

Base Catalysed Rearrangements involving Ylide Intermediates. Part 3.¹ A Novel Thermal [1,3] Sigmatropic Rearrangement

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The products (2) of a [3,2] sigmatropic rearrangement of ammonium ylides (1) undergo a thermal transformation by a [1,3] sigmatropic rearrangement giving amines (3).

In Part 2,¹ the competition between the [3,2] sigmatropic rearrangement and the Stevens [1,2] rearrangement of ammonium ylides (1) was examined. During this study it was found that the [3,2] sigmatropic rearrangement products (2) were thermally transformed into the Stevens rearrangement products (3).

(53%) was observed and some starting material (2h) was recovered (47%).

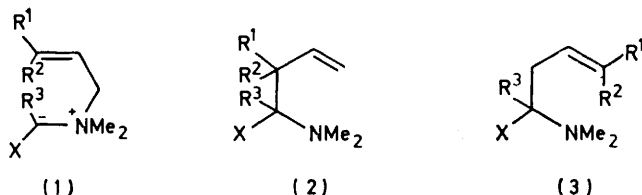
The structural requirements for the thermal rearrangement (2) \rightarrow (3) are an allyl residue in association with a group X where X is benzoyl, *p*-nitrophenyl, acetyl, and methoxycarbonyl. An obvious substituent effect is

Examples of the rearrangement reaction (2) \rightarrow (3)

Compound (2) †	R ¹	R ²	R ³	X	Temperature (°C)	Time	Yield (2) \rightarrow (3) (%)
(a)	Me	Me	H	PhCO	180	1 h	92
(c) (A + B) †	Ph	H	H	PhCO	120	15 h	100
(d) (A + B) †	<i>o</i> -MeC ₆ H ₄	H	H	PhCO	115	15 h	100
(e)	Me	Me	H	<i>p</i> -NO ₂ C ₆ H ₄	180	30 min	76
(f) (A) *	Ph	H	H	<i>p</i> -NO ₂ C ₆ H ₄	145	1 h	100
(g)	Me	Me	H	CH ₃ CO	160	2 h	87
(h)	Me	Me	H	MeO ₂ C	205	15 h	53
(j)	Me	Me	Me	MeO ₂ C	120	1 h	90
(k)	Me	Me	Me	PhCO	100	10 min	94

* The single diastereoisomer A was used for the rearrangement of compound (2f). † The letters used to refer to particular compounds match those used in the Table of Part 2.¹ ‡ The letters A and B refer to the use of mixtures of diastereoisomers for compounds (2c and d).

Examples of this thermal rearrangement (2) \rightarrow (3) are listed in the Table together with the experimental conditions used.²

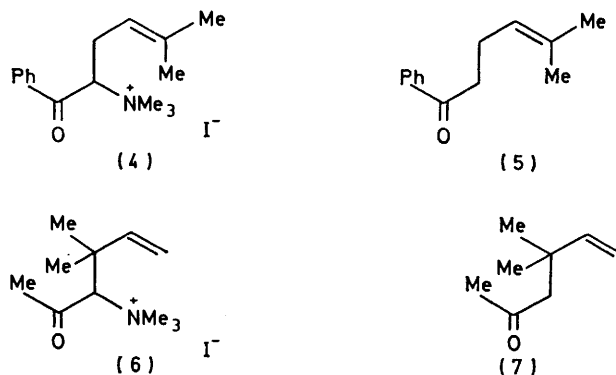


The constitutions of the products (3) were established mainly by their n.m.r. spectra and their relationship to the precursors (2). Definitive evidence was provided by the reduction by zinc and sulphuric acid of the methiodide (4) derived from the rearrangement product (3a). The reduction product (5) showed n.m.r. signals characteristic of the grouping $>C=CH_X-C(H_A)_2-C(H_B)_2-CO$ with the coupling constants J_{AX} 7 and J_{AB} 7 Hz and two vinylic methyl groups, τ 8.30 and 8.37.

