Reaction of Acridine with Pyrazolone Derivatives

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A mixture of acridine and a pyrazolone derivative was reacted in the solid state (without solvent). It is proposed that the enol tautomer (the C4-position) of the pyrazolone derivative attacks the C9-position of acridine through a nucleophilic reaction resulting in products where the C4-position of pyrazolone is connected to the C9-position of acridine. When the reaction of 3-methyl-1-phenyl-5-pyrazolone and acridine was carried out at low temperature (25°-50°), the reaction product was obtained even when the majority of the reaction mixture had not melted. The same reaction was also carried out in the presence of an ultrasonic wave at same temperature (25°-50°) and the reaction product was obtained in high yield. Under ultrasonic conditions, the reaction mixture was not melted. However, the interface between 3-methyl-1-phenyl-5-pyrazolone and acridine gradually changed from white to black. In this reaction, the dihydroacridine dimer is not obtained.

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Introduction.

Acheson *et al*. and Hamana *et al*. studied the reactions of pyridine and quinoline N-oxide compounds with nucleophilic reagents in solution [1-7]. Also mixtures of acridine and various vinyl ethers in methanol under reflux conditions have been shown to produce dihydroacridinyl derivatives [8].

We previously reported that the reaction of acridine with nucleophilic reagents without solvent produced condensation products [9,10]. For instance, acridine derivatives have been formed in high yields using mixture of acridinium chloride and aniline heated at 110 °C and from mixtures of acridine and methylindole derivatives heated at 160 °C. Pyrrole, pyrrole derivatives and imidazole derivatives have been used as nucleophilic reagents in the non-solvent reactions with acridine as well [11,12]. Furthermore, acridine and phenol or resorcinol were reacted at 210° without solvent [13], and finally an equimolar mixture of acridine and sodium amide was heated at 130 °C without solvent to successfully produce acridine derivatives [14].

Pyrrole is well known to be able to discharge a proton to become an anion. In the reaction, the proton migrates to the nitrogen atom of acridine to generate the N-protonated. In the N-protonated form, the C9-position of acridine is susceptible to nucleophilic substition [9,10].

In this study, we try to use the pyrazolone as the nucleophile in a non-solvent reaction with acridine. The carbonyl group at the C5-position of pyrazolone increases the electron density at the C4-position of pyrazole, which allows for nucleophilic substitution at the C9-position of acridine in manner similar to that of pyrole. It is possible that pyrazolone reacts at low temperature in the solid state.

This study reports on the reaction of π-deficient pyrazolone derivatives with acridine at low temperature without solvent. We examined 3-methyl-1-phenyl-5-pyrazolone, 3-methyl-5-pyrazolone, 1-(4'-nitrophenyl)-3-methyl-5-pyrazolone, 3-methyl-1-*p*-tolyl-5-pyrazolone, 1,3-dimethyl-5-pyrazolone and 2,3-dimethyl-1-phenyl-5-pyrazolone. The reaction mechanism is also discussed.

Results and Discussion.

Solid State Reaction of Acridine with Pyrazolone Derivatives.

Reaction of acridine with different pyrazolone derivatives at different temperatures is shown in Table 1 for a molar ratio of acridine:pyrazolone derivative of 1:2. The pyrazolone derivatives used were 3-methyl-1-phenyl-5-pyrazolone, 3-methyl-5-pyrazolone, 1-(4'-nitrophenyl)-3-methyl-5-pyrazolone, 3-methyl-1-*p*-tolyl-5-pyrazolone, 1,3-dimethyl-5-pyrazolone and 2,3-dimethyl-1-phenyl-5-pyrazolone. The resulting products were thus, 4-(9-acridinyl)-3-methyl-1-phenyl-5-pyrazol-5-ol **1** (black-purple needles)**,** 4-(9-acridinyl)-3-methyl-5-pyrazol-5-ol **2** (yellow needles), 4-(9-acridinyil)-1-(4'-nitrophenyl)-3-methyl-5-pyrazol-5-ol **3** (brown needles), 4-(9-acridinyl)-3-methyl-1-*p*-tolyl-5-pyrazol-5-ol **4 (**black-violet needles), 4-(9-acridinyl)-1,3-dimethyl-5-pyrazol-5-ol **5 (**red-brown needles) and 4-(9-acridinyl)-2,3-dimethyl-1-phenyl-5-pyrazol-5-ol **6 (**orange-yellow needles), respectively. The reaction conditions and yields of the reaction products are shown in Table 1 and Scheme 1.

