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A NEW EFFICIENT SYNTHESIS OF IMIDAZOLINONES AND QUINAZOLINONE BY INTRAMOLECULAR AZA-WITTIG REACTION

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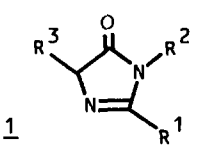
A new synthesis of imidazolinones and quinazolinones by intramolecular aza-Wittig reaction is described. Readily available azido substituted imides 4, 6, 10, and 12 reacted with triphenylphosphine or tributylphosphine to afford the corresponding imidazolinones 1, 7 and quinazolinones 2, 12 via the Staudinger reaction, followed by an intramolecular aza-Wittig reaction.

Recently we have demonstrated that ester carbonyls are reactive in the intramolecular aza-Wittig reaction[1], and the new synthesis of oxazoles under mild conditions has been developed utilizing this type of intramolecular imination. In comparison with aldehydes and ketones, ester-, amide-, and imide-carbonyls are generally unreactive in the intermolecular version. Now, we have applied intramolecular aza-Wittig reaction to the imide carbonyl group[2] because it may provide a new convenient route to two-nitrogen containing heterocycles, such as (4H)-imidazolin-5-one 1 and (3H)-quinazolin-4-one 2, that are often involved in some alkaloids[3] and drugs[3d, 4].



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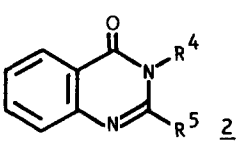
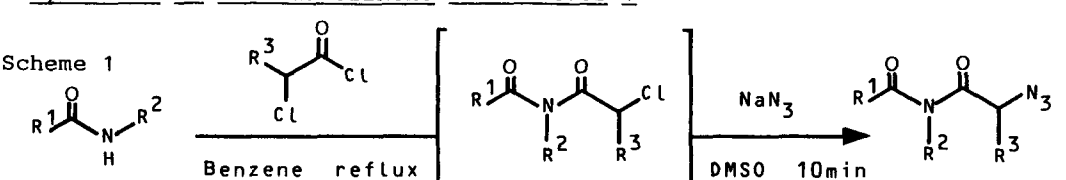


2

RESULTS AND DISCUSSION

Syntheses of 5-Imidazolinone Derivatives 1

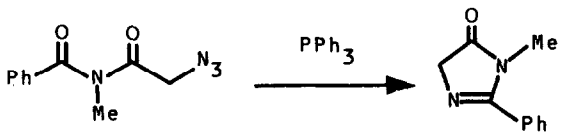
Scheme 1



3 3' 4

The starting azido derivatives 4 were readily prepared from amide derivatives 3 in one pot (Scheme 1): Amides 3 were treated with α-chloroacid chloride to give the corresponding imides 3', which, without purification, were further treated with sodium azide to afford the azides 4 (Table I). Taking our previous studies[1, 2] into consideration, the reaction of 4a with triphenylphosphine was examined first in toluene under reflux and the corresponding imidazolinone 1a was obtained in 82% yield (Scheme 2).

Scheme 2

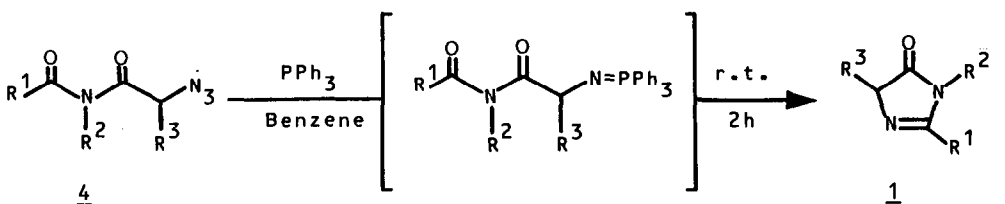


4a 1a

Temp.	Yield(%)
110°C	82
r.t.	89

But this cyclization proceeded even at room temperature in 89% yield. The present system 4a was very reactive in the aza-Wittig reaction in comparison with N-(azidoalkyl)phthalimides, whose cyclization with triphenylphosphine required heating at 120-140 °C[2]. This may be due to aromatic character of the cyclization product, 1. The other azides 4b-f were similarly treated with triphenylphosphine to afford imidazolinones 1b-f in good yields (Scheme 3 and Table I). In the case of 4g, imidazolinone 1g was obtained in only 16% yield because of the steric hindrance of phenyl group in the aza-Wittig reaction. Therefore, tributylphosphine was used instead of triphenylphosphine to improve the yield. Then, 1g was obtained in 70% yield.