The rearrangement (2) \rightarrow (3) can formally be regarded as a [1,3] sigmatropic shift.³⁻²² The experimental conditions required to effect most of the rearrangements (2) \rightarrow (3) are very mild compared with the conditions normally required (250–300°) to effect other thermal [1,3] alkyl migrations in unstrained systems.^{4,7,11,16,19} In only one case (2h) was a temperature $>200^\circ$ required and even under these conditions only partial conversion into the product (3h)

observed: the rate of rearrangement (2a) \rightarrow (3a) is much less than the corresponding reaction (2k) \rightarrow (3k). The role of the dimethylamino group in (2) was explored in the following way. The compound (2g) yielded a methiodide (6) which gave the unsaturated ketone (7) by reduction with zinc and sulphuric acid. This ketone (7) was quite stable under the thermal conditions (210°; 12 h) and showed no evidence that an isomerisation of the type (2) \rightarrow (3) was possible.

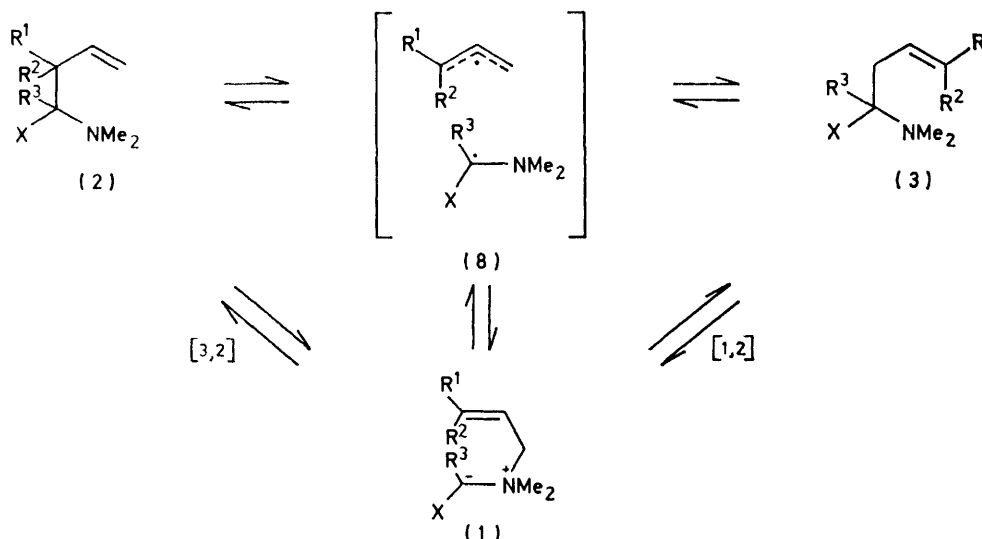
The stereoselectivity of the rearrangement was hopefully examined, but no conclusive result was obtained.



Optically active amine (2g) was prepared from the racemate by fractional crystallisation of the diastereoisomeric adducts with (+)-camphorsulphonic acid, fol-

lowed by treatment with sodium hydrogen carbonate. The amine (2g), $[\alpha]_D^{23} +47^\circ$, on heating at 160° for 2 h yielded racemic product (3g). This result unfortunately is not directly useful because the racemisation could be associated either with the rearrangement (2g) \rightarrow (3g) or with base catalysed racemisation of the starting material (2g) or the product (3g).

The thermal rearrangement (2) \rightarrow (3) is consistent with a mechanism involving cleavage of the amine (2) into the radical pair (8) which is stabilised by the substituents R^3 , X, and NMe_2 (Scheme). The remarkable



SCHEME Additional evidence concerning the mechanism of analogous [1,3] rearrangements in related systems is given in Part 4²²

substituent effects caused by the groups R^3 and NMe_2 are thus adequately explained. It should also be emphasised that the radical pair (8) is the same as that which has been suggested¹ as a possible intermediate in the Stevens rearrangement of ylide (1). Alternatively, a radical pair intermediate need not necessarily be involved and the reaction (2) \rightarrow (3) could be another example of a concerted-forbidden process.²³ A third possibility, indicated in the Scheme, involves a reverse [3,2] sigmatropic rearrangement²⁴ giving the ylide (1) which then yields the thermodynamically preferred product (3g) by a Stevens [1,2] rearrangement.