It seems that the normal pyrazolone derivatives react by nucleophilic attack to the C9-position of acridine, suggesting that the pyrazolone ring takes the resonance structure of the enol form. All of these reactions of acridine and pyrazolone derivatives were considered to occur *via* the enol form of pyrazolone where the C4-position of pyrazolone attacks the C9-position of acridine, except for 2,3-dimethyl-1-phenyl-5-pyrazolone. Because 2,3-dimethyl-1-phenyl-5-pyrazolone has groups substituted at the 1-, 2-and 3-positions, the pyrazolone ring cannot adopt the enol form and thus the product must occur *via* the keto form. All products obtained from these reactions are C9

Scheme 1

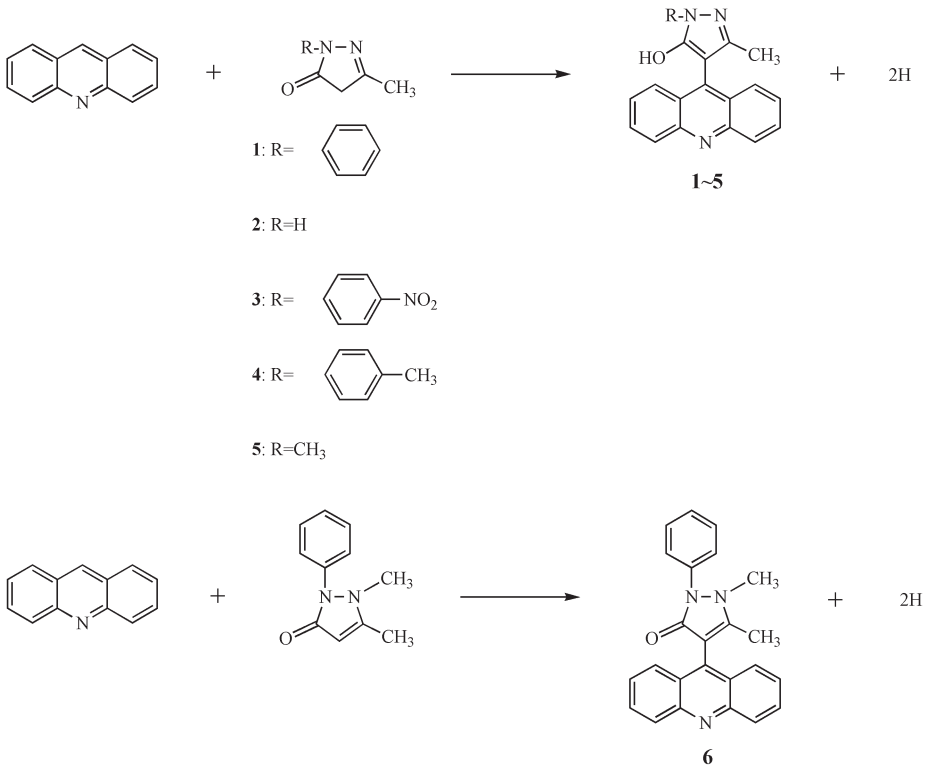


Table 1

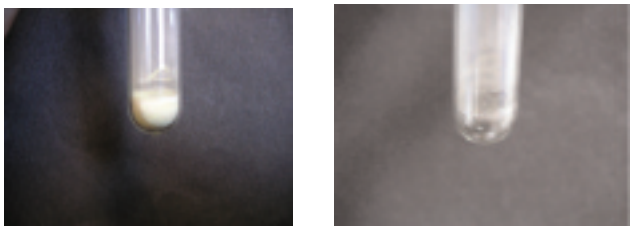
Reaction Products of Acridine with Pyrazolone Derivatives