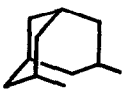
Scheme 3



4 1

Furthermore, we applied this method to the synthesis of another type of bicyclic imidazolinone 7 (Scheme 4). Azide 6 was readily obtained by bromination of 5, followed by treatment with sodium azide. The cyclization of 6 to 7 via the Staudinger reaction, followed by aza-Wittig reaction proceeded rather sluggishly at room temperature and heating to 100°C gave 7 in 75% yield. By using tributylphosphine, 7 was obtained in 69% yield even at room temperature.

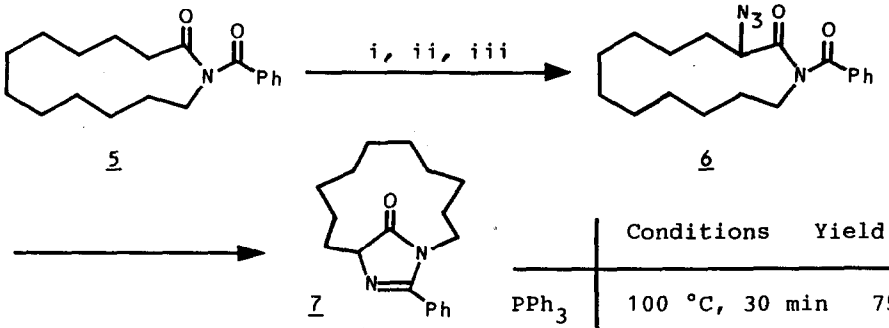
Table I

	R^1	R^2	R^3	Yields(%)[a] 4	1
a	Ph	Me	H	78	89
b	Me	Ph	H	74	91
c	Ph	Ph	H	46	85
d		Me	H	49	99
e	$-(CH_2)_3-$		H	59	80
f			H	52	91
g	$-(CH_2)_3-$		Ph	85	16(70)[b]

a) Isolated yields.

b) Treatment with tributylphosphine. See experimental.

Scheme 4



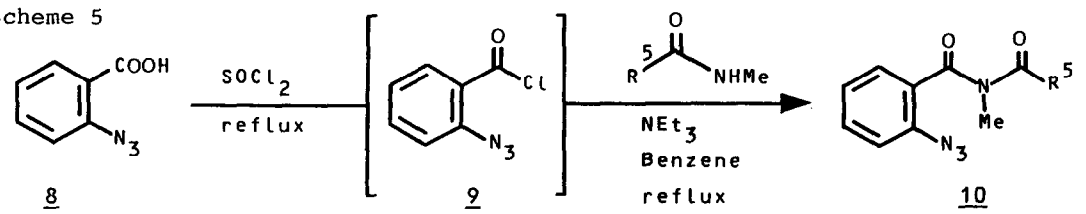
	Conditions	Yield(%)
PPh_3	100 °C, 30 min	75
PBu_3	r.t., 30 min	69

i) LDA/THF, -78°C, ii) Br_2, iii) NaN_3/DMSO.

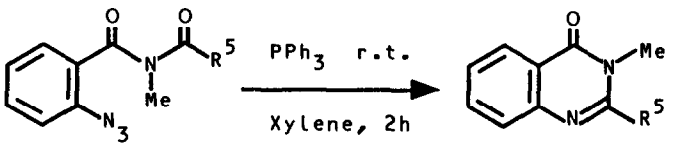
Syntheses of 4-Quinazolinone Derivatives 2

The intramolecular aza-Wittig reaction provided a convenient route to imidazolinones 1 in mild conditions, as described above. We extended this method to the synthesis of 6-membered quinazolinone 2. o-Azidobenzoic acid 8 was converted to the corresponding acid chloride 9[5], and required azides 10 were readily obtained from 9 and appropriate amides (Scheme 5 and Table II).

