, 247.
(2) Kornblum, N.; Powers, J. W.; Anderson, G. J.; Jones, W. J.; Larson, H. W.; Livand, O.; Weaver, W. N. *J. Am. Chem. Soc.* **1957**, *79*, 6562.
(3) Reddy, N. J.; Bokadia, M. M.; Sharma, T. C.; Donnelly, J. A. *J. Org. Chem.* **1981**, *46*, 638.
(4) Feurestein, W.; Kostanecki, St. Von. *Chem. Ber.* **1898**, *37*, 715.
(5) Chi-Hsiunglin; Chiu-Tsudin; Chin Lung Yeh. *J. Chem. Soc. (Taipei)* **1967**, *14* (1), 9.
(6) Sharma, T. C.; Pawar, S. R.; Reddy, N. J.; Donnelley, J. A. *Indian J. Chem.* **1981**, *20B*, 80.
(7) Mahanthy, P.; Panda, S. P.; Sabata, B. K.; Raut, M. K. *Indian J. Chem.* **1965**, *3*, 121.
(8) Ghiya, B. J.; Marathey, M. G. *J. Sci. Ind. Res. (India)* **1962**, *21B*, 28.
(9) Mahal, H. S.; Venkataraman, K. *J. Chem. Soc.* **1935**, 866.
(10) Fitzgerald, D. M.; O'Sullivan, F. J.; Philbin, E. M.; Wheeler, T. S. *J. Chem. Soc.* **1955**, 860.
(11) Wadodkar, P. N. *Indian J. Chem.* **1963**, *1*, 163.
(12) Kutumbey, K. R.; Marathey, M. G. *Chem. Ber.* **1963**, *96*, 913.

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Reaction of Acetylenedicarboxaldehyde Bis(diethyl acetal) with Bis(azidomethyl)benzene

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Three 1,1'-[phenylenebis(methylene)]bis(triazole-4,5-dicarboxaldehyde) tetrakis(ethyl acetals) (IIIa–c) were synthesized by the condensation of acetylenedicarboxaldehyde bis(diethyl acetal) with 1,2-, and 1,3-, and 1,4-bis(azidomethyl)benzene.

In continuation of our previous work on the condensation reaction of 1-benzyl-1*H*-triazole-4,5-dicarboxaldehyde with cyclic ketones to form polymethylene-bridged benzyl triazolotropones (*1*), we report in the present paper details on the preparation of the three bis(azidomethyl)benzenes (IIa–c) and on their reactions with acetylenedicarboxaldehyde bis(diethyl acetal) to give the phenylenebis(methylene)bis(triazole-4,5-dicarboxaldehyde) tetrakis(ethyl acetals) (IIIa–c). (See Scheme I.)

Experimental Section

Acetylenedicarboxaldehyde bis(diethyl acetal) was prepared from acetylene gas and triethyl orthoformate by the method described by Whol (*2*). Melting points were determined by using a Thomas-Hoover Unimelt instrument and are uncorrected. The nuclear magnetic resonance spectra were taken on a Varian A-60 spectrometer using tetramethylsilane as an internal reference, and shifts (δ) are reported in ppm.

Preparation of the Bis(azidomethyl)benzene (IIa–c). To a solution of 13.0 g (0.2 mol) of sodium azide in 70 mL of water and 70 mL of methanol was added 17.5 g (0.1 mol) of the bis(chloromethyl)benzene. The mixture was heated in a bomb flask at 100 °C for 2 days. The methanol was removed on a rotary evaporator at diminished pressure. The residue was

0021-9568/86/1731-0379$01.50/0

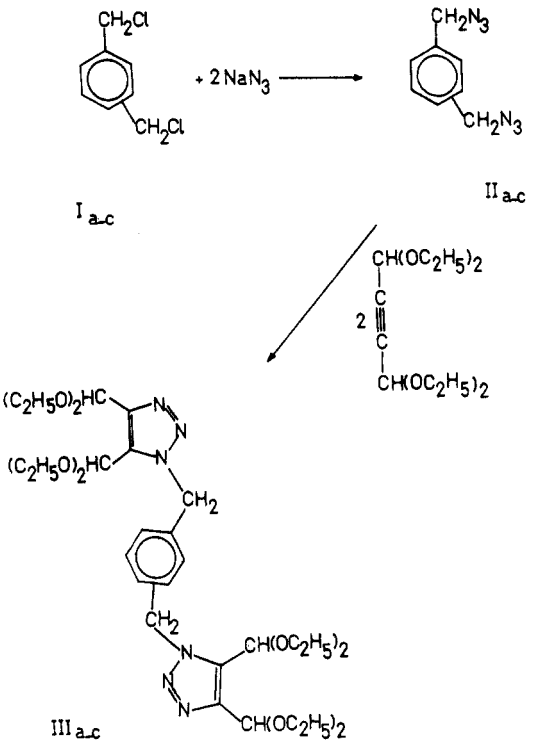
Table I. Physical Data and NMR Spectra of Prepared Compounds[a]