Pyrazolons	Products	Reaction conditions Temp.(°)	Time (hr)	Yield (%)
3-methyl-1-phenyl-5-pyrazolone	**1**	Room Temp.	5	26.6
		30	5	32.6
		40	5	33.7
		50	5	35.5
		100	5	41.9
		150	5	52.9
3-methyl- 5-pyrazolone	**2**	50	5	0
		100	5	70.9
		150	5	90.9
1-(4'-nitorophenyl)-3-methyl -5-pyrazolone	**3**	50	5	0
		100	5	75.5
		50	5	92.8
3-methyl-1-*p*-tolyl-5-pyrazolone	**4**	50	5	19.5
		100	5	53.5
		150	5	41.2
1,3-dimethyl- 5-pyrazolone	**5**	50	5	0
		100	5	67.5
		150	5	82.1
2,3-dimethyl-1-phenyl-5-pyrazolone	**6**	50	5	0
		100	5	0
		150	5	35.0

substituted acridines. The yields of products varied for different structures of pyrazolone derivatives.

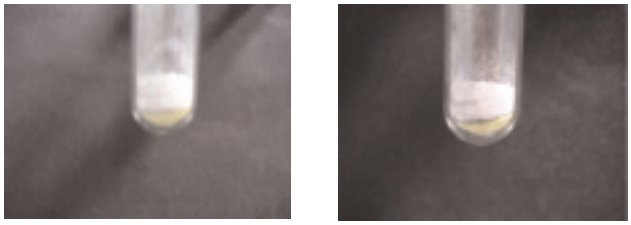
In particular, 3-methyl-1-phenyl-5-pyrazolone and 3-methyl-1-*p*-tolyl-5-pyrazolone reacted at lower temperatures (50°) even when it was observed that most of the raw material was not melted. Furthermore, the reaction occurred at room temperature for 3-methyl-1-phenyl-5-pyrazolone providing the product in high yield. Figure 1 (1) shows a photograph of the complete mixture of acridine and 3-methyl-1phenyl-5-pyrazolone before heating at 50°. Figure 1 (2) shows a color change of the mixture of acridine and 3-methyl-1phenyl-5-pyrazolone after heating at 50° for 5 hours. The color of the mixture changed from white to dark-gray. Figure 2 (1) shows a photograph of the interface of 3-methyl-1-phenyl-5-pyrazolone and acridine layers before heating at 50°. Figure 2 (2) shows the color change of the interface between the 3-methyl-1-phenyl-5-pyrazolone and acridine layers after heating at 50° for 5 hours. The color of the interface changed from white to black, suggesting that a reaction was occurring in the solid state.

2,3-Dimethyl-1-phenyl-5-pyrazolone is known to be acidic. In face, 2,3-dimethyl-1-phenyl-5-pyrazolone can even give up a proton in the keto form. As a result, a pyrazolone ring of 2,3-dimethyl-1-phenyl-5-pyrazolone takes an enamine structure under this reaction condition, and it is thought that an anion is generated at the C4-position of pyrazolone ring. Therefore, it was supposed that the reac-



(1) (2)

Fig.1 Change in state of mixture of acridine and 3-methyl-1phenyl-5-pyrazolone: (1) initially; (2) after heating for 5hr at 50°.



(1) (2)

Fig.2 Change in state of two-layer of acridine and 3-methyl-1phenyl-5-pyrazolone: (Upper part: 3-methyl-1-phenyl-5-pyrazolone, Lower part: acridine.) (1) initially; (2) after heating for 5hr at 50°.

Table 2

Yields of Products from Reaction of Acridine with Pyrazolone Derivatives after Ultrasonic Wave. Reaction Products are 4-(9-acridinyl)-3-methyl-1-phenyl-5-pyrazol-5-ol **1** and 4-(9-acridinyl)-3-methyl-1-*p*-tolyl-5-pyrazol-5-ol **4**.

Products	Reaction conditions		Yield (%)
	Temp.(°)	Time (hr)	
1	Room Temp	5	48.5
	50	5	59.6
4	Room Temp	5	0
	50	5	14.6

tion product was the keto-formed acridinium derivative in a low yield (Yield of 35%).

In this reaction system, it was found that the dihydroacridinyl derivatives were not produced. It is thought that most of the dihydroacridinyl derivatives were not able to form the reaction intermediate. Therefore, acridine does not draw hydrogen from pyrazolone derivatives, and acridine reacted with pyrazolone derivatives directly. It is supposed that C9-acridine substitution bodies are reaction products when obtained it.

Supersonic Reaction of Acridine with Pyrazolone Derivatives.

