Table 3. *Hydrogen-bond lengths* (Å) *and angles* (°)

The e.s.d.'s are in parentheses.

N–H···O	N···O	H···O	N–H	∠N–H···O
N(1)–H(7)···O(1)	2·822 (3)	1·83 (3)	1·01 (3)	166 (2)
N(1)–H(8)···O(1)	2·810 (3)	1·89 (3)	0·92 (3)	173 (2)
N(1)–H(9)···O(2)	2·904 (3)	2·01 (3)	0·92 (3)	165 (2)

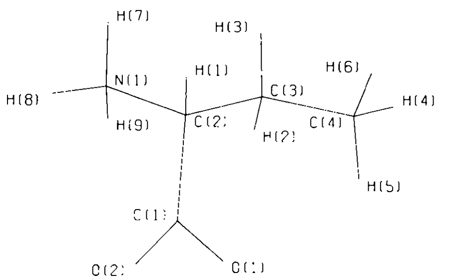

Fig. 1. Numbering of the atoms.

Discussion. The structure of the title compound was first reported by Ichikawa & Iitaka (1968). It was based on 630 reflexions from Weissenberg photographs, and yielded a final *R* of 0·12. The present structure determination is more accurate. The main difference between the atomic coordinates given in this paper and those of Ichikawa & Iitaka is that all the *y* coordinates have opposite signs. With our coordinates we can reproduce all interatomic distances and hydrogen bonds as given by Ichikawa & Iitaka. The cell dimensions, bond lengths and bond angles differ only slightly. For a detailed description of the structure we refer to their paper.

We thank Drs R. Olthof-Hazekamp for assistance in the use of the XRAY system.

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Methyl (*Z*)-4-Oxo-1,3-diphenyl-2-phenylimino-5-imidazolidinylideneacetate

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Abstract. $C_{24}H_{19}N_3O_3$, orthorhombic, *Pcab*, a = 10·97 (1), b = 14·99 (1), c = 25·66 (1) Å, Z = 8, μ = 0·093 mm^{-1} (Mo $K\alpha$ radiation), D_m = 1·28, D_c = 1·25 Mg m^{-3}, final R = 0·054. The product from 1,2,3-triphenylguanidine and dimethyl acetylenedicarboxylate possesses a five-membered-ring structure and not the six-membered-ring structure previously assigned.

Introduction. The 1:1 molar MeOH adducts produced by the reaction of acyclic guanidines with dimethyl acetylenedicarboxylate (DMAD) have been given both the imidazolidin-4-one structure (1) (Sasaki, Sakata & Iwanami, 1964) and the pyrimid-4-one structure (2) (Ruhemann & Stapleton, 1900), although the structural assignments were often unsupported. Some chemical degradative evidence favours the five-membered-ring structure (3) for the adduct from guanidine itself and DMAD (Katner & Zeige, 1971) but, in contrast, the adduct from 1,2,3-triphenylguanidine has been assigned the six-membered-ring structure (4), principally from its high-resolution mass

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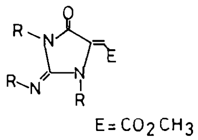
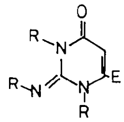
spectrum (Lown & Ma, 1967). The crystal structure of the adduct from 1,2,3-triphenylguanidine was therefore examined to gain some unambiguous information in this confused area.



(1)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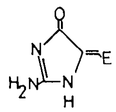
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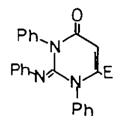

(4)

The adduct was prepared in 71% yield from 1,2,3-triphenylguanidine and DMAD in methanol (Lown & Ma, 1967), m.p. 490–492 K (found: C, 72·4; H, 4·8; N, 10·7%; calculated for $C_{24}H_{19}N_3O_3$: C, 72·5; H, 4·8; N, 10·6%); the published spectra were reproduced.

