(HPLC samples were quenched with ethyl bromide as above). The workup involved pouring the mixtures into an excess of water and collecting the solid products by filtration. The products were washed several times with water and then dried in a vacuum oven. Purification was effected by recrystallization or by liquid chromatography. The purity of the products was confirmed by HPLC, by ^{13}C NMR, and by elemental microanalysis (Galbraith Laboratories). Further characterization was obtained by field desorption mass spectrometry. Details of the purification, the percent isolated yields, and the melting points for the derivatives ethyl 4-cyanophenyl sulfide (**8**), ethyl 4-nitrophenyl sulfide (**14**), ethyl 2,4-dinitrophenyl sulfide (**15**),[10] bis[(4-cyanophenyl)thio]-methane (**10**), and 4-cyanophenyl 4-nitrophenyl sulfide (**9**) are given in Table I. The reactions of sodium 4-cyanophenyl sulfide (**7**) with *p*-dichlorobenzene or chlorobenzene was also examined, but it was found that even at reflux less than 10% of the sodium aryl sulfide underwent reaction with these aryl halides.

Thiophenoxide Exchange Reactions. A sample of 4-chlorobenzonitrile (1.00 g, 0.007 27 mol) was combined with sodium sulfide (0.57 g, 0.007 31 mol) and biphenyl (0.14 g, 0.000 908 mol) in dry DMF (20 mL). The reaction was allowed to proceed for 18 h at 130 °C. The mixture was sampled and examined by HPLC (ethyl bromide quench). Only sodium 4-cyanophenyl sulfide (**7**) was present. Then, bis(4-nitrophenyl) sulfide (**1**; 0.75 g, 0.002 71 mol) was added. Compound **1** was completely consumed within 1 h at 130 °C. It was replaced by the mixed thioether 4-cyanophenyl 4-nitrophenyl sulfide (**9**, 90% yield after 1 h). After 12 h compound **9** had also been consumed and replaced with bis-(4-cyanophenyl) sulfide (**3**; 92% yield based on the quantity of mixed thioether originally present).

The same procedure involving the initial formation of sodium 4-nitrophenyl sulfide (**12**) followed by addition of bis(4-cyanophenyl) sulfide (**3**) resulted in a small amount of exchange (less than 10% of compound **3** consumed over the course of 12 h).

(10) The synthesis of compound **11** from the interaction of sodium 4-cyanophenyl sulfide (**7**) with 2,4-dinitrochlorobenzene resulted in a product that contained the impurity bis[(4-cyanophenyl)thio]nitrobenzene as indicated by mass spectrometry. The impurity was less than 3% of the isolated product because no product other than compound **II** was indicated by HPLC. The structure of the bis-substituted impurity was not determined.

Acknowledgment. We thank M. M. Grade for the synthesis of several compounds used in this study and for the carrying out of some preliminary rate studies involving the synthesis of diaryl sulfides.

Registry No. **1**, 1223-31-0; **2**, 2253-67-0; **3**, 46836-99-1; **4**, 86047-00-9; **7**, 61628-44-2; **8**, 86047-01-0; **9**, 21969-10-8; **10**, 86047-02-1; **12**, 13113-79-6; **13**, 51256-42-9; **14**, 7205-60-9; **15**, 7343-55-7; C_2H_5Br, 74-96-4; CH_2Br_2, 74-95-3; Na_2S, 1313-82-2; 4-chlorobenzonitrile, 623-03-0; 1-chloro-4-nitrobenzene, 100-00-5; 2,4-dinitrobenzene, 97-00-7; 2-chlorobenzonitrile, 873-32-5.

Halogenated Ketenes. 38. Cycloaddition of α,β-Unsaturated Imines with Ketenes To Yield Both β- and δ-Lactams

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The cycloaddition of various types of α,β-unsaturated imines with diphenyl- and dichloroketenes yields both (2 + 2) and (4 + 2) cycloaddition products, i.e., β-lactams and δ-lactams, respectively. The cycloaddition products are dependent upon substitution in the imine and the ketene. The δ-lactams derived from dichloroketene are easily dehydrochlorinated to the corresponding 2-pyridones. All of the results are consistent with a two-step cycloaddition process involving a dipolar intermediate.

Several reports have appeared in the literature on the (2 + 2) cycloaddition of ketenes with imines to yield the biologically active β-lactams.[1-5] It has recently been established that these cycloadditions occur via a dipolar intermediate.[6] There are some scattered reports on the cycloaddition of α,β-unsaturated imines with ketenes, and some (4 + 2) cycloaddition products have been reported.[7,8] Since this reaction is occurring via a dipolar intermediate, α,β-unsaturated imines do, in fact, offer the possibility for ring closure to (4 + 2) cycloaddition products, and this could be a significant synthetic development for the preparation of δ-lactams and/or 2-pyridones. Therefore, this report describes a study of the reaction of diphenyl- and dichloroketenes with various α,β-unsaturated imines to determine the synthetic utility of this reaction for the preparation of β- and δ-lactams. During the course of this investigation, we learned of results on the cycloaddition of cyanoketenes with α,β-unsaturated imines.[9]

The cycloaddition of freshly distilled diphenylketene with α,β-unsaturated imines occurs readily in good yields at ambient temperatures to give the (2 + 2) cycloaddition products as shown in Scheme I. The reactions were complete within 1 h, and the solid products were easily isolated and purified by recrystallization. The structures of compounds **1a–k** are based primarily on the carbonyl band in the infrared at 1720–1742 cm^{-1}, proton and ^{13}C NMR, and elemental analysis. It is well established in the literature that the carbonyl absorption of β-lactams in the infrared occurs in the 1740 cm^{-1} range.[7,10] There was no evidence of any of the (4 + 2) cycloaddition products, the δ-lactams, in any of these cycloadditions. Some characteristic ^{13}C NMR data for compounds **1a–c,g** are shown in Table I.