A mixture including acridine (2.80 mmol) and the respective pyrazolone derivative (5.60 mmol) was placed in a sealed glass reaction tube (inside diameter, 15 mm) under nitrogen gas. The reaction tube was then immersed in an ultrasonic bath, and subjected to ultrasonic wave for 5 hours at either room temperature (25°) or at 50°. The pyrazolone derivatives were the same as those used in the previous reaction. Only the substrates 3-methyl-1-phenyl-5-pyrazolone and 3-methyl-1-*p*-tolyl-5-pyrazolone successfully reacted to give the acridine derivatives **1** and **4**, respectively. The yields of **1** and **4** are increased through ultrasonication for 5 hours at room temperature (25°) and 50° as shown in Table 2. The yield of the product **1** was 48.5% at room temperature (25°), and 59.6% at 50°. The yield of **4** was 14.6% at 50°.

Effect of Recrystallization Solvents on the Reaction Product of Acridine with 3-Methyl-1-phenyl-5-pyrazolone.

After the mixture of acridine and 3-methyl-1-phenyl-5-pyrazolone was heated at 50° for 5 hours, the product was dissolved in methanol, acetone, benzene, chloroform, DMSO and THF, and then allowed to cool for recrystallization purposes. After each solvent was removed, the residual substance was taken up into diethylether, and was recrystallized again in the same solvent as before. Black-purple needle of product **1** were only obtained from recrystallization in methanol or chloroform. The yield of **1** from recrystallization in methanol was 35.5% (see Table 1), but in chloroform was only 17.3%. It is thought that product **1** deteriorated in the chloroform.

Solvent Reaction of Acridine with 3-Methyl-1-phenyl-5-pyrazolone.

To investigate the solvent reaction, acridine and 3-methyl-1-phenyl-5-pyrazolone were dissolved in several solvents. In a series of reactions, a mixture of acridine (0.125 g, 0.70 mmol) and 3-methyl-1-phenyl-5-pyrazolone (0.244 g, 1.40 mmol) were completely dissolved in 50 ml of solvent, and were reacted at room temperature (25°) for 5 hours. The solvents used were methanol, acetone, benzene, chloroform, DMSO and THF. After the reaction, the solvent was removed and the residual substance was taken up into diethylether. The material was then recrystallized in the same solvent as used in the reaction. Of the solvents tested, only methanol produced the black-purple needles of product **1** in 18.4% yield. For the reactions carried in other solvents, **1** was not obtained. Notably, the yield of **1** for the synthesis in methanol was less than for the non-solvent reaction (see Table 1).

Conclusion.

Acridine and pyrazolone derivatives were investigated in the solid-state reaction, in the supersonic reaction, and in various solvents. As a result of the investigations, it was shown that the reaction occurs successfully in the solid

state. Furthermore, it appears that the reaction of acridine and 3-methyl-1-phenyl-5-pyrazolone is a very specific reaction involving the enol form of 3-methyl-1-phenyl-5-pyrazolone. Reaction product was provided in high yield.

EXPERIMENTAL PROCEDURES.

Measurements.

Melting points were determined on a Yamato melting point apparatus model MP-21 and were not corrected. The ir spectra were taken on a Shimadzu FT-IR 4200 spectrophotometer. ^{1}H nmr and ^{13}C nmr spectra were measured with a Bruker FT-NMR AVANC-E500 spectrometer. The EI mass spectra were taken on a Shimadzu GC-MS-QP1000EX or Shimadzu GC-MS-QP5050A spectrometers. The elemental analyses were measured by YANOCO CHN CODER NT-5.

Solid State Reaction of Acridine with Pyrazolone Derivatives.

Reaction of acridine with different pyrazolone derivatives at different temperatures is shown in Table 1 for a molar ratio of acridine:pyrazolone derivative of 1:2. The used pyrazolone derivatives were 3-methyl-1-phenyl-5-pyrazolone, 3-methyl-5-pyrazolone, 1-(4'-nitrophenyl)-3-methyl-5-pyrazolone, 3-methyl-1-*p*-tolyl-5-pyrazolone, 1,3-dimethyl-5-pyrazolone and 2,3-dimethyl-1-phenyl-5-pyrazolone. The resulting products were thus, 4-(9-acridinyl)-3-methyl-1-phenyl-5-pyrazol-5-ol **1,** 4-(9-acridinyl)-3-methyl-5-pyrazol-5-ol **2**, 4-(9-acridinyl)-1-(4'-nitrophenyl)-3-methyl-5-pyrazol-5-ol **3**, 4-(9-acridinyl)-3-methyl-1-*p*-tolyl-5-pyrazol-5-ol **4**, 4-(9-acridinyl)-1,3-dimethyl-5-pyrazol-5-ol **5** and 4-(9-acridinyl)-2,3-dimethyl-1-phenyl-5-pyrazol-5-ol **6**, respectively.

