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CONFORMATIONAL STUDIES ABOUT THE N–N' BOND BY NMR SPECTROSCOPY AND THEIR APPLICATION IN THE CONFIGURATIONAL ASSIGNMENT TO THE DIELS–ALDER ADDUCTS OF MALEIC ANNHYDRIDE[a]

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Abstract—Non-planar stable conformations about the N–N' bond in N-(diacylamino)imide derivatives of naphthalene-maleic anhydride adducts **2** and **3** and their reduced products **4** and **5** have been studied by NMR spectroscopy and have been demonstrated as a means of assigning *exo/endo* configurations to the Diels–Alder adducts. While N-(diacetylamino)imide system has proven to be a suitable probe for configurational assignment to Diels–Alder adducts, which show clear cut magnetic effects on the N'-substituents, the N'-acetyl-N'-aroyl-N-aminoimide system has been shown to be a versatile probe, applicable even to simple adducts. The N-(diaroylamino) imide system has shown little effect on the cage-protons, whereby it has been presumed that the aryl-rings of the N'-aroyl groups preferably reside away from the cage-moiety.

Restricted rotation about N–N bond in acyclic systems has been extensively studied by NMR spectroscopy.[1] The existence of non-planar ground states about the N–N' bond in N-(diacyl amino)imide systems **1** has been demonstrated with the help of a cage-moiety, having no symmetry about the plane of the imidyl-ring.[2-4] A preliminary communication explored the possibility of assigning *exo-/endo*-configurations to the Diels–Alder adducts of maleic anhydride with the help of the conformational studies of the N-(diacetyl amino)imide derivatives of those adducts.[5] Since the N-(diacetyl amino) imide system assumes a non-planar conformation about the N–N bond at room temperature, one of the N'-acetyl groups lying nearer to the cage-moiety, probes into the magnetic effect of the dienyl part of the adduct, whereby the configuration of the adduct could be determined. The utility of this technique for the determination of configurations of different types of Diels–Alder adducts of maleic anhydride is discussed in this article. The conformational studies of a series of N-(diacylamino)imide derivatives **2** and **3** of the Diels–Alder *exo* and *endo* adducts of naphthalene and maleic anhydride and their reduced products **4** and **5** have shown that the N'-acetyl-N'-aroyl system is another versatile probe for the direct determination of configuration of the adducts.

N',N'-Diacetyl Derivatives (**2b**, **3b**, **4b** *and* **5b**)

The NMR spectrum of the compound **2b** shows two singlets (each of 3H intensity, $\Delta\nu = 20$ Hz) for the two acetyl groups and normal resonances for the other protons (Table 1). The spectrum of the reduced product **4b** is very much similar to that of **2b** (Table 1). The spectra of the compounds **3b** and **5b** also show similar pattern to those of **2b** and **4b** respectively, but the signals of the N'-acetyl groups are largely separated ($\Delta\nu \approx 100$ Hz). In all these compounds one of the N'-acetyls resonates at the normal position, while the other is shielded by the cage-moiety.

In the spectra of the *exo*-compounds it is the CH=CH or $-CH_2-CH_2-$ system, while in the spectra of the *endo*-compounds it is the benzo group, that affects one of the N'-acetyl groups. Since it is well known that a benzo group has a greater influence than a –CH=CH– or $-CH_2-CH_2-$ system, the compounds **3b** and **5b**, which show a large shielding effect on one of the N'-acetyl groups, could be easily given *endo*-configuration while the compounds **2b** and **4b**, which show comparatively very small shielding effect, could be given *exo*-configuration.

The similarity between the $\Delta\nu$ values of N'-acetyl groups in **2b** and **4b** suggests that a compound of the type **6** would show a similar $\Delta\nu$ value for the *endo*- and *exo*-products, whereby it would not be possible to assign the configuration. In such simple systems where the cage-moiety has no clear-cut effects on the N'-substituents, introduc-

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[a] Part of the work was presented at the Fifth International Symposium on Magnetic Resonance held at Bombay (14–18 Jan 1974).