compd	mp, °C	bp, °C/mmHg	yield, %	^{1}H NMR ($CDCl_3$) ppm	assignment
IIa		112–115/1	88	7.22	(aromatic, 4 H, s)
				4.26	(ArCH_2–, 4 H, s)
IIb		118–120/1.5	88	7.18	(aromatic, 4 H, s)
				4.12	(ArCH_2–, 4 H, s)
IIc	30–32	118–120/1	92	7.22	(aromatic, 4 H, s)
				4.21	(ArCH_2–, 4 H, s)
IIIa		160–164/0.001	73	7.18	(aromatic, 4 H, s)
				5.99	(>C–H, 2 H, s)
				5.78	(ArCH_2–, 4 H, s)
				5.65	(>C–*H*, 2 H, s)
				3.25–4.0	(–O$CH_2$$CH_3$, 16 H, m)
				0.95–1.35	(–O$CH_2$$CH_3$, 24 H, m)
IIIb		160–163/0.001	78	7.20	(aromatic, 4 H, s)
				5.90	(>C–*H*, 2 H, s)
				5.75	(ArCH_2–, 4 H, s)
				5.65	(>C–*H*, 2 H, s)
				3.25–3.90	(–O$CH_2$$CH_3$, 16 H, m)
				0.95–1.35	(–O$CH_2$$CH_3$, 24 H, m)
IIIc	70–71[b]		43	7.28	(aromatic, 4 H, s)
				5.89	(>C–*H*, 2 H, s)
				5.73	(ArCH_2–, 4 H, s)
				5.65	(>C–*H*, 2 H, s)
				3.2–3.9	(–O$CH_2$$CH_3$, 16 H, m)
				0.95–1.35	(–O$CH_2$$CH_3$, 24 H, m)

[a] Elemental analyses (C, H, N) in agreement with theoretical values were obtained and submitted for review. [b] Analytical sample, recrystallized from petroleum ether. 1,2-Bis(azidomethyl)benzene (IIa). 1,3-Bis(azidomethyl)benzene (IIb). 1,4-Bis(azidomethyl)benzene (IIc). 1,1′-[1,2-phenylenebis(methylene)]bis(triazole-4,5-dicarboxyaldehyde) tetrakis(ethyl acetal) (IIIa). 1,1′-[1,3-phenylenebis(methylene)]bis(triazole-4,5-dicarboxaldehyde) tetrakis(ethyl acetal) (IIIb). 1,1′-[1,4-phenylenebis(methylene)]bis(triazole-4,5-dicarboxyaldehyde) tetrakis(ethyl acetal) (IIIc).

extracted three times with diethyl ether. The extracts were combined and dried over anhydrous $CaCl_2$. The ether was

Scheme I



evaporated, and the remaining liquid was cautiously distilled to give the product. The boiling points, yields, and NMR data are listed in Table I.

Preparation of the 1,1′-[Phenylenebis(methylene)]bis-(triazole-4,5-dicarboxaldehyde) Tetrakis(ethyl acetals) (IIIa–c). To a solution of 13.8 g (0.06 mol) of acetylenedicarboxaldehyde bis(diethyl acetal) in 15 mL of absolute ethanol was added 5.64 g (0.03 mol) of the corresponding bis(azidomethyl)benzene. The resulting mixture was heated in a bomb flask at 90 °C for 30 h. The alcohol was removed under reduced pressure. The resulting product was either distilled or recrystallized from petroleum ether. The boiling points, melting points, yields, and NMR data are listed in Table I.

Registry No. Ia, 612-12-4; Ib, 626-16-4; Ic, 623-25-6; IIa, 102437-79-6; IIb, 102437-80-9; IIc, 102437-81-0; IIIa, 102437-82-1; IIIb, 102437-83-2; IIIc, 102535-03-5; $(EtO)_2CHC{\equiv}CCH(OEt)_2$, 3975-08-4.

Literature Cited

(1) Abu-Orabi, S. T.; Harmon, R. E. *J. Heterocycl. Chem.*, in press.
(2) Wohl, A. *Chem. Ber.* **1912**, *45*, 339.

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Correction

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The correct spelling of the second author's name is Mohamed F. Hamoda, not Mahmoud F. Hamoda, as printed.