4-(9-Acridinyl)-3-methyl-1-phenyl-5-pyrazol-5-ol (**1**).

A mixture of acridine (0.501 g, 2.80 mmol) and 3-methyl-1-phenyl 5-pyrazolone (0.974 g, 5.60 mmol) was placed in a sealed glass reaction tube (inside diameter, 15 mm) under nitrogen gas. After the reaction tube was heated for 5 hours at 50° or 100° or 150°, the resulting product was dissolved in methanol. Then the reaction mixture was washed with diethylether to remove unreacted starting materials. Steam distillation of the methanol fraction removed the methanol and the resulting residue was washed with methanol to yield the acridine derivative, 4-(9-acridinyl)-3-methyl-1-phenyl-5-pyrazol-5-ol **1**, mp 267-268° as black purple needles; ir (potassium bromide): 3450(ν O-H), 1580, 1510, 1450, 1350(ν N-H), 770, 695(δ C-H) cm^{-1}; ms: EI (m/Z) 285; ^{1}H nmr (deuteriomethanol CD_3OD): δ 2.02 (s, 3H), 7.32 (t, 1H, *J*=4.44 Hz), 7.87 (q, 2H, *J*=4.94 Hz), 7.51 (t, 2H, *J*=4.77 Hz), 7.65 (t, 2H, *J*=4.75 Hz), 7.87 (d, 2H, *J*=4.79 Hz), 7.87 (d, 2H, *J*=4.79 Hz), 8.19 (t, 4H, *J*=4.61 Hz).

Anal. Calcd. For $C_{23}H_{17}ON_3$: C, 78.61; H, 4.88; N, 11.96. Found: C, 78.94; H, 4.93; N, 12.05.

4-(9-Acridinyl)-3-methyl-5-pyrazol-5-ol (**2**).

A mixture of acridine (0.501 g, 2.80 mmol) and 3-methyl-5-pyrazolone (0.549 g, 5.60 mmol) was placed in a sealed glass reaction tube (inside diameter, 15 mm) under nitrogen gas. After the reaction tube was heated for 5 hours at 50° or 100° or 150°, the resulting product was dissolved in methanol. Then the reaction mixture was washed with diethylether to remove unreacted starting materials. Steam distillation of the methanol fraction removed the methanol and the resulting residue was washed with methanol to yield the acridine derivative, 4-(9-acridinyl)-3-methyl-5-pyrazol-5-ol **2**, mp 301-302° as yellow needles; ir (potassium bromide): 3460(ν O-H), 1590, 1500, 1420, 1315(ν N-H), 760(δ C-H) cm^{-1}; ms: EI (m/Z) 275; ^{1}H nmr (deuteriomethanol CD_3OD): δ 1.97 (s, 3H), 7.58, 7.63 (tt, 3H, *J*=4.58 Hz, *J*=4.54 Hz), 7.87 (q, 2H, *J*=4.94 Hz), 7.96 (d, 1H, *J*=5.04 Hz), 8.02 (quintet, 3H, *J*=4.15 Hz).

Anal. Calcd. For $C_{17}H_{13}ON_3$: C, 74.17; H, 4.76; N, 15.26. Found: C, 75.55; H, 4.76; N, 14.45.

4-(9-Acridinyl)-1-(4'-nitrophenyl)-3-methyl-5-pyrazol-5-ol (**3**).