Table 1: NMR spectral data

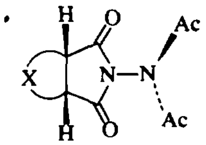
Comp. No.	δA	δB	δ(1 + 4)	δ(2 + 3)	δ(9 + 10)	δ(5 + 6 + 7 + 8)
2a	(m-1H) 4·41	(m-1H) 4·41	(m-2H) 4·60	(t-2H) 3·03	(m-2H) 6·67	(m-4H) 7·44
2b	(ds-3H) 2·36; 1:1; 20 Hz	(ds-3H) 2·36; 1:1; 20 Hz	(m-2H) 4·68	(t-2H) 3·23	(m-2H) 6·78	(m-4H) 7·42
2c	(ds-3H) 2·61; 2:1; 6 Hz	(m-5H) 7·58	(m-2H) 4·53	(dt-2H) 3·01; 2:1; 13·5 Hz	(dm-2H) 6·33; 1:2; 57 Hz	(m-4H) 7·58
2d	(ds-3H) 2·67; 2:1; 5 Hz	(ds-3H) 2·46; 2:1 3 Hz (m-4H) 7·37	(m-2H) 4·44	(dt-2H) 2·92; 2:1; 15·5 Hz	(dm-2H) 6·22; 1:2; 61 Hz	(m-4H) 7·37
2e	(m-5H) 7·63	(m-5H) 7·63	(m-2H) 4·56	(t-2H) 3·12	(m-2H) 6·30	(m-4H) 7·63
2f	(s-3H) 2·48; (m-4H) 7·59	(s-3H) 2·48; (m-4H) 7·59	(m-2H) 4·58	(t-2H) 3·17	(m-2H) 6·36	(m-4H) 7·59
3a	(m-1H) 3·81	(m-1H) 3·81	(m-2H) 4·53	(t-2H) 3·15	(m-2H) 6·86	(nm-4H)7·31
3b	(ds-3H) 1·71; 1:1; 99 Hz	(ds-3H) 1·71; 1:1; 99 Hz	(m-2H) 4·65	(t-2H) 3·39	(m-2H) 6·93	(m-4H) 7·43
3c	(ds-3H) 1·75; 1:7; 61 Hz	(m-5H) 7·58	(m-2H) 4·65	(t-2H) 3·31	(m-2H) 6·91	(m-4H) 7·58
3d	(m-5H) 7·55	(m-5H) 7·55	(m-2H) 4·60	(t-2H) 3·31	(m-2H) 6·90	(m-4H) 7·55
4a	(nm-1H) 4·70	(nm-1H) 4·70	(m-2H) 3·70	(m-2H) 2·96	(q-2H) 1·63	(nm-4H) 7·46
4b	(ds-3H) 2·43; 1 : 1; 20 Hz	(ds-3H) 2·43; 1:1; 20 Hz	(m-2H) 3·68	(m-2H) 3·05	(bm-4H) 1·21–2·30	(nm-4H) 7·41
4c	(ds-3H) 2·69; 1:1; 9 Hz	(m-5H) 7·63	(m-2H) 3·60	(dm-2H) 2·84; 1:1; 23 Hz	(bm-4H) 0·95–2·35	(m-4H) 7·63
4d	(ds-3H) 2·71; 1:1; 9·5 Hz	(ds-3H) 2·51; 1:1; 5 Hz (m-4H) 7·42	(m-2H) 3·54	(dm-2H) 2·76; 1:1; 26 Hz	(bm-4H) 0·80–2·32	(m-4H)7·42
4e	(ds-3H) 2·64; 2:1; 9·5 Hz	(ds-3H) 2·41; 1:2; 3 Hz (m-4H) 7·58	(m-2H) 3·58	(dm-2H) 2·85; 1:2; 21 Hz	(bm-4H) 1·04–2·35	(m-4H) 7·58
4f	(m-5H) 7·71	(m-5H) 7·71	(m-2H) 3·66	(m-2H) 2·97	(bm-4H) 1·11–2·11	(m-4H) 7·71
4g	(ds-3H) 2·47; 1:1; 3 Hz (m-4H) 7·63	(ds-3H) 2·47; 1 : 1; 3 Hz (m-4H) 7·63	(m-2H) 3·68	(m-2H) 3·00	(bm-4H) 1·11–2·20	(m-4H) 7·63
4h	(s-3H) 2·38; (m-4H) 7·58	(s-3H) 2·38; (m-4H) 7·58	(m-2H) 3·65	(m-2H) 2·95	(bm-4H) 1·10–2·16	(m-4H) 7·58
5a	(m-1H) 3·90	(m-1H) 3·90	(m-2H) 3·66	(m-2H) 3·16	(q-4H) 1·79	(nm-4H) 7·33
5b	(ds-3H) 1·70; 1:1; 104 Hz	(ds-3H) 1·70; 1:1; 104 Hz	(m-2H) 3·76	(m-2H) 3·40	(q-4H) 1·81	(nm-4H) 7·37
5c	(ds-3H) 1·77; 1:7; 72 Hz	(m-5H) 7·41	(m-2H) 3·74	(m-2H) 3·28	(q-4H) 1·78	(m-4H) 7·41
5d	(ds-3H) 1·77; 1:7; 72 Hz	(ds-3H) 2·32; 7:1; 6 Hz (m-4H) 7·33	(m-2H) 3·71	(m-2H) 3·26	(q-4H) 1·76	(m-4H) 7·33
5e	(ds-3H) 1·78; 1:10; 73 Hz	(ds-3H) 2·35; 10:1; 3·5 Hz (m-4H) 7·33	(m-2H) 3·73	(m-2H) 3·30	(q-4H) 1·75	(m-4H) 7·33
5f	(ds-3H) 1·78; 1:4·5; 73 Hz	(s-3H) 2·42; (m-4H) 7·43	(m-2H) 3·73	(m-2H) 3·30	(q-4H) 1·80	(m-4H) 7·43
5g	(m-5H) 7·54	(m-5H) 7·54	(m-2H) 3·71	(m-2H) 3·28	(q-4H) 1·76	(m-4H) 7·54
5h	(ds-3H) 2·23; 1:1; 6·5 Hz (m-4H) 7·35	(ds-3H) 2·23; 1:1; 6·5 Hz (m-4H) 7·35	(m-2H) 3·70	(m-2H) 3·25	(q-4H) 1·76	(m-4H) 7·35
5i	(s-3H) 2·33; (m-4H) 7·37	(s-3H) 2·33; (m-4H) 7·37	(m-2H) 3·67	(m-2H) 3·25	(q-4H) 1·70	(m-4H) 7·37

