(CH), 1200 (P=O), 1280, 975, 735 cm^{-1}; MS *m/e* 212, 168, 92.

Titration of an aliquot of the aqueous layer indicated 93.4% iodide ion release.

An authentic sample of *N,N,N',N'*-tetramethylphenylphosphonamide, mp 82.5–84 °C, with NMR, IR, and mass spectra identical with those for the product just described, was prepared by condensation of phenylphosphonic dichloride with dimethylamine in diethyl ether solution at temperature below 15 °C.

Competitions between Iodo- and Bromobenzene. The following procedure is representative. Into a three-necked, 125-mL round-bottom flask equipped with a cold finger condenser charged with solid CO_2 and 2-propanol, nitrogen inlet, and magnetic stirrer, was condensed ca. 50 mL of ammonia distilled from sodium. To the ammonia were added freshly resublimed potassium *tert*-butoxide (0.47 g, 4.2×10^{-3} mol), *N,N,N',N'*-tetramethylphosphonamide (0.54 g, 4.0×10^{-3} mol), iodobenzene (0.0626 g, 3.07×10^{-4} mol), and bromobenzene (0.526 g, 3.35×10^{-3} mol), and the mixture was stirred briefly. (The iodo- and bromobenzene were previously weighed and mixed in a small vial, and it was shown by weighing that no appreciable residue remained in the vial or in the pipet used to transfer the mixture.) The flask was placed in the photochemical reactor equipped with only four lamps symmetrically disposed, and the lamps were turned on for 45 s. The mixture was quickly acidified with ammonium nitrate, and 50 mL of diethyl ether was added. The entire process, from addition of the phenyl halides to acidification, took about 60 s. The ammonia was allowed to evaporate, 25 mL of water was added, the phases were separated, and the water phase was extracted with diethyl ether (2×50 mL). The aqueous phase was diluted to 100 mL, and 25 mL aliquots were titrated with 0.07 M $AgNO_3$ potentiometrically. The iodide and bromide ion end points were easily identified; in a representative run, 4.71 mL of $AgNO_3$ solution was required to attain the iodide ion end point and a further 0.14 mL for the bromide ion end point.

Rate constant ratios were reckoned by means of eq 7.[30]

$$k_{PhI}/k_{PhBr} = ([ArBr]_0/[Br^-]_t) \ln ([PhI]_0/([PhI]_0 - [I^-]_t)) \quad (7)$$

Acknowledgment. This research was supported in part by the National Science Foundation. Acquisition of the Finnigan Model 4000 GC/MS equipment was assisted by the NSF Chemical Instrumentation Program.

Registry No. $PhP(OBu)O^-K^+$, 71774-84-0; $Ph_2PO^-K^+$, 19115-01-6; $(EtO)_2PS^-K^+$, 71774-85-1; $(Me_2N)_2PO^-K^+$, 71774-86-2; PhI, 591-50-4; PhBr, 108-86-1; $Ph_2P(O)OBu$, 20610-34-8; Ph_3PO, 791-28-6; $PhP(S)(OEt)_2$, 6231-03-4; $PhP(O)(NMe_2)_2$, 3732-83-0; PhH, 71-43-2.

(30) Bunnett, J. F. In "Investigation of Rates and Mechanisms of Reactions", 3rd ed.; Lewis, E. S., Ed.; Wiley-Interscience: New York, 1974; Part I, p 159.

Occurrence of an S_N(ANRORC) Mechanism in the Chichibabin Amination of 4-Phenylpyrimidine[1]

Koos Breuker and Henk C. van der Plas*

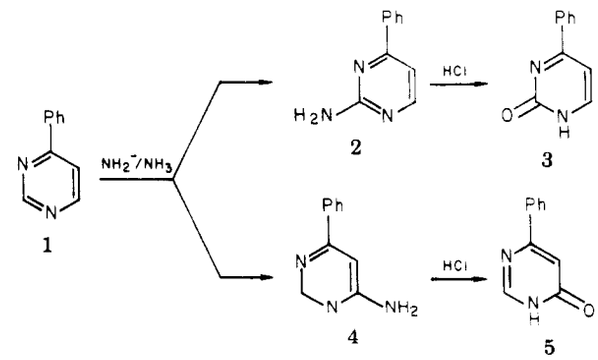
Laboratory of Organic Chemistry, Agricultural University, 6703 BC Wageningen, The Netherlands