Scheme 5



Scheme 6



10 2

Table II

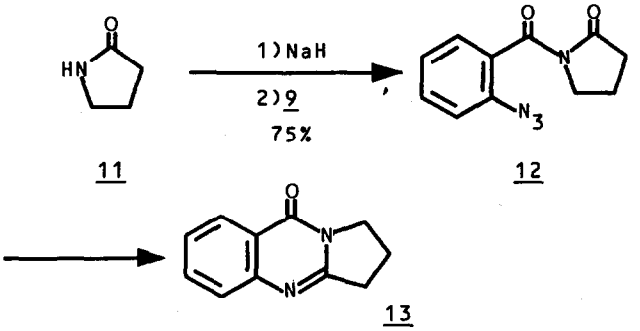
	R^5	Yields(%)[a] 10	2
a	Me	44	99
b		84	99
c		70	99
d		51	95

a) Isolated yields

The azides 10 were treated with triphenylphosphine in the same manner for the synthesis of imidazolinones (Scheme 6). As expected, the cyclization proceeded smoothly at room temperature. Although 2b-d have the substituent labile to acid and/or heating, they were obtained in high yield by this method.

Furthermore, we attempted to synthesize a natural product, deoxyvacisinone 13[3a] as an application (Scheme 7).

Scheme 7



11 12 13

	Conditions	Yields(%)
PPh_3	140°C, 5h	99.5
PBu_3	r.t., 3h	99

Azide 12 was obtained from 9 and pyrrolidone 11 in the presence of sodium hydride as a base at room temperature. Then azide 12 was treated with triphenylphosphine but the cyclization required heating at 140°C for 5h. As the reacting carbonyl group is located in the five membered ring, it was expected to require a strained transition state in the four center reaction[6]. When tributylphosphine was used instead of triphenylphosphine, 13 could be obtained in 99% yield even at room temperature for 2h.

There are many known methods for the syntheses of imidazolinones[7] and quinazolinones[3d, 8]. However, a strong acid as the dehydrating agent and/or heating are required generally in these known syntheses. By using

the intramolecular aza-Wittig reaction as the key-step, the cyclization to these nitrogen heterocycles proceeds in high yields under milder conditions. Clearly, the intramolecular aza-Wittig method holds promise as a general synthetic route to 5- and 6-membered nitrogen heterocycles.

EXPERIMENTAL

General Melting points were determined with a Yanagimoto micro melting point apparatus and were uncorrected. Elemental analyses were performed with a Perkin-Elmer 240 B elemental analyzer. ^{1}H NMR spectra were taken at 25°C with a Varian Gemini 200 instrument at 200 MHz (Me_4Si as internal standard). IR spectra were obtained on a JASCO IRA-1 spectrometer. Column chromatography was performed on Fuji-Davison silica gel BW-300.

Preparations of α-Azido Substituted Imides 4

To a stirred solution of amide 3 (1.00 mmol) in benzene (5.0 ml) was added 2-chloroacetyl chloride or 2-chloro-2-phenylacetyl chloride (1.10 mmol) at room temperature under nitrogen. The mixture was heated to reflux for 2 h, and then, evaporated under reduced pressure. Dimethylsulfoxide (DMSO) (5.0 ml) was added to the residue to give a solution, to which was added NaN_3 (3.00 mmol) in one portion. After stirring for 10 min at room temperature, the mixture was poured onto water and extracted with ether (3 x 30 ml). The combined extracts were dried (Na_2SO_4) and evaporated under reduced pressure, and the residue was chromatographed on a silica gel column, eluting with dichloromethane-hexane system (ethyl acetate-hexane system for 4g), to afford 4. The yields, physical and analytical data were summarized in Tables I, III and IV, respectively.

Synthesis of Imidazolinones 1 by Intramolecular Aza-Wittig Reaction of 4

To a solution of imide derivative 4 (1.00 mmol) in benzene (10 ml) was added triphenylphosphine or tributylphosphine (1.10 mmol). The mixture was stirred for 2 h at room temperature and evaporated under reduced pressure. The residue was chromatographed on a silica gel column, eluting with chloroform-ethyl acetate system (ethyl acetate-methanol or ethanol system for 1e-g), to afford imidazolinones 1. The yields, physical and analytical data were summarized in Tables I, V and VI.