NMR spectra were recorded in $CDCl_3$ at 44°C. Total number of protons and the multiplicity of the bands are indicated in brackets.

In case of multiplicity due to slow rotation, the ratio of the intensity of the downfielded signal to the upfielded and the separation (in Hz) are indicated.

s = singlet, t = triplet, q = quartet, m = multiplet, ds = double singlet, dt = double triplet, dm = double multiplet, nm = narrow multiplet, bm = broad multiplet

TMS as internal reference.



X = Chain of four Carbons

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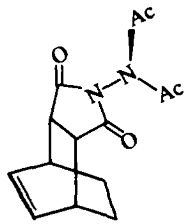

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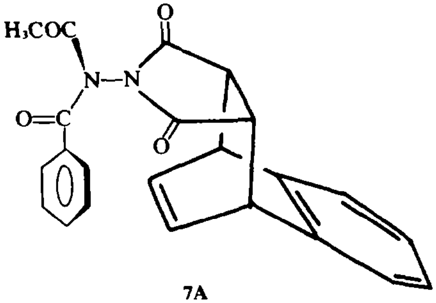
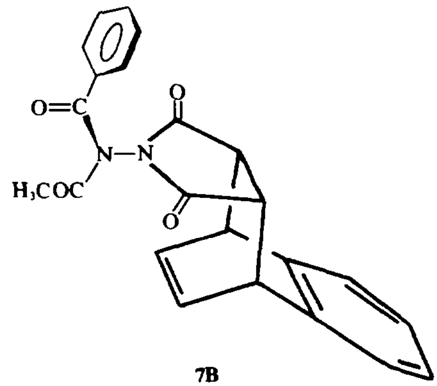