Received June 11, 1979

On treatment of 4-phenylpyrimidine with potassium amide in liquid ammonia, a mixture of 6-amino-4-phenylpyrimidine and 2-amino-4-phenylpyrimidine is obtained. It was proven that in the formation of the 2-amino compound the pyrimidine ring is opened but that in the formation of the 6-amino compound *no* ring opening is involved. ^1H and ^{13}C NMR spectroscopy gave evidence for formation of two σ adducts between the amide ion and 4-phenylpyrimidine, i.e., the 6-amino-1(or 3),6-dihydropyrimidinide ion and the 2-amino-1(or 3),2-dihydropyrimidinide ion.

During the last decade the amination of aza aromatics containing groups with considerable leaving character has been a subject of continuous interest; special attention is paid to the use of potassium amide in liquid ammonia as an aminating reagent. Sound proof is obtained—on the basis of application of ^{15}N-labeled compounds and NMR spectroscopy—that in an overwhelming number of cases the replacement of the leaving group by the amino group takes place for the greater part according to a so-called S_N(ANRORC) mechanism, describing a reaction sequence involving an *A*ddition of the *N*ucleophile to the heterocycle, *R*ing *O*pening, and *R*ing *C*losure reaction.[2]

Scheme I



In order to extend these studies we became interested as to whether the Chichibabin amination of aza aromatics by potassium amide (i.e., the replacement of a hydrogen by an amino group) would also occur according to an S_N(ANRORC) mechanism. Up to now only one example of a Chichibabin reaction has been reported in which the

(1) (a) Part 78 on pyrimidines from this laboratory. For part 77 see: N. J. Kos, H. C. van der Plas, and A. van Veldhuizen, *J. Org. Chem.*, **44**, 3140 (1979). (b) Part 25 on the S_N(ANRORC) mechanism. For part 24 see: H. C. van der Plas, *Acc. Chem. Res.*, **11**, 462 (1978). For part 23 see: C. A. H. Rasmussen and H. C. van der Plas, *Tetrahedron Lett.*, 3841 (1978). (c) Part 23 on σ adducts. For part 22 see: A. Nagel, H. C. van der Plas, G. Geurtsen, and A. van Veldhuizen, *J. Heterocycl. Chem.*, **16**, 301 (1979).

(2) For a review, see the first reference in 1b.

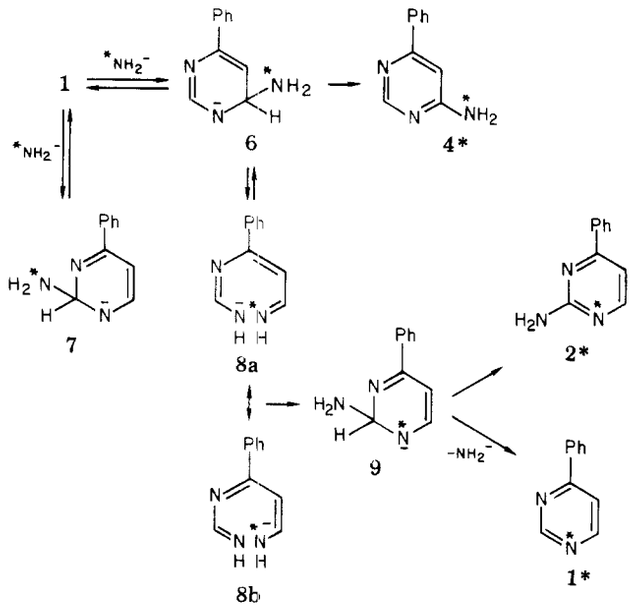
0022-3263/79/1944-4677$01.00/0

Table I. ^{15}N Excess in Recovered 1*, in the Amino Compounds 2* and 4*, and in the Oxo Compounds 3* and 5*

compd	% ^{15}N excess[a]	% ANRORC process[b]
1*	3.2 (2.4)	38 ± 5
2*	8.7 (6.3)	92 ± 5
3*	8.3 (5.6)	
4*	8.5 (6.5)	5 ± 5
5*	0.3 (0.4)	

[a] The values in parentheses refer to duplicate experiments. [b] Average values.