EXPERIMENTAL

The general directions are the same as in Part 1.²⁵

Thermal Rearrangement of 3,3-Dimethyl-2-dimethylamino-1-phenylpent-4-en-1-one (2a). Formation of 2-Dimethylamino-5-methyl-1-phenylhex-4-en-1-one (3a).—3,3-Dimethyl-2-dimethylamino-1-phenylpent-4-en-1-one¹ (6.0 g) was kept in a sealed evacuated tube at 180° for 1 h, then distilled giving 2-dimethylamino-5-methyl-1-phenylhex-4-en-1-one (5.5 g, 92%) as a yellow oil, b.p. $85\text{--}87^\circ$ at 0.02 mmHg (Found: C, 77.7; H, 9.1; N, 6.3. $C_{15}H_{21}NO$ requires C, 77.9; H, 9.1; N, 6.1%); τ 1.9—2.8 (m, 5 aromatic H), AMNX system, τ_A 4.94, τ_M, τ_N 7.1—7.7, τ_X 6.00 ($J_{AM} = J_{AN} = ca. 7$, $J_{MX} 6$, $J_{NX} 8$ Hz, $=CH_A-CH_MCH_N-CH_X-CO$), τ 7.67 (s, NMe_2) and 8.40br (s, $=CMe_2$).

2-Dimethylamino-5-methyl-1-phenylhex-4-en-1-one Methiodide (4).—2-Dimethylamino-5-methyl-1-phenylhex-4-en-

1-one¹ (5.0 g) and methyl iodide (50 ml) were heated under reflux for 24 h, cooled, and diluted with ether (250 ml). The precipitate was collected and crystallisation from ether-ethyl acetate-methanol yielded 2-dimethylamino-5-methyl-1-phenylhex-4-en-1-one methiodide (4) (6.0 g, 80%), m.p. 139° (Found: C, 51.1; H, 6.4; N, 4.2. $C_{15}H_{21}NO, CH_3I$ requires C, 51.5; H, 6.4; N, 3.8%).

5-Methyl-1-phenylhex-4-en-1-one (5).—2-Dimethylamino-5-methyl-1-phenylhex-4-en-1-one methiodide (4) (4.0 g), zinc dust (10.0 g), and 2N-sulphuric acid (25 ml) were kept at 100° for 15 min, then the mixture was steam-distilled until no more oily droplets appeared in the distillate.

Extraction with ether (2×75 ml) and evaporation followed by distillation gave 5-methyl-1-phenylhex-4-en-1-one (5) (1.5 g, 71%) as an oil, b.p. $85\text{--}87^\circ$ at 0.6 mmHg (Found: C, 82.7; H, 8.4. $C_{13}H_{16}O$ requires C, 83.0; H, 8.5%); τ 1.9—2.8 (m, 5 aromatic H), A_2B_2X system, τ_A 7.0, τ_B 7.6, τ_X 4.80 [$J_{AB} 7$, $J_{AX} 2$, $J_{BX} 7$ Hz, $=CO-C(H_A)_2-C(H_B)_2-CH_X-$], and 8.30 and 8.37 (two s, $=CMe_2$). Its 2,4-dinitrophenylhydrazone crystallised as orange prisms, m.p. 134° (Found: C, 61.7; H, 5.4; N, 15.3. $C_{19}H_{20}N_4O_4$ requires C, 62.0; H, 5.4; N, 15.2%).