7A



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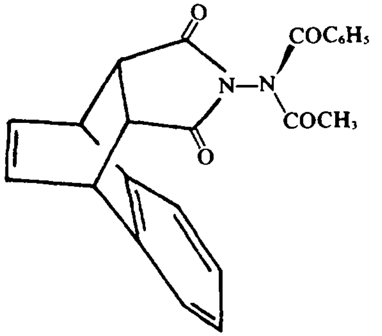

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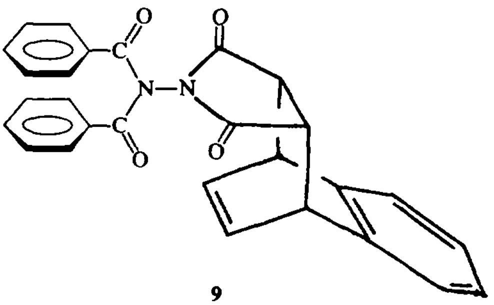

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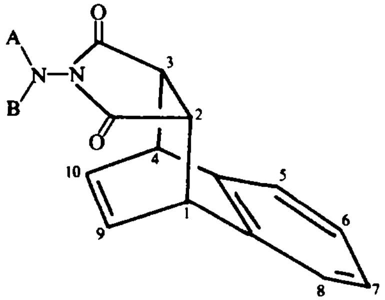
tion of a group, which can strongly influence the resonances of the cage-protons, at the *exo*-cyclic nitrogen would give fruitful results. N'-acetyl-N'-aroyl system described below is found to be satisfactory for this purpose.

N'-Acetyl-N'-Aroyl Derivatives (**2c**, **2d**, **3c**, **4c–4e** *and* **5c–5f**)

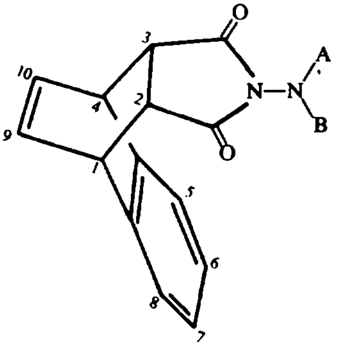
The NMR spectrum of **2c** (Fig 1) shows two multiplets ($\Delta\nu = 57$ Hz) for the C_9 and C_{10} olefinic protons, two singlets ($\Delta\nu = 6$ Hz) for the acetyl protons, two triplets for the C_2 and C_3 protons and normal resonances for the other protons (Table 1). The spectral multiplicity evidences for the restricted rotation, non-planar ground states and the presence of two stable conformers **7A** and **7B** about the N–N' bond. The spectrum of the other isomeric compound **3c** (Fig 2) shows two singlets (intensity ratio 1:7; $\Delta\nu = 61$ Hz) for the acetyl protons and normal resonances for the other protons (Table 1). The multiplicity of the acetyl signals evidences for the presence of two different conformers about the N–N' bond. The C_2 and C_3 protons would be expected to exhibit two signals due to the two conformers, but they show only one signal presumably because the other signal, due to its low intensity and multiplet nature, might have been merged in the noise.