Crystals were grown by evaporation of an ethyl acetate solution, and preliminary oscillation and Weissenberg photographs showed the crystal system to be orthorhombic. Cell dimensions, determined on an Enraf–Nonius CAD-4 four-circle diffractometer, and the density showed $Z = 8$. Intensities were collected with Mo $K\alpha$ radiation by an $\omega/2\theta$ scan with standard reflections checked hourly. The space group was found to be *Pcab* from systematic absences ($0kl$: l = odd, $h0l$: h = odd, $hk0$: k = odd). Lorentz and polarization corrections were applied to the 2232 observed reflections, equivalent reflections merged and structure amplitudes derived for the 1675 with $I > 3\sigma(I)$. The structure was solved with *MULTAN* 77 (Main, Lessinger, Woolfson, Germain & Declercq, 1977) and refined by full-matrix least squares to $R = 0{\cdot}138$ with isotropic temperature factors. H atom positions were calculated and assigned isotropic temperature factors of 0·085 Å^2. Non-hydrogen atoms were then refined with anisotropic temperature factors by blocked-matrix least squares until convergence at $R = 0{\cdot}054$. Weights were computed from the Chebyshev series $w = [131{\cdot}96t_0(x) + 175{\cdot}71t_1(x) + 52{\cdot}54t_2(x) + 8{\cdot}73t_3(x)]^{-1}$ where $(x) = F_o/F_{max}$ (Carruthers & Watkin, 1979). Data reduction and refinement were performed with *CRYSTALS* (Carruthers, 1975) and all calculations were made on the Oxford University ICL 1906A computer.

Atomic coordinates are given in Table 1.*

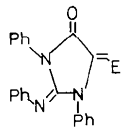
* Lists of structure factors and anisotropic thermal parameters have been deposited with the British Library Lending Division as Supplementary Publication No. SUP 35566 (15 pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

Table 1. *Final atomic coordinates with e.s.d.'s in parentheses*

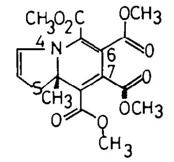
$$B_{eq} = 8\pi^2(U_{11}U_{22}U_{33})^{1/3}.$$