Scheme II



*N = ^{15}N

S_N(ANRORC) mechanism partly operates, i.e., in the amination of phenyl-*s*-triazine into aminophenyl-*s*-triazine by potassium amide in liquid ammonia.[3]

Since pyrimidines were found to be very appropriate systems to undergo S_N(ANRORC) substitutions,[2] we started an investigation on the Chichibabin amination of 4-phenylpyrimidine (**1**). The choice of 4-phenylpyrimidine instead of the parent substance pyrimidine was mainly based on the fact that in introductory experiments the amination of pyrimidine by potassium amide led to a more complex mixture of compounds.

On treatment of 1 equiv of **1** with 4 equiv of potassium amide in liquid ammonia at −33 °C for 70 h, two products were formed. On the basis of NMR spectroscopy these products were identified as the isomers 2-amino-4-phenylpyrimidine (**2**) and 6-amino-4-phenylpyrimidine (**4**). (Scheme I). The two compounds did not show depression of the melting points when mixed with reference compounds. The yields were 60 and 15%, respectively, the remainder being mainly starting material.

In order to investigate whether in the formation of the two amino compounds a ring opening would be involved, we carried out the amination with ^{15}N-labeled potassium amide. If an S_N(ANRORC) process occurs, it leads to incorporation of ^{15}N in the ring; if not, the label will only be present in the exocyclic amino group.

(3) G. Simig and H. C. van der Plas, *Recl. Trav. Chim. Pays-Bas*, **95**, 125 (1976).

Table II. Proton NMR Spectra of 1, 6, and 7[g]

proton	compd 1[a]	6[b]	7[b]
H-2	9.22	7.2	4.26
H-5	7.64	4.92[c]	5.43[d]
H-6	8.69	4.71[c]	*e*
ortho H[f]	8.0	7.5	7.5
meta and para H[f]	7.4	7.2	7.2

[a] In $CDCl_3$. [b] In KNH_2/NH_3. [c] $J_{5,6}$ = 4 Hz. [d] $J_{5,6}$ = 5 Hz. [e] This signal probably lies under the multiplets of the phenyl signals. [f] The hydrogens present in the phenyl group. [g] Chemical shifts in δ units, relative to Me_4Si.

Table III. ^{13}C NMR Chemical Shifts and Coupling Constants of 1, 6, and 7[a]

atoms	compd 1[b]	6[c]	7[c]
	Chemical Shifts		
C2	159.3	156.8	84.6
C4	164.0	146.3	
C5	117.2	93.4	88.1
C6	157.5	63.7	155.6
C1	138.4	143.0	
C2′,6′	127.3	125.4	
C3′,5′	129.1	127.9	
C4′	131.2	126.3	
	$^1J_{CH}$ Coupling		
C2-H	199	176	149
C5-H	170	157	158
C6-H	179	149	168

[a] Chemical shifts in ppm relative to Me_4Si. Coupling constants ($^1J_{CH}$) in Hz. [b] In $CDCl_3$. [c] In KNH_2/NH_3.

In order to be able to establish whether ^{15}N was present in the pyrimidine ring and/or in the amino group of the labeled amino products **2*** and **4***, these compounds were converted into the corresponding 4-phenylpyrimidin-2-one (**3***) and 4-phenylpyrimidin-6-one (**5***) by hydrolysis in concentrated hydrochloric acid. The excess of ^{15}N in compounds **2***, **3***, **4***, and **5*** as found by mass spectrometric measurement is given in Table I.

These results clearly show that the formation of **2** almost exclusively occurs via the ring opening, ring closure mechanism. On the contrary, product **4** appears to be formed without following this S_N(ANRORC) process.