The N'-benzoyl group in N'-acetyl-N'-benzoyl-N-amino[2.2.1] bicyclo-5-heptene-2,3-*endo*-dicarboximide has shown a large shielding effect on the cage-olefinic protons in the conformation in which the benzoyl group lies *syn* to the cage-moiety.[3] Similar shielding effect of the N'-aroyl group on the cage-protons has been observed in the N'-acetyl-N'-aroyl-N-amino camphorimide.[6] Therefore, the strong shielding effect of the N'-aroyl



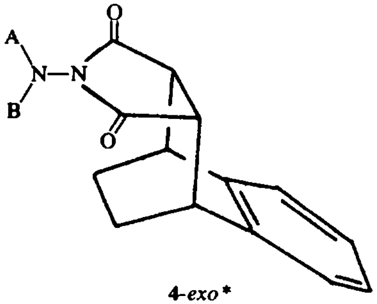
2-*exo**

No	Compound	
	A	B
2a	H	H
2b	$COCH_3$	$COCH_3$
2c	$COCH_3$	COC_6H_5
2d	$COCH_3$	$COC_6H_4 \cdot CH_3$-o
2e	COC_6H_5	COC_6H_5
2f	$COC_6H_4 \cdot CH_3$-o	$COC_6H_4 \cdot CH_3$-o



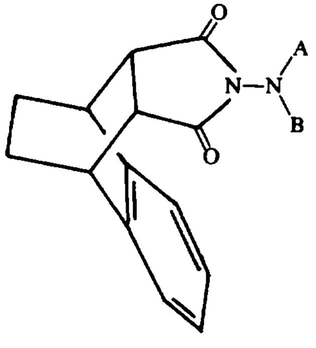
3-*endo**

No	Compound	
	A	B
3a	H	H
3b	$COCH_3$	$COCH_3$
3c	$COCH_3$	COC_6H_5
3d	COC_6H_5	COC_6H_5



4-*exo**

No	Compound	
	A	B
4a	H	H
4b	$COCH_3$	$COCH_3$
4c	$COCH_3$	COC_6H_5
4d	$COCH_3$	$COC_6H_4 \cdot CH_3$-o
4e	$COCH_3$	$COC_6H_4 \cdot CH_3$-p
4f	COC_6H_5	COC_6H_5
4g	$COC_6H_4 \cdot CH_3$-o	$COC_6H_4 \cdot CH_3$-o
4h	$COC_6H_4 \cdot CH_3$-p	$COC_6H_4 \cdot CH_3$-p



5-*endo**

No	Compound	
	A	B
5a	H	H
5b	$COCH_3$	$COCH_3$
5c	$COCH_3$	COC_6H_5
5d	$COCH_3$	$COC_6H_4 \cdot CH_3$-o
5e	$COCH_3$	$COC_6H_4 \cdot CH_3$-m
5f	$COCH_3$	$COC_6H_4 \cdot CH_3$-p
5g	COC_6H_5	COC_6H_5
5h	$COC_6H_4 \cdot CH_3$-o	$COC_6H_4 \cdot CH_3$-o
5i	$COC_6H_4 \cdot CH_3$-p	$COC_6H_4 \cdot CH_3$-p

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*The prefixes *endo* and *exo* are used in the sense that substituents on the same side of the bicyclo[2.2.2] octene ring as the benzene ring are *endo*, those on the other side are *exo*.

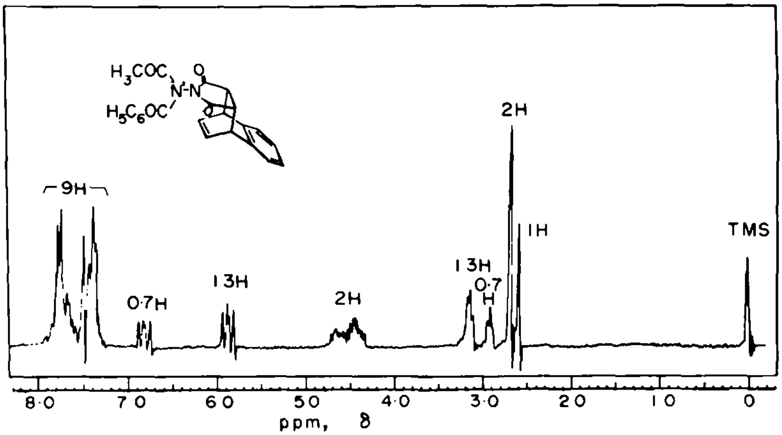

Fig 1. 60 MHz NMR spectrum of N'-acetyl-N'-benzoyl-N-amino-1,2,3,4-tetrahydro-1,4-ethylenonaphthalene-2,3-*exo*-dicarboximide (**2c**) in $CDCl_3$ at 44°C.