Thermal Rearrangement of 2-Dimethylamino-1,3-diphenylpent-4-en-1-one (Diastereoisomers A and B) (2c). Formation of 2-Dimethylamino-1,5-diphenylpent-4-en-1-one (3c).—2-Dimethylamino-1,3-diphenylpent-4-en-1-one (2c) (diastereoisomers A and B)¹ (5.0 g) was kept in a sealed evacuated tube at 120° for 15 h and 2-dimethylamino-1,5-diphenylpent-4-en-1-one (3c) (5.0 g, 100%) was obtained as a pale yellow oil; τ 1.8—2.8 (m, 10 aromatic H), ABMNX system, τ_A 3.50, τ_B 3.85, τ_M, τ_N 7.1—7.5, τ_X 5.74 ($J_{AB} 16$, $J_{BM} = J_{BN} = 7$, $J_{MX} 5$, $J_{NX} 8$ Hz, $CH_A=CH_B-CH_MCH_N-CH_X-CO$), and 7.60 (s, NMe_2). It was characterised as its methiodide, which crystallised from ether-ethyl acetate-methanol as prisms, m.p. 180° (Found: C, 57.0; H, 5.6; N, 3.3. $C_{20}H_{24}INO$ requires C, 57.0; H, 5.7; N, 3.3%).

Thermal Rearrangement of 2-Dimethylamino-1-phenyl-3-(o-tolyl)pent-4-en-1-one (Diastereoisomers A and B) (2d). Formation of 2-Dimethylamino-1-phenyl-5-(o-tolyl)pent-4-en-1-one (3d).—2-Dimethylamino-1-phenyl-3-(o-tolyl)pent-4-en-1-one (2d) (diastereoisomers A and B)¹ (500 mg) was kept in a sealed evacuated tube at 115° for 15 h and 2-dimethyl-

amino-1-phenyl-5-(o-tolyl)pent-4-en-1-one (3d) (500 mg, 100%) was obtained as a pale yellow oil; τ 1.8–2.9 (m, 9 aromatic H), ABMNX system, τ_A 3.34, τ_B 4.00, τ_M , τ_N 7.0–7.6, τ_X 5.78 (J_{AB} 15.5, $J_{BM} = J_{BN} = 7$, J_{MX} 6, J_{NX} 8 Hz, $CH_A=CH_B-CH_MH_N-CH_X-CO$), 7.58 (s, NMe_2), and 7.75 (s, CH_3). It was characterised as its *methiodide* which crystallised from ether–ethyl acetate–methanol as prisms, m.p. 145° (Found: C, 57.8; H, 6.0; N, 3.3. $C_{21}H_{26}INO$ requires C, 57.9; H, 6.0; N, 3.2%).

Thermal Rearrangement of 4-Dimethylamino-3,3-dimethyl-4-(p-nitrophenyl)but-1-ene (2e). *Formation of 5-Dimethylamino-2-methyl-5-(p-nitrophenyl)pent-2-ene* (3e) (with D. J. Yarrow).—4-Dimethylamino-3,3-dimethyl-4-*p*-nitrophenylbut-1-ene **1** (2e) (1.0 g) was kept in a sealed evacuated tube at 180° for 30 min then distilled giving *5-dimethylamino-2-methyl-5-(p-nitrophenyl)pent-2-ene* (3e) (0.76 g, 76%) as a pale brown oil, b.p. 121–122° at 0.2 mmHg (Found: C, 67.9; H, 7.9; N, 11.3. $C_{14}H_{20}N_2O_2$ requires C, 67.7; H, 8.1; N, 11.3%), identical to a sample described in Part 2.¹

Thermal Rearrangement of 4-Dimethylamino-4-p-nitrophenyl-3-phenylbut-1-ene (*Diastereoisomer A*) (2f). *Formation of 4-Dimethylamino-4-p-nitrophenyl-1-phenylbut-1-ene* (3f) (with K. Reynard).—4-Dimethylamino-4-*p*-nitrophenyl-3-phenylbut-1-ene (2f) (*diastereoisomer A*) (150 mg) was kept in a sealed evacuated tube at 145° for 1 h giving *4-dimethylamino-4-p-nitrophenyl-1-phenylbut-1-ene* (3f) (150 mg, 100%), identical to a sample described in Part 2.¹