3-Azido-1-benzoylazacyclotridecan-2-one 6

To tetrahydrofuran (THF) (10 ml) was added a 1.5 M solution of lithium diisopropylamide in cyclohexane (1.15 ml, 1.73 mmol) at -78°C, followed by adding dropwise a solution of 1-benzoylazacyclotridecan-2-one 5 (500 mg, 1.66

mmol) in THF (2.0 ml) at -78 °C. After the mixture was stirred for 1 h, bromine, dried over P_2O_5, (256 mg, 1.66 mmol) was added dropwise, and after 10 min, the mixture was slowly warmed to room temperature while the stirring was continued. After 10 min, the mixture was poured onto water and extracted with ether (5 x 50 ml). The combined extracts were dried (Na_2SO_4) and evaporated under reduced pressure. The obtained residue was chromatographed on a silica gel column, eluting with ethyl acetate-hexane (1:20), to afford 354 mg (56.1 %) of 3-bromo-1-benzoylazacyclotridecan-2-one as colorless oil: δ ($CDCl_3$) 7.71-7.37(m, 5H), 4.88(dd, 1H, J=9.4 and 5.0Hz), 4.23(ddd, 1H, J=14.0, 6.0 and 4.8Hz), 3.51(ddd, 1H, J=14.0, 8.2 and 4.6Hz), 2.33-1.63(m, 4H), and 1.60-1.15(m, 14H); $\nu_{max.}$ (CCl_4) 2970, 2870, 1690, 1670, 1450, 910, and 720 cm^{-1} (Found: C, 59.72; H, 6.82; N, 3.94. $C_{19}H_{26}BrNO_2$ requires C, 60.00; H, 6.89; N, 3.68%). Then to a solution of sodium azide (150 mg, 2.31 mmol) in DMSO (5.0 ml) was added 3-bromo-1-benzoylazacyclotridecan-2-one (300 mg, 0.79 mmol) at room temperature. After stirring for 10 min, the mixture was poured onto ice-water and extracted with ether (5 x 50 ml). The combined extracts were dried (Na_2SO_4) and evaporated under reduced pressure. The obtained residue was chromatographed on a silica gel column, eluting with ethyl acetate-hexane (1:4), to afford 234 mg (90.0 %) of 6 as pale yellow crystals: m.p. 63-66 °C; δ ($CDCl_3$) 7.67-7.45(m, 5H), 4.27(ddd, 1H, J=13.6, 4.4 and 3.4Hz), 3.88(dd, 1H, J=7.2 and 6.6Hz), 3.55(ddd, 1H, J=13.6, 10.2 and 2.6Hz), 2.05-1.61(m, 4H), 1.50-1.15(m, 14H); $\nu_{max.}$ (CCl_4) 2950, 2870, 2100, 1690, 1670, and 1450 cm^{-1} (Found: C, 66.69; H, 7.50; N, 16.12. $C_{19}H_{26}N_4O_2$ requires C, 66.64; H, 7.65; N, 16.36%).

14-Phenyl-1,13-diazabicyclo[10.2.1]pentadec-13-en-15-one 7.

Cyclization with triphenylphosphine. To a solution of 6 (100 mg, 0.29 mmol) in xylene (b.p. 138.5-141.5°C, 5.0 ml) was added triphenylphosphine (80 mg, 0.31 mmol) under nitrogen and stirred at room temperature for 1 h. The mixture was heated at 100 °C for 30 min with continued stirring. The cooled mixture was evaporated under reduced pressure and the residue was chromatographed on a silica gel column, eluting with ethyl acetate-hexane (1:1 v/v), to afford 65 mg (75 %) of imidazolinone 7 as pale yellow solid: m.p. 78-81 °C; δ ($CDCl_3$) 7.61-7.38(m, 5H), 4.28(dd, 1H, J=5.6 and 3.8Hz, C_{12}-H), 3.93(ddd, 1H, J=14.2, 5.9 and 3.0 Hz, C_2-H), 3.46(ddd, 1H, J=14.2, 9.5 and 1.5Hz, C_2-H), 2.45-2.29(m, 1H, C_{11}-H), 2.17-2.03(m, 1H, C_{11}-H), 1.89-1.58(m, 2H, C_3-H), 1.50-0.80(m, 14H); $\nu_{max.}$ (CCl_4) 2950, 2870, 1735, 1625, 1600, 1500, 1450, 910, and 695 cm^{-1} (Found: C, 76.21; H, 8.49; N, 9.31. $C_{19}H_{26}N_2O$ requires C, 76.47; H, 8.78; N, 9.39%).

Cyclization with tributylphosphine. To a solution of 6 (100 mg, 0.29 mmol)

in xylene (5.0 ml) was added tributylphosphine (65 mg, 0.32 mmol) under nitrogen and stirred at room temperature for 30 min. The mixture was evaporated under reduced pressure and the residue was treated in the same manner described above, to afford 60 mg (69 %) of 7.