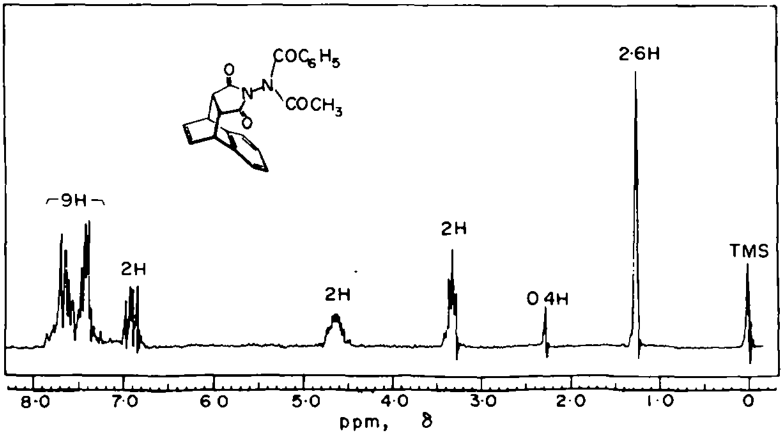

Fig 2. 60 MHz NMR spectrum of N'-acetyl-N'-benzoyl-N-amino-1,2,3,4-tetrahydro-1,4-ethylenonaphthalene-2,3-*endo*-dicarboximide (**3c**) in $CDCl_3$ at 44°C.

group on the C_9 and C_{10} olefinic protons in one of the conformers of **2c** strongly suggests an *exo*-configuration to this compound, while the absence of such a shielding effect in **3c** suggests an *endo*-configuration.

The reduced products **4c** and **5c** also exhibit similar behavior. The C_9 and C_{10} protons in **5c** are least affected by the N'-substituents and show a well resolved AA' BB' type signal, whereas the C_9 and C_{10} protons in **4c** exhibit a very broad multiplet (δ 0·95–2·35) due to the superimposition of two AA' BB' type signals caused by the restricted rotation about the N–N' bond. Hence **4c** could be assigned *exo*-configuration while **5c** could be assigned *endo*-configuration. All the other N'-acetyl-N'-aroyl compounds also show similar NMR spectra and the configurations could be assigned.

The *endo*-isomers show high preference for the conformation **8** in which the N'-aroyl group lies *anti* to the cage-moiety (about 7 times the other), probably due to the large steric hindrance in the other conformation. The *exo*-isomers **2c** and **2d** show some preference for more crowded conformation **7A**, in which the N'-aroyl group lies *syn* to the cage-moiety (about twice the other), as it could be observed from the intensities of the shielded and deshielded signals of the C_9 and C_{10} protons. This type of conformational preference has been observed in a series of N'-acetyl-N'-aroyl-N-amino [2.2.1] bicyclo-5-heptene-2,3-*endo*- dicarboximide derivatives. Surprisingly, the compounds **4c**–**4e**, in which the C_9–C_{10} double bond is saturated, have shown both the conformers about the N–N' bond in almost the same populations (Table 1). Therefore the unusual preference for the crowded conformation in the compounds **2c**, **2d** may be due to some intra-molecular interaction (attraction) between the cage-olefinic bond and the N'-aroyl ring.