These different routes of formation of both amino products can reasonably be explained if we assume an initial attack of the ^{15}N-labeled amide ion on position 6 of the starting material **1**, yielding the 6-amino-1(or 3),6-dihydropyrimidinide ion **6** (Scheme II). The 4(6) position in pyrimidine is the preferred position for nucleophilic attack.[4] Adduct **6** can aromatize into the 6-amino compound **4***, having its ^{15}N in the amino group. In an alternative route **6** may undergo a ring opening into the resonance-stabilized species **8a** ↔ **8b**. Ring closure into the 2-amino-1(or 3),2-dihydropyrimidine **9**, followed by aromatization, yields the 2-amino compound **2***, having the ^{15}N incorporated in the ring.

In order to substantiate this mechanism we investigated by ^1H and ^{13}C NMR spectroscopy whether one or more of these suggested intermediates could be detected. Therefore, we measured the NMR spectra of **1** in the KNH_2/NH_3 system. The data are collected in Tables II and III.

(4) R. G. Shepherd and J. L. Fedrick, *Adv. Heterocycl. Chem.*, **4**, 291 (1965); J. A. Zoltewicz and L. S. Helmick, *J. Am. Chem. Soc.*, **94**, 682 (1962); R. E. van der Stoel and H. C. van der Plas, *Recl. Trav. Chim. Pays-Bas*, **97**, 116 (1978).

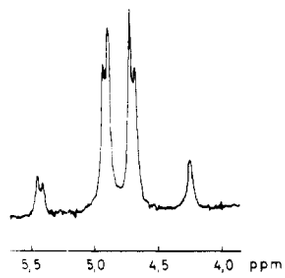


Figure 1. Part of the ^1H NMR spectrum, 20 min after dissolving 1 in KNH_2/NH_3 at −50 °C.

In the ^1H NMR spectrum no signal of unreacted 1 was detected. A doublet at 4.71 ppm and a doublet at 4.92 ppm were observed, being ascribed to the protons at positions 6 and 5, respectively, in **6**. This assignment was based on comparison with the spectrum of 6-deuterio-4-phenylpyrimidine in KNH_2/NH_3, showing the absence of a signal at 4.71 ppm and at the same time disappearance of the coupling in the signal at 4.92 ppm. The signal for H-2 in **6** lies under the phenyl multiplet of the meta and para hydrogens at 7.2 ppm as indicated by the integration. The large upfield shift of 8.69 − 4.71 = 3.98 ppm for H-6, observed on comparison of the spectrum of 1 in KNH_2/NH_3 with that of 1 in deuterated chloroform, confirms the presence of a 1:1 anionic σ adduct formed by attack of the amide ion on the C-6 of the pyrimidine ring.

In addition, in the region of 4–5.5 ppm two small signals were also observed: a doublet at 5.43 ppm and a somewhat broadened singlet at 4.26 ppm. These signals are ascribed to H-5 and H-2, respectively, in the 2-amino-1(or 3),2-dihydropyrimidinide ion (**7**) (see Figure 1). These assignments could be made by measuring the spectra of the deuterated compounds 6-deuterio-4-phenylpyrimidine and 2-deuterio-4-phenylpyrimidine in KNH_2/NH_3. The spectrum of 6-deuterio-4-phenylpyrimidine shows a singlet at 5.43 ppm (besides the signal at 4.92 ppm as discussed before); the spectrum of 2-deuterio-4-phenylpyrimidine has no peak at 4.26 ppm. The signal for H-6 in **7** probably lies under the phenyl multiplet. The upfield shift for H-2, observed on the formation of adduct **7**, amounts to 9.22 − 4.26 = 4.96 ppm. The ratio of the adducts **6** and **7**, 20 min after dissolving 1 in KNH_2/KH_3, is about 80:20. It is evident that adduct **7** cannot act as an intermediate in the formation of **2** (in that case no ring opening is involved), since the ^{15}N-labeling experiments show that about 95% of **2** has been formed according to an S_N(ANRORC) process. We found, however, that on standing of the reaction mixture for several hours, the amount of **7** diminishes and finally disappears. Apparently we are dealing with the kinetically favored formation of **7**, which slowly converts via 1 into the more stable adduct **6**. A somewhat similar result has been found on dissolving quinoline in KNH_2/NH_3:[5] the adduct at position 2 slowly converts into the adduct at position 4.