Preparation of Azido Derivatives 10.

o-Azidobenzoic acid 8 (1.00 mmol) was placed into a two necked flask. Thionyl chloride (0.80 ml, 1.33 g, 11.2 mmol) was added into the flask, followed by heating at 80 °C under nitrogen for 2 h. The cooled mixture was evaporated under reduced pressure and the residue was dissolved in dry benzene (5.0 ml). To this solution was added the corresponding amide (1.00 mmol) and triethylamine (1.00 mmol) under nitrogen, and the mixture was heated at 80 °C for 2 h. The cooled mixture was filtered, and the filtrate was evaporated under reduced pressure. The residue was chromatographed on a silica gel column, eluting with ethyl acetate-hexane (1:4), to afford 10. The yields, physical and analytical data were summarized in Tables II, VII and IX.

Synthesis of Quinazolinones 2 by Intramolecular Aza-Wittig Reaction of 10.

To a solution of imide derivative 10 (1.00 mmol) in xylene (5.0 ml) was added triphenylphosphine (1.00 mmol) under nitrogen and stirred at room temperature for 2 h. The mixture was evaporated under reduced pressure and the residue was chromatographed on a silica gel column, eluting with ethyl acetate-hexane system, to afford quinazolinone 2. The yields, physical and analytical data were summarized in Tables II, VIII and X.

1-(2-Azidobenzoyl)azacyclopent-2-one 12.

To a solution of pyrrolidone 11 (160 mg, 1.88 mmol) in dry THF (5.0 ml) was added sodium hydride (60 % dispersion in mineral oil, 80 mg, 2.0 mmol) at 0 °C under nitrogen and stirred for 15 min. To this mixture was added dropwise the solution of 9, which was obtained from 8 (300 mg, 1.84 mmol) as described above, in dry THF (3.0 ml) at 0 °C, and the mixture was stirred at room temperature for 2 h. The mixture was poured onto ice-water and extracted with dichloromethane (6 x 50 ml). The combined extracts were dried (Na_2SO_4) and evaporated under reduced pressure, and the residue was chromatographed on a silica gel column, eluting with ethyl acetate-hexane (1:2), to afford 316 mg (74.6 %) of 12 as pale yellow solid; m.p. 82-84 °C; δ ($CDCl_3$) 7.53-7.16(m, 4H), 3.99(t, 2H, J=7.2Hz), 2.60(t, 2H, J=7.8Hz), 2.15(tt, 2H, J=7.2 and 7.8Hz); $\nu_{max.}$ (CCl_4) 2150, 1760, 1680, 1610, and 1500 cm^{-1} (Found: C, 57.21; H, 4.41; N, 24.33. $C_{11}H_{10}N_4O_2$ requires C, 57.39; H,

4.38; N, 24.34%).

Deoxyvacisinone 13.

Cyclization with triphenylphosphine. To a solution of 12 (200 mg, 0.87 mmol) in xylene (5.0 ml) was added triphenylphosphine (230 mg, 0.88 mmol) and the mixture was stirred for 1 h at room temperature. The mixture was heated at 140 °C for 5 h with continued stirring. The cooled mixture was evaporated under reduced pressure, and the residue was chromatographed on a silica gel column, eluting with ethyl acetate-hexane (2:1), to afford 161 mg (99.5 %) of 13 as white solid; m.p. 106-108 °C; δ ($CDCl_3$) 8.30(dd, 1H, J=8.2 and 2.0Hz), 7.79-7.42(m, 3H), 4.22(t, 2H, J=7.4Hz), 3.20(t, 2H, J=8.0Hz), 2.30(tt, 2H, J=7.4 and 8.0Hz); $\nu_{max.}$ (CCl_4) 1690, 1640, 1620, and 1480 cm^{-1} (Found: C, 70.69; H, 5.12; N, 14.76. $C_{11}H_{10}N_2O$ requires C, 70.95; H, 5.41; N, 15.04%).

Cyclization with tributylphosphine. To a solution of 12 (200 mg, 0.87 mmol) in xylene (5.0 ml) was added tributylphosphine (0.22 ml, 179 mg, 0.88 mmol). The mixture was stirred for 1 h at room temperature, and heated at 50 °C for 3 h with continued stirring. The mixture was treated in the same manner as above to afford 160 mg (98.9 %) of 13.