Table 2. M.ps., Elemental Analyses and Characteristic IR peaks[a]

Compd. No.	A	B	M. °C	Found		Calculated		IR
				C%	H%	C%	H%	ν_{max} cm^{-1}
2a	H	H	185–187	69·60	5·33	70·00	5·00	3325 m, 3260 m, 3200 m, 1778 m, 1695 s, 1620 m, 760 m,
2b	$COCH_3$	$COCH_3$	177–179	66·71	4·47	66·66	4·93	1790 w, 1750 s, 1732 s, 754 m.
2c	$COCH_3$	COC_6H_5	170–171	70·68	4·87	71·50	4·66	1790 w, 1740 s, 1730 s, 1603 w, 755 m.
2d	$COCH_3$	$COC_6H_4{\cdot}CH_3$-o	170–172	71·78	5·10	72·00	5·00	1790 w, 1744 s, 1730 s, 1605 w, 760 w.
2e	COC_6H_5	COC_6H_5	195–197	75·34	4·33	75·00	4·46	1793 w, 1743 s, 1720 s, 1605 w, 760 m.
2f	$COC_6H_4{\cdot}CH_3$-o	$COC_6H_4{\cdot}CH_3$-o	185–187	75·44	4·98	75·63	5·04	1790 w, 1739 s, 1725 s, 1700 m, 760 m.
3a	H	H	190–191	69·60	4·92	70·00	5·00	3340 m, 3260 w, 1780 w, 1690 s, 1580 w, 760 w.
3b	$COCH_3$	$COCH_3$	198–201	66·30	4·88	66·66	4·92	1800 w, 1749 s, 1733 s, 750 w.
3c	$COCH_3$	COC_6H_5	202–204	71·88	4·33	71·50	4·66	1790 w, 1740 s, 1710 s, 1608 w, 750 w.
3d	COC_6H_5	COC_6H_5	193–195	74·87	4·52	75·00	4·46	1798 w, 1747 s, 1730 s, 1607 w.
4a	H	H	192–194	68·80	5·66	69·42	5·78	3320 m, 3275 m, 1785 w, 1705 s, 1600 m, 750 m.
4b	$COCH_3$	$COCH_3$	215–217	66·00	5·60	66·25	5·52	1793 w, 1736 s, 755 w.
4c	$COCH_3$	COC_6H_5	192–195	70·52	5·02	71·13	5·15	1790 w, 1743 s, 1733 s, 1607 w, 760 m.
4d	$COCH_3$	$COC_6H_4{\cdot}CH_3$-o	183–186	71·33	5·32	71·64	5·47	1790 w, 1748 s, 1733 s, 1608 w, 760 m, 750 m.
4e	$COCH_3$	$COC_6H_4{\cdot}CH_3$-p	190–192	72·00	5·32	71·64	5·47	1800 m, 1740 s, 1720 s, 1615 m, 760 m.
4f	COC_6H_5	COC_6H_5	218–221	74·20	4·67	74·66	4·88	1805 w, 1734 s, 1712 s, 1604 w, 755 w.
4g	$COC_6H_4{\cdot}CH_3$-o	$COC_6H_4{\cdot}CH_3$-o	209–212	74·77	5·33	75·31	5·43	1740 s, 1725 s, 1698 s, 1605 w, 760 w, 738 w.
4h	$COC_6H_4{\cdot}CH_3$-p	$COC_6H_4{\cdot}CH_3$-p	230–234	75·50	5·46	75·31	5·43	1793 w, 1739 s, 1715 s, 1690 s, 1615 m, 758 w.
5a	H	H	227–230	69·02	5·66	69·42	5·78	3315 m, 3260 w, 1690 s, 1610 w.
5b	$COCH_3$	$COCH_3$	193–196	66·30	5·32	66·25	5·52	1807 w, 1745 s, 1725 s.
5c	$COCH_3$	COC_6H_5	219–222	70·44	4·98	71·13	5·15	1798 w, 1740 s, 1708 s, 1602 w.
5d	$COCH_3$	$COC_6H_4{\cdot}CH_3$-o	184–186	70·86	5·22	71·64	5·47	1805 w, 1770 s, 1744 s, 1705 s, 1605 w, 763 m.
5e	$COCH_3$	$COC_6H_4{\cdot}CH_3$-m	179–182	71·24	5·50	71·64	5·47	1800 w, 1744 s, 1710 s, 1615 w, 753 m.
5f	$COCH_3$	$COC_6H_4{\cdot}CH_3$-p	205–207	71·33	5·51	71·64	5·47	1800 w, 1743 s, 1714 s, 1614 m, 756 m.
5g	COC_6H_5	COC_6H_5	210–213	75·02	5·00	74·66	4·88	1744 s, 1706 s, 1607 w, 757 w.
{**5h**	$COC_6H_4{\cdot}CH_3$-o	$COC_6H_4{\cdot}CH_3$-o	194–196	74·80	5·23	75·31	5·43	1740 s, 1700 s, 1603 w, 750 w, 740 m.
{**5i**	$COC_6H_4CH_3$-p	$COC_6H_4{\cdot}CH_3$-p	224–227	75·34	5·08	75·31	5·43	1734 s, 1690 s, 1616 m, 752 w, 740 w.