Thermal Rearrangement of (\pm)-4,4-Dimethyl-3-dimethylaminohex-5-en-2-one (2g). *Formation of (\pm)-3-Dimethylamino-6-methylhept-5-en-2-one* (3g).—(\pm)-4,4-Dimethyl-3-dimethylaminohex-5-en-2-one **1** (2g) (4.0 g) was kept in a sealed evacuated tube at 160° for 2 h. Distillation gave (\pm)-3-dimethylamino-6-methylhept-5-en-2-one (3g) (3.5 g, 87%) as an oil, b.p. 78° at 8 mmHg (Found: C, 70.8; H, 11.2; N, 8.2. $C_{10}H_{19}NO$ requires C, 71.0; H, 11.2; N, 8.3%); n.m.r.: AX_2Y system, τ_A 4.94, τ_X 7.5–7.8, τ_Y 7.02 [J_{AX} 7, J_{XY} 7 Hz, $=CH_A-C(H_X)_2-CH_Y-CO$], 7.72 (s, NMe_2), 7.85 (s, CH_3CO), and 8.3 (m, two vinylic Me). It was characterised as the *picrate* which crystallised from ethanol as yellow prisms, m.p. 123° (Found: C, 48.4; H, 5.6; N, 14.3. $C_{16}H_{22}N_4O_8$ requires C, 48.2; H, 5.2; N, 14.1%).

Resolution of (\pm)-4,4-Dimethyl-3-dimethylaminohex-5-en-2-one (2g).—The racemate (2g) (1.64 g) in chloroform (20 ml) was added to (+)-camphorsulphonic acid (2.4 g) in ethanol (20 ml). Ether (100 ml) was then added, and after 3 days the precipitate was collected and crystallised from ether–ethyl acetate giving (+)-4,4-dimethyl-3-dimethylaminohex-5-en-2-one *camphorsulphonate* (1.2 g) as prisms, m.p. 116° (Found: C, 59.7; H, 8.7; N, 3.5. $C_{20}H_{30}NO_5S$ requires C, 59.7; H, 9.0; N, 3.5%).

(+)-4,4-Dimethyl-3-dimethylaminohex-5-en-2-one *camphorsulphonate* (1.0 g) was added to a saturated solution of sodium hydrogencarbonate (50 ml) and the solution was extracted with ether (3 \times 25 ml). Evaporation yielded (+)-4,4-dimethyl-3-dimethylaminohex-5-en-2-one (400 mg), $[\alpha]_D^{23} +47^\circ$ (c 1.9% in methanol), identical in all other respects with the racemate.

Thermal Rearrangement of (+)-4,4-Dimethyl-3-dimethylaminohex-5-en-2-one (2g). *Formation of (\pm)-3-Dimethylamino-6-methylhept-5-en-2-one* (3g).—(+)-4,4-Dimethyl-3-dimethylaminohex-5-en-2-one (2g) (500 mg) was kept in a sealed evacuated tube at 160° for 2 h giving (\pm)-3-dimethylamino-6-methylhept-5-en-2-one (3g) (500 mg, 100%), $[\alpha]_D^{20}$ 0.0 (c 2% in MeOH), identical with the material already described.

4,4-Dimethylhex-5-en-2-one (7).—4,4-Dimethyl-3-dimethylaminohex-5-en-2-one (10.0 g) and methyl iodide (20 ml) were heated under reflux for 30 min, cooled, and diluted with ether (100 ml). The ether was decanted and the residual product was dissolved in water (25 ml) and heated with 5*N*-sulphuric acid (20 ml) and zinc dust (10.0 g) at 100° for 15 min. The solution was cooled, filtered, and extracted with ether (2 \times 50 ml). Evaporation of the dried extracts followed by distillation gave *4,4-dimethylhex-5-en-2-one* (7) (4.2 g, 57%) as an oil, b.p. 45–50° at 10 mmHg (lit.,²⁶ 45–50° at 30 mmHg) (Found: C, 76.4; H, 11.3. $C_8H_{14}O$ requires C, 76.3; H, 11.1%); n.m.r.: ABX system, τ_A 5.05, τ_B 5.07, τ_X 4.02 (J_{AB} 2, J_{AX} 18, J_{BX} 10 Hz, $CH_AH_B=CH_X-$), 7.55 (s, CH_2CO), 7.90 (s, CH_3CO), and 8.88 (s, $>CMe_2$). The ketone (7) remained unchanged after heating at 160° for 2 h and at 210° for 12 h.