^{13}C NMR spectroscopy of the solution of 1 in KNH_2/NH_3 confirms the formation of the two adducts **6** and **7**. The assignment of the signals, based upon selected proton decoupling, is given in Table III. Characteristic are the large upfield shifts of C-6 in **6** (157.5 − 63.7 = 93.8 ppm) and of C-2 in **7** (159.3 − 84.6 = 74.7 ppm) and the decrease in ^{13}C–H coupling constants (from 179 to 149 Hz and from 199 to 149 Hz), indicating the decrease in s character of the carbon–hydrogen bonding orbital on the site of amide attack, compared with 1 in $CDCl_3$ as a solvent.

All NMR data are in agreement with our assumption concerning **6** as intermediate in the amination reaction. The result is the first example of the formation of an aminodiazine formed by a Chichibabin reaction via an S_N(ANRORC) mechanism (Scheme II).

As seen in Scheme II, the formation of **2*** from **6** goes via intermediate **9**. This intermediate is quite analogous to adduct **7**, however, with the important difference of the position of the ^{15}N label. Since **7** is in equilibrium with **1**, **9** must be in equilibrium with **1***, the starting material 1 in which ^{15}N is incorporated. Checking the recovered starting material for the presence of ^{15}N, we found indeed that about 35–40% of ^{15}N was incorporated into **1**. This result gives additional evidence for the occurrence of the 2-amino-1(or 3),2-dihydropyrimidinide **9** as intermediate in the reaction course.

It is of interest to mention that we found no double labeling in the mass spectra of **2*** and **4***. Therefore, **1*** and **9** cannot be present in the liquid ammonia containing the potassium amide. If this had been the case, it should have led to the 2- and 6-amino compounds being *doubly* labeled. Since this is not found, we reach the conclusion that the formation of **4***, **2***, and **1*** takes place after the addition of the ammonium salt (neutralizing the amide ions) and that in KNH_2/NH_3 only **6** is present. A similar result has been found before.[6] The reaction **6** → **8** → **9** may proceed due to the temperature rise.

Experimental Section

Melting points are uncorrected. The ^1H NMR spectra of solutions in $CDCl_3$ were recorded on a Hitachi Perkin-Elmer R-24B spectrometer, using tetramethylsilane (δ 0) as internal standard. The ^1H NMR spectra of the solutions in liquid ammonia containing potassium amide and all ^{13}C NMR spectra were recorded on a Varian XL-100-15. In liquid ammonia the chemical shifts of the protons were measured against the ammonia signal (δ 0.95) as standard. In the ^{13}C NMR spectra trimethylamine (δ 47.5) was used as the reference compound. All these values are converted to the Me_4Si scale by adding the indicated values. Typical spectral parameters for ^{13}C NMR spectra were as follows: spectral width 5120 Hz (1.25 Hz/point), acquisition time 0.8 s, pulse delay 1.2 s, pulse width 10–20 μs.

Column chromatography was carried out over Merck silica gel 60 (70–230 mesh ASTM).

The excess of ^{15}N in the compounds investigated was calculated from the (M + 1)/M ratio, measured on an AEI MS-902 mass spectrometer.

Preparation of 6- and 2-Deuterio-4-phenylpyrimidine. 6-Deuterio-4-phenylpyrimidine. A suspension of 4-hydrazino-6-phenylpyrimidine in D_2O was refluxed for 90 min. After evaporation of the solvent NMR spectroscopy of the residue revealed a complete exchange of the protons of the hydrazino group. This product was subsequently heated with D_2O and silver acetate according to procedures previously described in the literature for similar conversions.[7] The resulting crude material was purified by preparative TLC using silica gel and 2% methanol in chloroform as eluent; yield 38%.

2-Deuterio-4-phenylpyrimidine. This compound was prepared in an analogous way as 6-deuterio-4-phenylpyrimidine by starting from 2-hydrazino-4-phenylpyrimidine.

