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Microwave-Induced 1,3-Dipolar Cycloaddition of 2-Aroyl-aziridines

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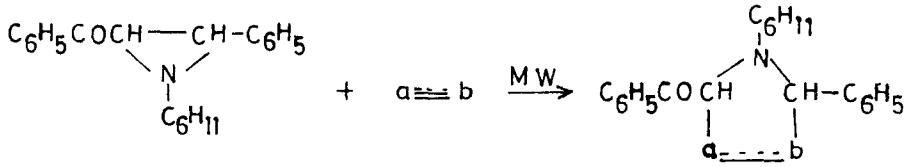
Abstract: 2-Aroyl-aziridines react with a variety of multiple bonds under microwave irradiation in an Erlenmeyer flask at ambient pressure to give the corresponding cycloadducts in high yields within few minutes.

The application of microwave energy to effect a variety of chemical processes has blossomed rapidly in the past few years. From its use in moisture analysis[1] it has been applied to a number of organic reactions, such as esterification, etherification, hydrolysis, Claisen, Diels-Alder (4+2), Reformatsky, Knoevenagel and Bischler Napieralski reactions[2]. However, to our knowledge, there are no literature reports on 1,3-dipolar cycloaddition reactions. Many reviews[2] have been published on microwaves and most reports describe some very important accelerations in reaction rate. In this letter we report the first example of 1,3-dipolar cycloaddition of t-2-benzoyl-1-cyclohexyl-3-phenyl aziridine to multiple bonds under microwave irradiations within a few minutes and without solvent. The reaction involving cleavage of the 2,3-bond of 2-aroyl-aziridines to an azomethine ylide intermediate and subsequent [2+3] cycloaddition to a multiple bond leads to the formation of oxazolidines, imidazolidines, naphtho-oxazoles and pyrrolines in high yields.



1 2 3

In a typical experiment, t-2-benzoyl-1-cyclohexyl-3-phenylaziridine[3] 1 (0.305g, 1 mmol) and p-nitrobenzaldehyde (0.15g, 1 mmol) were mixed together without solvent in an Erlenmeyer flask and placed in a commercial microwave oven operating at 2450 MHz and irradiated for 15 mins. The reaction mixture was allowed to reach room temperature then extracted with benzene. The residue, after removal of solvent, is purified by thin layer chromatography to afford t-4-benzoyl-3-cyclohexyl-c-5-(4-nitro phenyl)-r-2-phenyl oxazolidine[4] in 75% yields. Similarly 2-benzoyl-1-cyclohexyl-3-phenyl-aziridine was reacted with 1-nitrosonaphth-2-ol, which underwent 1,3-dipolar cycloaddition to the nitrogen-oxygen bond and subsequent cleavage of the intermediate oxadiazolidine to a nitrone[5] and cyclisation of the latter afforded both 2-benzoyl naphtho[1,2-d]oxazole and 2-phenyl naphtho[1,2-d]oxazole in good yields (Table). When cinnamylidene aniline or DMAD was reacted under identical conditions and usual work-up , the corresponding 1-cyclohexyl-2-benzoyl-3-styryl 4,5-diphenyl imidazolidine[6] or 1-cyclohexyl-5-phenyl 3,4-carbomethoxy-2-benzoyl-2-pyrroline[7] was obtained in 10-12 mins and 70-75% yields. The conventional methods needs 18-20 hr.

to reach completion. All the compounds obtained were confirmed by infrared and ^1H NMR spectra and finally by comparison (mp, TLC) with authentic samples.

In conclusion, it is noteworthy to mention that this simple and easily reproducible technique affords various cycloadducts in just one-pot, in shorter reaction time and with better yields than the classical homogeneous reactions in solvents[8].

Table: Cycloaddition of t-2-Benzoyl-1-cyclohexyl-3-phenyl-aziridine **1** with different dipolarophiles **2**.

Entry	Dipolarophile a⎓b	Product type	Microwave		Lit.. Conditions	
			Time min.	Yield[a] %	Time hr.	Yield %
1	4-Nitro benzaldehyde	t- 4-Benzoyl-3-cyclohexyl-c-5-(4-nitrophenyl)-r-2-phenyloxazolidine[4]	15	80	11	82
2	Dimethyl acetylene dicarboxylate	1-Cyclohexyl-5-phenyl 3,4-carbo-methoxy-2-benzoyl-2-pyrroline[7]	10	70	18	-
3	1-Nitrosonaphth-2-ol	2-Benzoyl naphtho[1,2-d]oxazole[5]	12	50	-	41.5
		2-Phenyl naphtho[1,2-d]oxazole[5]	12	35	-	40
4	Cinnamylidene aniline	1-Cyclohexyl-2-benzoyl 3-styryl 4,5-diphenylimidazolidine[6]	12	75	28	45
5	4-Chloro cinnamylidene aniline	1-Cyclohexyl-2-benzoyl-3-styryl-4-(4-chlorophenyl)-5-phenyl imidazolidine[6].	10	70	24	50

[a]Yields refer to the yield of pure isolated products.

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References:

1 Hesek, J.A. and Wilson, R.C., *Anal. Chem.*, **1974**, *46*, 1160.
2 For recent reviews on microwave irradiation see: Caddick, S., *Tetrahedron*, **1995**, *51*, 10403;Mingos, D.M. and Baghurst, D.R., *Chem. Soc. Rev.*, **1991**, *20*, 1-47; Giguere, R.J. in *Organic Synthesis: Theory and Application*, JAI Press, **1989**, *1*,103-172; Bose, A.K., Manhas, M.S., Ghosh, M., M., Raju, V.S., Bari, S.S., Newaz, S.N., Banik, B.K.,Choudhury, A.G.and Borakat, K.J., *J. Org. Chem.*, **1991**, *56*, 6968.
3 For the preparation of t-2-benzoyl-1-cyclohexyl-3-phenyl aziridine see: Cromwell, N.H., Barker, N.G., Wankel, R.A., Vanderhorst, P.J., Olson, F.W. and Anglin, J.H., *J. Am. Chem. Soc.*, **1951**, *37*, 1044.
4 Dallas, G., Lown, J.W. and Moser, J.P., *J. Chem. Soc.(C).*, **1970**, 2383.
5 Lown, J.W. and Moser, J.P., *Chem. Commun.*,**1970**, 247.
6 Sain, B. and Sandhu, J.S., *Ind. J. Chem.*, **1985**, *24B*, 291.
7 Padwa, A.and Hamilton, L., *Tetrahedron Lett.*, **1965**, *48*, 4363.
8 The reaction (entry 1) also proceeds smoothly when carried out under ultrasound in homogeneous medium.

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