Thermal Rearrangement of Methyl 3,3-Dimethyl-2-dimethylaminopent-4-enoate (2h). *Formation of Methyl 2-Dimethylamino-5-methylhex-4-enoate* (3h) (with E. Dickinson).—Methyl 3,3-dimethyl-2-dimethylaminopent-4-enoate **1** (2h) (1.0 g) was kept in a sealed evacuated tube at 205° for 15 h and the products were separated by preparative t.l.c. [silica gel, chloroform–ether (1 : 1)] into unchanged methyl 3,3-dimethyl-2-dimethylaminopent-4-enoate (2h) (47%) and *methyl 2-dimethylamino-5-methylhex-4-enoate* (3h) (53%); n.m.r.: AM_2X system, τ_A 4.90, τ_M 7.4–7.7, τ_X 6.87 [J_{AM} 7.5, J_{MX} 7.5 Hz, $=CH_A-C(H_M)_2-CH_X-$], 6.30 (s, CO_2Me), 7.68 (s, NMe_2), and 8.30 and 8.38 (two s, vinylic CMe_2). Methyl 2-dimethylamino-5-methylhex-4-enoate was characterised as its *picrate*, yellow prisms, m.p. 110°, from ethanol (Found: C, 46.1; H, 5.2; N, 13.8. $C_{16}H_{22}N_4O_9$ requires C, 46.4; H, 5.3; N, 13.5%).

Thermal Rearrangement of Methyl 2-Dimethylamino-2,3,3-trimethylpent-4-enoate (2j). *Formation of Methyl 2,5-Dimethyl-2-dimethylaminohex-4-enoate* (3j).—Methyl 2-dimethylamino-2,3,3-trimethylpent-4-enoate **1** (2j) (5.0 g) was kept in a sealed evacuated tube at 120° for 1 h. Distillation yielded *methyl 2,5-dimethyl-2-dimethylaminohex-4-enoate* (3j) (4.5 g, 90%) as a liquid, b.p. 110° at 15 mmHg (Found: C, 66.6; H, 10.7; N, 7.1. $C_{11}H_{21}NO_2$ requires C, 66.3; H, 10.5; N, 7.0%), identical with the sample already described in Part 2.¹

Thermal Rearrangement of 2-Dimethylamino-2,3,3-trimethyl-1-phenylpent-4-en-1-one (2k). *Formation of 2,5-Dimethyl-2-dimethylamino-1-phenylhex-4-en-1-one* (3k).—A mixture (5 g) of 2-dimethylamino-2,3,3-trimethyl-1-phenylpent-4-en-1-one (2k) (40%) and 2,5-dimethyl-2-dimethylamino-1-phenylhex-4-en-1-one **1** (60%) was kept in a sealed evacuated tube at 100° for 10 min, then distilled, giving *2,5-dimethyl-2-dimethylamino-1-phenylhex-4-en-1-one* (3k) (4.7 g, 94%) as a pale brown oil, b.p. 89–90° at 0.05 mmHg (Found: C, 78.3; H, 9.1; N, 5.7. $C_{16}H_{23}NO$ requires C, 78.4; H, 9.4; N, 5.7%); τ 1.4–2.8 (m 5 aromatic H), ABX system, τ_X 5.15, τ_A , τ_B 7.3–7.7 ($J_{AX} = J_{BX} = 7.5$ Hz, $=CH_X-CH_AH_B$), 7.71 (s, NMe_2), 8.55 and 8.72 (two s, $=CMe_2$), and 8.82 (s, $CH_3-C-NMe_2$).

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