Amination. The amination reaction was performed as described before;[8] however, 10 mol % of potassium nitrate was added.[9] After 70 h the potassium amide was destroyed by the

(5) J. A. Zoltewicz, L. S. Helmick, T. M. Oestreich, R. W. King, and P. E. Kandetzki, *J. Org. Chem.*, **38**, 1947 (1973).

(6) C. A. H. Rasmussen, H. C. van der Plas, P. Grotenhuis, and A. Koudijs, *J. Heterocycl. Chem.*, **15**, 1121 (1978).

(7) E. A. Oostveen and H. C. van der Plas, *Recl. Trav. Chim. Pays-Bas*, **93**, 114 (1974); H. C. van der Plas, P. Smit, and A. Koudijs, *Tetrahedron Lett.*, 9 (1968).

(8) A. Nagel and H. C. van der Plas, *Chem. Pharm. Bull.*, **23**, 2678 (1975). For labeled KNH_2/NH_3 see: A. Nagel and H. C. van der Plas, *Heterocycles*, **7**, 205 (1977).

(9) F. W. Bergstrom, *J. Org. Chem.*, **2**, 411 (1937).

addition of ammonium chloride. The ammonia was evaporated and ether/methanol was added to the residue. The mixture was separated into its components by column chromatography. Elution with petroleum ether (bp 60–80 °C)/chloroform (1:1) and chloroform gave the starting material 1 and 2-amino-4-phenylpyrimidine (2): mp 165–166 °C from petroleum ether (bp 60–80 °C); yield 60% (average of three experiments). Further elution with 4% methanol in chloroform gave 6-amino-4-phenylpyrimidine (4): mp 222–225 °C from benzene/ethanol (50/50); yield 15%.

Hydrolysis of the Amino Compounds. A 10-mg sample of 2- or 6-amino-4-phenylpyrimidine was heated with concentrated hydrochloric acid in a Carius tube (150 °C during 15 h). After neutralization the 4-phenylpyrimidones were purified by column chromatography using 3% methanol in chloroform as eluent.

Acknowledgment. We are indebted to Dr. C. A. Landheer and Mr. W. P. Combé for the mass spectrometric data and to Mr. A. van Veldhuizen for the NMR spectra of the adducts.

Registry No. 1, 3438-48-0; 1*, 71734-70-8; 2, 2305-87-5; 2*, 71734-71-9; 3, 38675-31-9; 3*, 71734-72-0; 4, 3435-29-8; 4*, 71734-73-1; 5, 4891-69-4; 5*, 40889-17-6; 6, 71734-74-2; 6*, 71734-75-3; 7, 71734-76-4; 7*, 71734-76-4; 6-deuterio-4-phenylpyrimidine, 71734-77-5; 2-deuterio-4-phenylpyrimidine, 71734-78-6; 4-hydrazino-6-phenylpyrimidine, 35594-08-2; 2-hydrazino-4-phenylpyrimidine, 71734-79-7.

Stereoselectivity in the Cycloaddition Reactions of 2-Ethoxy-3-methylisoquinolinium Salts[1]

Teh-Kuei Chen and Charles K. Bradsher*

Paul M. Gross Chemical Laboratory, Duke University, Durham, North Carolina 27706

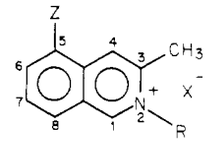
Received February 27, 1979

With styrenes, norbornenes, and cyclopentadiene, 2-ethoxy-3-methylisoquinolinium ion and its 5-nitro derivative undergo polar cycloaddition more readily than do their 2-methyl counterparts. Characteristic of these cycloadditions is a stereoselectivity which in many instances is 100% and which in nearly all instances is believed to have a polar origin.