Table III Physical Data of α-Azidoimides 4

4	IR, cm^{-1} (CH_2Cl_2)	^1H NMR, δ ($CDCl_3$)
a	2120, 1710, 1700	7.64-7.46(m, 5H), 4.40(s, 2H), 3.24(s, 3H)
b	2100, 1720, 1710	7.60-7.15(m, 5H), 4.35(s, 2H), 2.14(s, 3H)
c	2110, 1720, 1695	7.56-7.51(m, 2H), 7.44-7.24(m, 6H), 7.16-7.11(m, 2H), 4.45(s, 2H)
d	2120, 1710, 1680	7.66(dd, 1H, J=0.8 and 1.9Hz), 7.31(dd, 1H, J=3.6 and 0.8Hz), 6.62(dd, 1H, J=1.9 and 3.6Hz), 4.36(s, 2H), 3.45(s, 3H)
e	2120, 1750, 1710	4.74(s, 2H), 3.87(t, 2H, J=7.0Hz), 2.62(t, 2H, J=8.0Hz), 2.13(tt, 2H, J=7.0 and 8.0Hz)
f	2120, 1690	5.14-5.11(m, 1H), 4.41(s, 2H), 3.03-2.95(m, 1H), 2.15-1.58(m, 12H)
g	2110, 1740, 1700	7.45-7.37(m, 5H), 6.08(s, 1H), 4.01-3.74(m, 2H), 2.68-2.38(m, 2H), 2.19-1.90(m,2H)

Table IV Analytical Data of α-Azidoimides 4

4	m.p. (°C)	Molecular Formula	Analysis(%) Found C, H, N (Calcd. C, H, N)	4	m.p. (°C)	Molecular Formula	Analysis(%) Found C ,H ,N (Calcd. C ,H ,N)
a	69-72	$C_{10}H_{10}N_4O_2$	54.78 4.70 25.47 (55.04 4.62 25.68)	e	oil	$C_6H_8N_4O_2$	42.90 4.71 33.37 (42.86 4.80 33.32)
b	44-47	$C_{10}H_{10}N_4O_2$	54.76 4.67 25.72 (55.04 4.62 25.68)	f	61-64	$C_{12}H_{16}N_4O_2$	58.45 6.49 22.18 (58.05 6.50 22.57)
c	51-53	$C_{15}H_{12}N_4O_2$	64.21 4.28 19.86 (64.28 4.32 19.99)	g	49-52	$C_{12}H_{12}N_4O_2$	58.83 4.83 23.24 (59.01 4.95 22.94)
d	48-50	$C_8H_8N_4O_3$	46.19 3.94 26.66 (46.16 3.87 26.91)				

Table V Physical Data of 5-Imidazolinones 1

1	IR, cm^{-1} (CH_2Cl_2)	^1H NMR, δ ($CDCl_3$)
a	1730, 1620, 1330	7.66-7.50(m, 5H), 4.30(s, 2H), 3.16(s, 3H)
b	1750, 1600, 1330	7.56-7.40(m, 3H), 7.23-7.18(m, 2H), 4.27(q, 2H, J=2.2Hz), 2.09(t, 3H, J=2.2Hz)
c	1750, 1620, 1600, 1330	7.44-7.07(m, 10H), 4.51(s, 2H)
d	1740, 1640, 1620, 1330	7.64(d, 1H, J=1.6Hz), 7.17(d, 1H, J=3.6Hz), 6.60(dd, 1H, J=1.6 and 3.6Hz), 4.30(s, 2H), 3.39,(s, 3H)
e	1720, 1660, 1350	4.41(s, 2H), 3.56(t, 2H, J=7.0Hz), 2.70-2.60(m, 2H), 2.52-2.37(m, 2H)
f	1720, 1640, 1350	4.43-4.38(m, 1H), 4.13(s, 2H), 3.05(t, 1H, J=5.8Hz), 2.15-1.98(m, 6H), 1.87-1.61(m, 6H)
g	1720, 1680, 1600, 1340	7.70-7.18(m, 5H), 5.48(s, 1H), 3.43(t, 2H, J=7.0Hz), 2.69-2.58(m, 2H), 2.42-2.27(m, 2H)