A mixture of acridine (0.501 g, 2.80 mmol) and 1-(4'-nitrophenyl)-3-methyl-5-pyrazolone (1.226 g, 5.60 mmol) was placed in a sealed glass reaction tube (inside diameter, 15 mm) under nitrogen gas. After the reaction tube was heated for 5 hours at 50° or 100° or 150°, the resulting product was dissolved in methanol. Then the reaction mixture was washed with diethylether to remove unreacted starting materials. Steam distillation of the methanol fraction removed the methanol and the resulting residue was washed with methanol to yield the acridine derivative, 4-(9-acridinyl)-1-(4'-nitrophenyl)-3-methyl-5-pyrazol-5-ol **3**, mp 301-302° as brown needles; ir (potassium bromide): 3300(ν O-H), 1600, 1580(ν N-H), 1510, 1420(ν N-O), 820(δ C-H) $^{-1}$; ms: EI (m/Z) 396; ^{1}H nmr (deuteriomethanol CD_3OD): δ 1.99 (d, 3H, *J*=4.95 Hz), 6.61 (d, 2H, *J*=5.49 Hz), 7.64 (t, 2H, *J*=2.58 Hz), 7.98 (t, 2H, *J*=2.75 Hz), 8.15 (d, 2H, *J*=5.22 Hz), 8.34 (d, 2H, *J*=6.19 Hz).

Anal. Calcd. For $C_{23}H_{16}O_3N_4$: C, 68.69; H, 4.07; N, 14.13. Found: C, 68.02; H, 4.04; N, 15.08.

4-(9-Acridinyl)-3-methyl-1-*p*-tolyl-5-pyrazol-5-ol (**4**).

A mixture of acridine (0.501 g, 2.80 mmol) and 3-methyl-1-*p*-tolyl-5-pyrazolone (1.054 g, 5.60 mmol) was placed in a sealed glass reaction tube (inside diameter, 15 mm) under nitrogen gas. After the reaction tube was heated for 5 hours at 50° or 100° or 150°, the resulting product was dissolved in methanol. Then the reaction mixture was washed with diethylether to remove unreacted starting materials. Steam distillation of the methanol fraction removed the methanol and the resulting residue was washed with methanol to yield the acridine derivative, 4-(9-acridinyl)-3-methyl-1-*p*-tolyl-5-pyrazol-5-ol **4**, mp 268-269° as black violet needles; ir (potassium bromide): 3450(ν O-H), 2910, 2850(ν C-H), 1600, 1550, 1510(ν N-H), 1300, 820, 720(δ C-H) cm^{-1}; ms: EI (m/Z) 365; ^{1}H nmr (deuteriomethanol CD_3OD): δ 2.02 (s, 3H), 2.41 (s, 3H), 7.65 (t, 2H, *J*=4.98 Hz), 7.71 (d, 2H, *J*=5.01 Hz), 7.94 (t, 2H, *J*=4.58 Hz), 8.178(t, 4H, *J*=6.23 Hz).

Anal. Calcd. For $C_{24}H_{19}ON_3$: C, 78.88; H, 5.24; N, 11.50. Found: C, 78.16; H, 5.39; N, 11.31.

4-(9-Acridinyl)-1,3-dimethyl-5-pyrazol-5-ol (**5**).

A mixture of acridine (0.501 g, 2.80 mmol) and 1,3-methyl-5-pyrazolone (0.627 g, 5.60 mmol) was placed in a sealed glass reaction tube (inside diameter, 15 mm) under nitrogen gas. After the reaction tube was heated for 5 hours at 50° or 100° or 150°, the resulting product was dissolved in methanol. Then the reaction mixture was washed with diethylether to remove unreacted starting materials. Steam distillation of the methanol fraction removed the methanol and the resulting residue was washed with methanol to yield the acridine derivative, 4-(9-acridinyl)- 1,3-

methyl-5-pyrazol-5-ol **5**, mp 300-302° as red brown needles; ir (potassium bromide): 3450(ν O-H), 2910(ν C-H), 1620, 1560, 1480(ν N-H), 760(δ C-H) cm^{-1}; ms: EI (m/Z) 289; ^1H nmr (deuteriomethanol CD_3OD): δ 1.94 (s, 3H), 3.67 (s, 3H), 7.60 (t, 2H, *J*=4.59 Hz), 7.89 (t, 2H, *J*=4.61 Hz), 8.00 (d, 2H, *J*=5.1Hz), 8.19 (d, 2H, *J*=5.25Hz).

Anal. Calcd. For $C_{18}H_{15}ON_3$: C, 74.72; H, 5.23; N, 14.52. Found: C, 74.93; H, 5.32; N, 14.52.

