Trimethylamine *N*-Oxide as a Precursor of Azomethine Ylides

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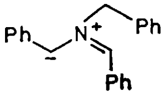
The first azomethine ylide devoid of a stabilizing group, generated by treating trimethylamine *N*-oxide with lithium di-isopropylamide, undergoes a ready [3 + 2] cycloaddition with various simple alkenes to give high yields of the corresponding pyrrolidines.



(A)

The reactivity of *N*-oxides on treatment with strong bases has been shown to be of anionic[1] (quinuclidine *N*-oxide) or radical[2] type (benzylic *N*-oxides).† We have now found that trimethylamine *N*-oxide (1), on treatment with lithium di-isopropylamide, generates the azomethine ylide (I). This reaction is of importance since azomethine ylides were hitherto known to be formed only from precursors bearing electron-withdraw-

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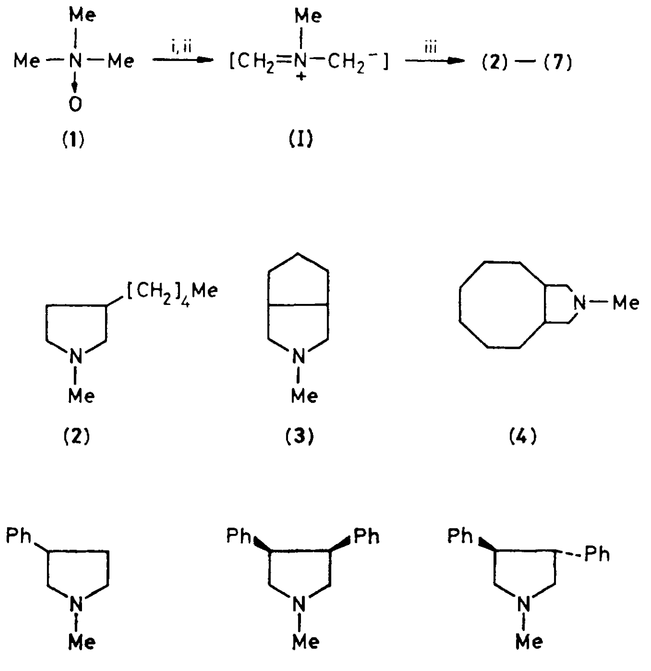
† The reactivity of *N*-substituted dibenzylamine *N*-oxides on treatment with n-butyl-lithium to give *cis*-2,3-diphenylaziridines, as recently reported by H. Takayama and T. Nomoto (*J. Chem. Soc., Chem. Commun.*, 1982, 408) could also in our opinion be considered to be radical like, since they failed to trap the postulated azomethine ylide (A) with an excess of dipolarophile (unspecified).



Scheme 1. Reagents: i, (**1**) (freshly sublimed) (1 mmol); LiPr$^i_2$N (4 mmol), tetrahydrofuran, −78 °C; ii, alkene (hept-1-ene, cyclopentene, cyclo-octene, styrene, *trans*-stilbene, or *cis*-stilbene) (1.1 mmol); iii, work-up (after warming to room temperature) by pouring into water and acid–base extraction with methylene dichloride. (**2**) 63%; m.p. (picrate) 134—135 °C; (**3**) 42%; m.p. (picrate) 203—205 °C; (**4**) 90%; m.p. (picrate) 218—219 °C; (**5**) 57%; m.p. (picrate) 158—160 °C; lit.[4] 155—158 °C; (**6**) 62%; m.p. 67—69 °C; (**7**) 72%; m.p. (picrate) 186—187 °C. All compounds have spectral data (mass, n.m.r.) and elemental analyses consistent with the structures.

ing or conjugative substituents, by thermal ring opening of aziridines or to a lesser extent by deprotonation of iminium salts.[3] To the best of our knowledge no example of a non-stabilized azomethine ylide has been reported.

The intermediate (I) is a very reactive species since it is trapped by simple alkenes (hex-1-ene, cyclopentene, or cyclohexene) whereas activated dipolarophiles (ethylenedicarboxylic esters, phenylmaleimide, *etc.*) were required for reactions with stabilized 1,3 dipoles.

Thus, the pyrrolidines (**2**), (**3**), and (**4**) were obtained from the reactions in Scheme 1. That the intermediate is (I) an azomethine ylide is strongly suggested by the following. (i) Reaction with styrene takes place easily, giving 1-methyl-3-phenylpyrrolidine (**5**) without the polymerization which might have been induced if a radical species had been present. (ii) *cis*- or *trans*-Stilbene gives *cis*- or *trans*-1-methyl-3,4-diphenylpyrrolidine (**6**) or (**7**) as expected for a [2 + 3] cycloaddition process.

We expect that this reaction may be useful synthetically for producing non-activated pyrrolidine ring compounds.

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References

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