Table VI Analytical Data of 5-Imidazolinones 1

1	m.p. (°C)	Molecular Formula	Analysis(%) Found C, H, N (Calcd. C, H, N)	1	m.p. (°C)	Molecular Formula	Analysis(%) Found C, H, N (Calcd. C, H, N)
a	90-93	$C_{10}H_{10}N_2O$	69.16 5.81 15.86 (68.95 5.79 16.08)	d	90(dec)	$C_8H_8N_2O_2$	58.25 5.01 16.80 (58.53 4.91 17.06)
b	93-96	$C_{10}H_{10}N_2O$	68.75 5.80 15.84 (68.95 5.79 16.08)	e	61-64	$C_6H_8N_2O$ +MeOH(0.26)	56.49 6.49 20.77 (56.76 6.88 21.15)
c	116-119	$C_{15}H_{12}N_2O$	76.37 5.22 11.59 (76.25 5.20 11.86)	f	121-124	$C_{12}H_{16}N_2O$	70.67 7.69 13.46 (70.56 7.90 13.71)

continued from preceding page.

1	m.p. (°C)	Molecular Formula	Found C, H, N (Calcd. C, H, N)
g	105-107	$C_{12}H_{12}N_2O$	71.75 6.00 13.93 (71.98 6.04 13.99)

Table VII Physical Data of Imides 10

10	IR, cm^{-1} (CH_2Cl_2)	^1H NMR, δ ($CDCl_3$)
a	2150, 1710, 1700	7.56-7.20(m, 4H), 3.12(s, 3H), 2.47(s, 3H)
b	2140, 1700, 1660, 1600	7.54-7.19(m, 4H), 3.30(s, 3H), 2.05-1.91(m, 1H), 1.12-1.04(m, 2H), 0.83-0.74(m, 2H)
c	2140, 1680, 1650, 1620, 1600	7.64(d, 1H, J=15.6Hz), 7.52-7.15(m, 9H), 6.82(d, 1H, J=15.6Hz), 3.34(s, 3H)
d	2140, 1690, 1660, 1640, 1600	7.53-7.16(m, 5H), 6.22-5.96(m, 3H), 3.27(s, 3H), 1.84(d, 3H, J=5.6Hz)

Table VIII Physical Data of 4-Quinazolinones 2

2	IR, cm^{-1} (CH_2Cl_2)	^1H NMR, δ ($CDCl_3$)
a	1670, 1600, 1480	8.27(dd, 1H, J=1.0, and 8.0Hz), 7.72-7.27(m, 3H), 3.64(s, 3H), 2.63(s, 3H)
b	1680, 1620, 1600, 1480, 1420	8.25(dd, 1H, J=1.2 and 8.0Hz), 7.73-7.40(m, 3H), 3.80(s, 3H), 2.11-1.98(m, 1H), 1.32-1.05(m, 4H)
c	1670, 1580, 1560, 1480	8.30(dd, 1H, J=2.2 and 8.2Hz), 8.02(d, 1H, J=15.4Hz), 7.60-7.41(m, 8H), 7.14(d, 1H, J=15.4Hz), 3.78(s, 3H)
d	1690, 1630, 1560, 1480	8.28-8.23(m, 1H), 7.76-7.37(m, 4H), 6.62-6.13(m, 3H), 3.68(s, 3H), 1.91(d, 3H, J=5.8Hz)

Table IX Analytical Data of Imides 10

10	m.p. (°C)	Molecular Formula	Analysis(%) Found C, H, N (Calcd. C, H, N)
a	<30	$C_{10}H_{10}N_4O_2$	54.84 4.57 25.40 (55.04 4.62 25.68)
b	54.5-55.5	$C_{12}H_{12}N_4O_2$	58.73 4.97 22.92 (59.01 4.95 22.94)
c	71-74	$C_{17}H_{14}N_4O_2$	66.35 4.79 18.03 (66.66 4.61 18.29)
d	54-59	$C_{14}H_{14}N_4O_2$	62.04 5.33 20.79 (62.21 5.22 20.73)

Table X Analytical Data of 4-Quinazolinones 2

2	m.p. (°C)	Molecular Formula	Found C, H, N (Calcd. C, H, N)	2	m.p. (°C)	Molecular Formula	Found C, H, N (Calcd. C, H, N)
a	104 -107	$C_{10}H_{10}N_2O$	69.22 5.80 15.97 (68.95 5.79 16.08)	c	171	$C_{17}H_{14}N_2O$	77.73 5.23 10.30 (77.84 5.38 10.68)
b	77-80	$C_{12}H_{12}N_2O$	71.76 5.80 13.72 (71.98 6.04 13.99)	d	140 -143	$C_{14}H_{14}N_2O$	74.36 5.97 12.49 (74.31 6.24 12.38)

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