The discovery[2-5] that 2,3-dimethylisoquinolinium salts (1) will undergo 1,4-addition both regioselectively and stereoselectively with alkyl vinyl ethers or cyclopentadiene was of theoretical interest but of limited synthetic value since less reactive alkenes (e.g., styrene) failed to react under any of the conditions tried. It was demonstrated that the reactivity of the isoquinolinium salt could be greatly enhanced if a nitro group was introduced at position 5 (cf. 2), permitting reaction with such relatively unactivated alkenes as β-pinene or norbornene.[6]

A promising alternate approach was suggested by earlier observations[7,8] that 2-alkoxyisoquinolinium salts show high reactivity toward the attack of nucleophiles. One of the implications of the electrophilic addition model[9-11] for cationic polar cycloaddition[12,13] is that enhanced electrophilicity of the cation should result in increased tendency toward cycloaddition.[14] The purpose of the present investigation was to determine whether cycloaddition was favored by replacing the *N*-alkyl group of quaternary isoquinolinium salts by *N*-ethoxyl and to learn whether the observed stereochemistry of addition was explicable in terms of the explanations presented earlier,[11] in particular with those parts dealing with stereoselectivity due to coulombic repulsion.

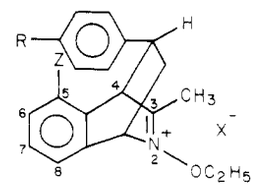
2-Ethoxy-3-methylisoquinolinium tetrafluoroborate (3, X = BF_4) can be prepared[15] conveniently by the action of triethyloxonium tetrafluoroborate on the *N*-oxide of 3-methylisoquinoline. With ethyl vinyl ether, 3 appeared



1, Z = H; R = CH_3
2, Z = NO_2; R = CH_3
3, Z = H; R = OC_2H_5
4, Z = NO_2; R = OC_2H_5

to induce immediate polymerization, and no pure cycloadduct was obtained. Similar results were obtained when the cation was 2-ethoxy-5-nitro-3-methylisoquinolinium tetrafluoroborate (4, X = BF_4).

The less reactive nucleophile *p*-methoxystyrene underwent simple cycloaddition, affording after 10 days a 96% yield of the expected adduct 5. From the ^1H NMR of the



5, R = OCH_3; Z = H
6, R = H; Z = NO_2
7, R = OCH_3; Z = NO_2

adduct 5 it is clear that only a single racemate is formed

(1) This research was supported by U.S. Public Health Service Research Grant No. HL02170 from the National Heart and Lung Institute of the National Institutes of Health.

(2) Bradsher, C. K.; Day, F. H. *Tetrahedron Lett.* **1971**, 409.
(3) Bradsher, C. K.; Day, F. H.; McPhail, A. T.; Wong, P.-S. *Tetrahedron Lett.* **1971**, 4205.
(4) Bradsher, C. K.; Day, F. H. *J. Heterocycl. Chem.* **1974**, *11*, 23.
(5) Bradsher, C. K.; Day, F. H.; McPhail, A. T.; Wong, P.-S. *J. Chem. Soc., Chem. Commun.* **1975**, 156.
(6) Day, F. H.; Bradsher, C. K.; Chen, T.-K. *J. Org. Chem.* **1975**, *40*, 1195.
(7) Feely, W. E.; Beavers, E. M. *J. Am. Chem. Soc.* **1959**, *81*, 4004.
(8) Katritzky, A. R.; Lunt, E. *Tetrahedron* **1969**, *25*, 4291.
(9) Bradsher, C. K.; Carlson, G. L. B.; Porter, N. A.; Westerman, I. J.; Wallis, T. G. *J. Org. Chem.* **1978**, *43*, 822.
(10) Westerman, I. J.; Bradsher, C. K. *J. Org. Chem.* **1978**, *43*, 3002.
(11) Westerman, I. J.; Bradsher, C. K. *J. Org. Chem.* **1979**, *44*, 727.
(12) Schmidt, R. R. *Angew. Chem., Int. Ed. Engl.* **1973**, *12*, 212.
(13) Bradsher, C. K. *Adv. Heterocycl. Chem.* **1974**, *16*, 289.
(14) Westerman, I. J.; Bradsher, C. K. *J. Org. Chem.* **1971**, *36*, 969.
(15) Reichardt, C. *Chem. Ber.* **1966**, *99*, 1769.

0022-3263/79/1944-4680$01.00/0