[a] IR spectra were recorded in Nujol. m = medium, s = strong and w = weak.

N′,N′-Diaroyl Derivatives (**2e**, **2d**, **3d**, **4f**–**4g** *and* **5g**–**5i**)

An N′,N′-diaroyl system, with two effective groups, would show a promising effect on the cage-protons, so as to make the configurational assignment more easier. But, surprisingly, the N′,N′-diaroyl system has shown little effect on the cage protons. The spectrum of **2e** shows a normal signal at δ 6·3 for the C_9 and C_{10} protons, a triplet at δ 3·2 for the C_2 and C_3 protons and normal signals for the other protons also (Table 1). The N′-benzoyl group of the compound **2c** shielded the C_9 and C_{10} protons to a very high field δ 5·86 and still more shielding would be expected in the compound **2e**, but the observation is exactly the reverse. All the other N′,N′-diaroyl compounds also exhibit similar behavior (Table 1). Non-planar ground states about the tetraacyl N–N′ system is presumed beyond doubt. Therefore, in order to explain the observed spectral data, a folded type conformation **9** was suggested for the N′,N′-diaroyl system. Thus the N-(diaroylamino)imide system is least informative about the configuration of the Diels–Alder adducts.

EXPERIMENTAL

The NMR spectra were recorded on a Varian A-60D spectrometer and the spectral data of the compounds are recorded in Table 1. Infrared spectra were recorded for Nujol mulls on a Perkin-Elmer-257 spectrophotometer. Melting points, chemical analyses and IR bands of the compounds are recorded in Table 2.

The *endo* and *exo* adducts with benzo bicyclo[2.2.2] octenes were obtained according to the method of Takeda *et al.*[7] Both the isomeric adducts were transformed into N-amino imides **3a** and **2a** with equimolar amounts of hydrazine hydrate at room temperature in ethanol medium. The N-amino imides **2a** and **3a** on refluxing with hydrazine hydrate[8] (slightly excess than two moles) in ethanol in presence of animal charcoal gave the reduced products **4a** and **5a**. These reduced products were identical with those obtained by catalytic reduction of **2a** and **3a**. All the N-amino imides were recrystallised from ethanol and gave benzal derivatives with benzaldehyde which showed a characteristic one proton singlet near δ.

The N′,N′-diacetyl derivatives **2b**, **3b**, **4b** *and* **5b** were prepared by heating the corresponding N-amino imides with excess of acetic anhydride the product was washed with water, dried and recrystallised from ethanol.

N′-Acetyl-N′-benzoyl derivatives **2c**, **2d**, **3c**, **4c**–**4e** *and* **5c**–**5f** were prepared in two stages. N′-Monobenzoyl derivatives were obtained by refluxing equimolar amounts of N-amino imide, benzoyl chloride and pyridine in benzene for about three hrs. After removing the solvent, the product was washed with water, dried and recrystallised from ethanol. The mono-benzoyl derivatives thus obtained were acetylated by heating on water bath with excess of acetic anhydride and few drops of pyridine.

N′,N′-Diaroyl derivatives **2e**, **2f**, **3d**, **4f**–**4h** and **5g**–**5i** were obtained by heating the N-amino imides with slightly more than two moles of the respective aroyl chlorides and few drops of pyridine at 120–125°C for 2–3 h. The products were washed with water and were recrystallised either from ethanol or ethyl acetate.

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