	x	y	z	B_{eq} (Å^2)
O(7)	0·4380 (4)	0·2560 (3)	0·1788 (1)	6·1
O(10)	0·7059 (3)	0·1942 (3)	0·0220 (1)	4·8
O(11)	0·5238 (4)	0·1738 (4)	−0·0131 (1)	6·4
N(1)	0·6674 (4)	0·1079 (3)	0·1356 (1)	3·6
N(3)	0·5862 (4)	0·1569 (3)	0·2112 (1)	3·7
N(6)	0·7505 (4)	0·0426 (3)	0·2110 (1)	4·1
C(11)	0·7347 (4)	0·0485 (3)	0·1008 (2)	3·5
C(12)	0·6741 (5)	−0·0112 (4)	0·0676 (2)	4·0
C(13)	0·7406 (6)	−0·0670 (4)	0·0329 (2)	4·6
C(14)	0·8670 (6)	−0·0641 (4)	0·0343 (2)	5·4
C(15)	0·9266 (5)	−0·0047 (5)	0·0687 (2)	5·2
C(16)	0·8613 (5)	0·0519 (4)	0·1024 (2)	4·5
C(2)	0·6764 (4)	0·0976 (3)	0·1907 (2)	3·6
C(31)	0·5771 (4)	0·1866 (3)	0·2647 (2)	3·5
C(32)	0·6591 (5)	0·2514 (4)	0·2829 (2)	5·1
C(33)	0·6473 (7)	0·2858 (5)	0·3328 (3)	7·1
C(34)	0·5570 (7)	0·2553 (6)	0·3635 (2)	6·6
C(35)	0·4751 (7)	0·1897 (6)	0·3466 (2)	6·7
C(36)	0·4841 (5)	0·1541 (4)	0·2955 (2)	5·4
C(4)	0·5198 (5)	0·2004 (4)	0·1725 (2)	4·4
C(5)	0·5750 (5)	0·1688 (3)	0·1226 (2)	3·8
C(61)	0·7581 (5)	0·0246 (3)	0·2649 (2)	3·6
C(62)	0·8556 (5)	0·0617 (4)	0·2938 (2)	5·0
C(63)	0·8700 (6)	0·0357 (5)	0·3452 (2)	6·0
C(64)	0·7929 (6)	−0·0263 (5)	0·3686 (2)	5·7
C(65)	0·6977 (6)	−0·0632 (4)	0·3406 (2)	5·8
C(66)	0·6804 (5)	−0·0376 (4)	0·2883 (2)	5·2
C(8)	0·5350 (3)	0·2030 (4)	0·0767 (2)	4·7
C(9)	0·5849 (5)	0·1874 (4)	0·0244 (2)	4·3
C(10)	0·7621 (6)	0·1734 (5)	−0·0275 (2)	5·9
H(121)	0·5829	−0·0143	0·0680	
H(131)	0·6989	−0·1087	0·0085	
H(141)	0·9159	−0·1024	0·0107	
H(151)	1·0208	−0·0040	0·0700	
H(161)	0·9047	0·0938	0·1256	
H(321)	0·7266	0·2721	0·2592	
H(331)	0·7063	0·3337	0·3453	
H(341)	0·5483	0·2801	0·3997	
H(351)	0·4100	0·1681	0·3710	
H(361)	0·4261	0·1068	0·2824	
H(621)	0·9126	0·1062	0·2772	
H(631)	0·9390	0·0623	0·3660	
H(641)	0·8075	−0·0443	0·4057	
H(651)	0·6413	−0·1079	0·3575	
H(661)	0·6120	−0·0640	0·2675	
H(81)	0·4630	0·2436	0·0786	
H(101)	0·8479	0·1804	−0·0262	
H(102)	0·7226	0·2133	−0·0547	
H(103)	0·7374	0·1095	−0·0364	

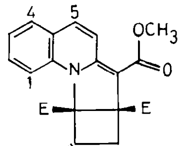
Discussion. Bond distances and angles are in Tables 2 and 3. The product from 1,2,3-triphenylguanidine and DMAD has in fact the five-membered ring structure (5) (Fig. 1). The plane of the ester group is at 45° to the plane of the exocyclic double bond and at 36° to the plane of the nearest phenyl group [attached to N(1)]. The ester methyl group deviates from the plane of the ester function [C(9), O(10) and O(11)] towards the adjacent phenyl group by 0·2 Å and lies 3·7 Å above the plane of that aromatic ring, accounting for the high-field ^{1}H NMR signal of the methyl protons (6·89τ). The three phenyl groups and the ester group are arranged like a propeller. The molecule is chiral in the crystal with enantiomeric pairs related by glide planes. The five-membered ring is slightly puckered. If a plane is defined by the ring C atoms, N(1) lies almost in the plane but N(3) deviates by 0·03 Å (Table 4).



(5)



(6)



(7)

Table 2. *Interatomic distances* (Å) *and e.s.d.'s*

O(7)–C(4)	1·235 (6)	C(12)–C(13)	1·422 (7)
O(10)–C(9)	1·332 (6)	C(13)–C(14)	1·388 (8)
O(10)–C(10)	1·447 (7)	C(14)–C(15)	1·414 (8)
O(11)–C(9)	1·189 (6)	C(15)–C(16)	1·407 (8)
N(1)–C(2)	1·426 (5)	C(31)–C(32)	1·404 (7)
N(1)–C(5)	1·404 (6)	C(31)–C(36)	1·379 (7)
N(1)–C(11)	1·460 (6)	C(32)–C(33)	1·387 (8)
N(3)–C(2)	1·429 (6)	C(33)–C(34)	1·346 (11)
N(3)–C(4)	1·394 (6)	C(34)–C(35)	1·401 (11)
N(3)–C(31)	1·448 (6)	C(35)–C(36)	1·417 (8)
N(6)–C(2)	1·269 (6)	C(61)–C(62)	1·415 (7)
N(6)–C(61)	1·413 (6)	C(61)–C(66)	1·398 (8)
C(4)–C(5)	1·493 (7)	C(62)–C(63)	1·385 (8)
C(5)–C(8)	1·357 (7)	C(63)–C(64)	1·393 (8)
C(8)–C(9)	1·470 (7)	C(64)–C(65)	1·383 (9)
C(11)–C(12)	1·404 (7)	C(65)–C(66)	1·409 (8)
C(11)–C(16)	1·390 (7)		