4-(9-Acridinyl)-2,3-dimethyl-1-phenyl-5-pyrazol-5-ol (**6**).

A mixture of acridine (0.501 g, 2.80 mmol) and 2,3-methyl-1-phenyl-5-pyrazolone (1.054 g, 5.60 mmol) was placed in a sealed glass reaction tube (inside diameter, 15 mm) under nitrogen gas. After the reaction tube was heated for 5 hours at 50° or 100° or 150°, the resulting product was dissolved in methanol. Then the reaction mixture was washed with diethylether to remove raw materials of non-reaction. Steam distillation of the methanol fraction removed the methanol and the resulting residue was washed with methanol to yield the acridine derivative, 4-(9-acridinyl)-2,3-methyl-1-phenyl-5-pyrazol-5-ol **6**, mp 238-239° as orange yellow needles; ir (potassium bromide): 1660(ν C-O), 1580, 1500, 1440(ν N-H), 740(δ C-H) cm^{-1}; ms: EI (m/Z) 365; ^1H nmr (deuteriomethanol CD_3OD): δ 2.02 (s, 3H), 3.34 (s, 3H), 7.36 (t, 1H, *J*=7.43 Hz), 7.55 (quintet, 6H, *J*=7.58 Hz), 7.77 (t, 2H, *J*=7.65 Hz), 7.99 (t, 2H, *J*=8.85 Hz), 8.28 (t, 2H, *J*=8.7 Hz).

Anal. Calcd. For $C_{24}H_{19}ON_3$: C, 78.86; H, 5.24; N, 11.50. Found: C, 78.86; H, 5.07; N, 11.43.

Supersonic Reaction of Acridine with Pyrazolone Derivatives.

A mixture including acridine (2.80 mmol) and the respective pyrazolone derivative (5.60 mmol) was placed in a sealed glass reaction tube (inside diameter, 15 mm) under nitrogen gas. The reaction tube was then immersed in an ultrasonic bath, and subjected to ultrasonic wave for 5 hours at either room temperature (25°) or at 50°.

The pyrazolone derivatives were the same as those used in the previous reaction. The Pyrazolone derivatives were 3-methyl-1-phenyl-5-pyrazolone, 3-methyl-5-pyrazolone, 1-(4'-nitrophenyl)-3-methyl-5-pyrazolone, 3-methyl-1-*p*-tolyl-5-pyrazolone, 1,3-dimethyl-5-pyrazolone and 2,3-dimethyl-1-phenyl-5-pyrazolone.

The crude product for these samples was dissolved in methanol and washed with diethylether to unreacted starting materials. Steam distillation of the methanol fraction removed the methanol and the resulting residue was washed with methanol to give the acridine derivatives **1** and **4**. The yield of the product **1** was 48.5% at room temperature (25°), and 59.6% at 50°. The yield of **4** was 14.6% at 50°.

Effect of Recrystallizaion Solvent on the Reaction Product of Acridine with 3-Methyl-1-phenyl-5-pyrazolone.

After the reaction mixture of acridine and 3-methyl-1-phenyl-5-pyrazolone was heated at 50° for 5 hours, the product was dissolved in methanol, acetone, benzene, chloroform, DMSO or THF. And these solvent were used in a recrystallization process. After each solvent was removed, the residual substance was taken up into diethylether. Again, the product was recrystallized in the same solvent as before. Black-purple needles of **1** (mp 267-268°) were obtained from recrystallization in methanol or chloroform. The recrystallization yield of **1** from methanol was 35.5%, but the yield of the product **1** from chloroform was only 17.3%.

Solvent Reaction of Acridine with 3-Methyl-1-phenyl-5-pyrazolone.

A mixture of acridine (0.125 g, 0.70 mmol) and 3-methyl-1-phenyl-5-pyrazolone (0.244 g, 1.40 mmol) was completely dissolved in 50 ml of solvent, and was reacted at room temperature (25°) for 5 hours. The solvents used were methanol, acetone, benzene, chloroform, DMSO and THF. After the reaction, solvent was removed and the residual substance was taken up into diethylether. The product material was then recrystallized in the same solvent as used in the reaction. In the case of methanol, black-purple needle of product **1** (mp 267-268°) were obtained at 18.4%. For the reactions in other solvents, **1** was not obtained.

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