Table 3. *Interbond angles* (°), *e.s.d.'s* 0·2–0·7°

C(9)–O(10)–C(10)	116·6	C(12)–C(11)–C(16)	120·9
C(2)–N(1)–C(5)	110·8	C(11)–C(12)–C(13)	120·8
C(2)–N(1)–C(11)	120·3	C(12)–C(13)–C(14)	118·6
C(5)–N(1)–C(11)	128·1	C(13)–C(14)–C(15)	119·8
C(2)–N(3)–C(4)	113·0	C(14)–C(15)–C(16)	121·9
C(2)–N(3)–C(31)	126·0	C(11)–C(16)–C(15)	118·0
C(4)–N(3)–C(31)	119·7	N(3)–C(31)–C(32)	118·9
C(2)–N(6))–C(61)	124·4	N(3)–C(31)–C(36)	119·1
N(1)–C(2)–N(3)	104·5	C(32)–C(31)–C(36)	121·9
N(1)–C(2)–N(6)	121·5	C(31)–C(32)–C(33)	120·2
N(3)–C(2)–N(6)	134·1	C(32)–C(33)–C(34)	118·9
O(7)–C(4)–N(3)	127·1	C(33)–C(34)–C(35)	121·9
O(7)–C(4)–C(5)	128·4	C(34)–C(35)–C(36)	120·4
N(3)–C(4)–C(5)	104·5	C(31)–C(36)–C(35)	116·6
N(1)–C(5)–C(4)	107·2	N(6)–C(61)–C(62)	118·8
N(1)–C(5)–C(8)	133·2	N(6)–C(61)–C(66)	120·8
C(4)–C(5)–C(8)	119·5	C(62)–C(61)–C(66)	119·9
C(5)–C(8)–C(9)	127·8	C(61)–C(62)–C(63)	118·3
O(10)–C(9)–O(11)	122·6	C(62)–C(63)–C(64)	122·0
O(10)–C(9)–C(8)	113·6	C(63)–C(64)–C(65)	120·1
O(11)–C(9)–C(8)	123·8	C(64)–C(65)–C(66)	119·2
N(1)–C(11)–C(12)	121·3	C(61)–C(66)–C(65)	120·6
N(1)–C(11)–C(16)	117·8		

Table 4. *Deviations* (Å) *of atoms from plane defined by* C(2), C(3) *and* C(4)

Equation of the plane: $7·26x + 11·22y + 0·93z = 6·18$. The mean isotropic e.s.d. is 0·004 Å.

O(7)	0·04	C(8)	0·05
N(1)	0·00	C(11)	−0·21
N(3)	0·03	C(31)	0·35
N(6)	−0·06		

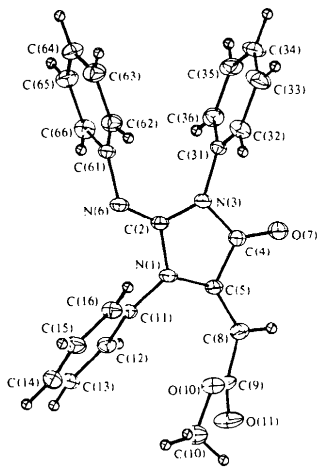


Fig. 1. Methyl (*Z*)-4-oxo-1,3-diphenyl-2-phenylimino-5-imidazolidinylideneacetate (5).

The phenyl groups attached to N(3) and N(6) are bent away from each other slightly such that N(6) deviates from the best plane through its phenyl group by 0·2 Å and N(3) deviates by 0·1 Å from its phenyl-ring plane (Fig. 2). The rather large bond angles N(6)–C(2)–N(3) and C(2)–N(3)–C(31) [134·1 (4) and 126·0 (4)° respectively] reflect this repulsion between the phenyl groups. The angle between the best planes of these two rings is 38° and C(31) and C(61) are 3·2 Å apart.

Since the ester group is 45° out of the plane of the exocyclic double bond, it cannot fully conjugate with the enaminic system. The C(8)–CO_2R bond length for

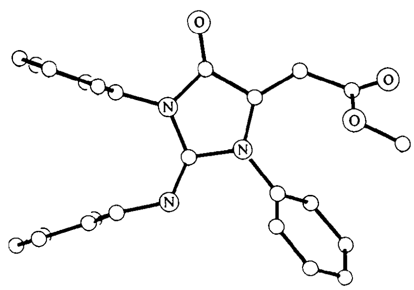


Fig. 2. View of (5) showing apparant repulsion between the phenyl groups attached to N(3) and N(6).

Table 5. $C—CO_2R$ *bond length for* (5), (6) *and* (7)

Ester group	Angle between plane of ester group and the plane of the enaminic system (°)	$C—CO_2R$ bond length (Å)
(5) 8-ester	45	1·470 (7)
(6) 6-ester	10	1·439 (7)
(6) 7-ester	70	1·504 (6)
(6) 8-ester	43	1·464 (7)
(7) 7-ester	0	1·443 (8)

(5) is compared in Table 5 with the equivalent parameters (from crystal structure data) for the 6- and 7-esters of (6) (Abbott, Acheson, Eisner, Watkin & Carruthers, 1976) and the 7-ester of (7) (Abbott, Acheson, Forder, Watkin & Carruthers, 1977). The 6-ester of (6) and the 7-ester of (7) are almost coplanar with an enaminic system and have shorter $C—CO_2R$ bonds while the 7-ester of (6), which is at 70° to the conjugated system, has a longer bond length (1·504 Å). There is a close similarity between the $C—CO_2R$ bond length for the 8-ester of (6) and the 8-ester of (5) which both make similar angles (43 and 45°) with their respective enaminic systems.

It may be concluded that mass spectrometry is an unreliable method for differentiating between the structural possibilities (1) and (2), and an alternative method using ^1H and ^{13}C NMR spectra which is based on the established structure (5) will be published elsewhere (Acheson & Wallis, 1980).

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N,N'-Tetramethylenedibenzamide (TMDB)*

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Abstract. $C_{18}H_{20}N_2O_2$, monoclinic, $P2_1/c$, $a = 5·118\,(3)$, $b = 5·324\,(3)$, $c = 28·410\,(5)$ Å, $\beta = 97·05\,(5)°$, $Z = 2$. The crystal structure was solved by direct methods. All H atoms have been located. $R_w = 4·7\%$. The planes of the phenyl ring and the amide group are rotated with respect to each other due to steric hindrance. Hydrogen bonds connect molecules related by translation in the *a* direction.

Introduction. The present investigation reports the crystal structure of TMDB, a model compound of an aromatic–aliphatic polyamide. TMDB was prepared as described by Gaymans & Harkema (1977). Intensities were measured on a Philips PW 1100 diffractometer (Mo $K\alpha$ radiation, $\lambda = 0·71069$ Å, graphite monochromator). Reflections up to $\theta = 30°$ were measured with the $\omega/2\theta$ scan mode.

The number of reflections measured was 1612. All reflections were used in the refinement. No absorption correction was applied. Details of the solution of the structure, the weighting scheme, the scattering factors

* The Structure of Model Compounds of Aromatic and Aromatic–Aliphatic Polyamides. III. Part II: Harkema, Gaymans, van Hummel & Zylberlicht (1979).

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