The α,β-unsaturated imines were selected so that the steric requirement on both the N substituent and substitution on the carbon skelton would be varied. A wide

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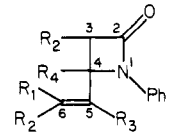
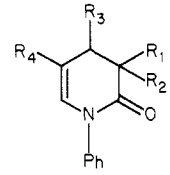
Table I. Characteristic ^{13}C NMR Data for Compounds 1a-c,g



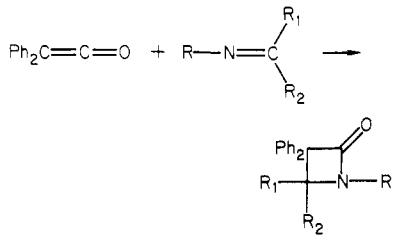
compd	R	R_1	R_2	R_3	R_4	chemical shift, ppm C_2	C_3	C_4	C_6	C_5 and Ph
1a	Ph	Ph	H	H	H	166.5	70.62	66.01	117.05	122.78-140.12
1b	Ph	Ph	H	CH_3	H	166.6	70.49	70.50	116.77	123.97-140.46
1c	Ph	Ph	H	H	CH_3	166.9	68.82	74.56	118.17	124.13-138.76
1g	Ph	CH_3	CH_3	H	CH_3	167.09	68.87	75.10	117.79	123.86-139.30

Table II. ^{13}C NMR Data for 3,4-Dihydro-2-pyridones 2a-d

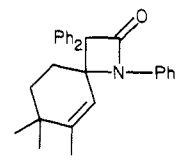


compd	R_1	R_2	R_3	R_4	chemical shift, ppm C_2	C_3	C_4	C_5	C_6 and Ph
2a	Ph	Ph	$(CH_3)_2N$	H	170.56	41.19	59.34	102.35	125.6-142.71
2b	Cl	Cl	Ph	H	159.2	84.7	53.74	107.6	123.84-133.7
2c	Cl	Cl	Ph	CH_3	158.14	83.9	60.24	116.9	116.96-138.25
2d	Cl	H	Ph	CH_3	163.19	60.47	50.83	118.76	125.38-140.06

Scheme I

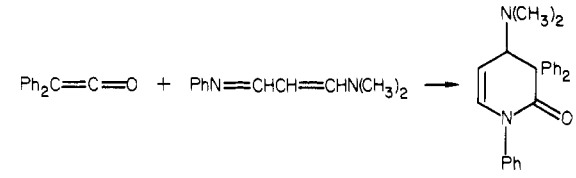


	R	R_1	R_2
1a	Ph	H	PhCH=CH
b	Ph	H	PhCH=C—CH_3
c	Ph	CH_3	PhCH=CH
d	Ph	H	
f	*i*-Pr	H	PhCH=CH
g	Ph	CH_3	$(CH_3)_2C$=CH
h	Ph	H	
i	C_6H_{11}	H	PhCH=CH
j	Ph	H	$(Ph)_2C$=CH
k	*t*-Bu	H	PhCH=CH



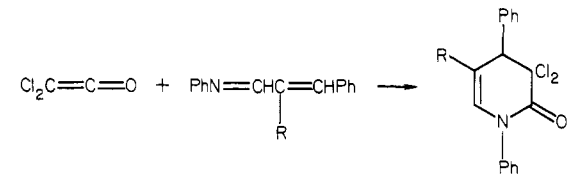
1e

Scheme II



2a

Scheme III



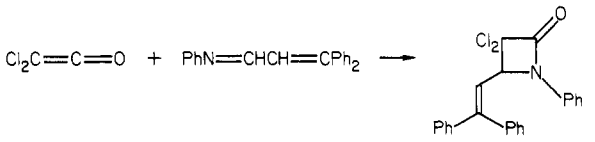
2b, R = H
c, R = CH_3

variation in the substituents on the imine did not alter the course of the (2 + 2) cycloaddition with diphenylketene as shown in Scheme I. There was, however, one exception to this general trend, and that was the *N*-phenyl imine of 3-(dimethylamino)-2-propenal, as this imine yielded a (4 + 2) cycloaddition products, **2a**, a 3,4-dihydro-2-pyridone (Scheme II). The structure of compound **2a** was assigned on the basis of the carbonyl band in the infrared at 1675 cm^{-1}, the proton and ^{13}C NMR data, and elemental analysis. The carbonyl group of δ-lactams occurs in the infrared in the 1660-cm^{-1} range.[7,8] The ^{13}C NMR data for this compound are recorded in Table II.

The in situ generation of dichloroketene from dichloroacetyl chloride with triethylamine in the presence of the *N*-phenyl imine of cinnamaldehyde and the *N*-phenyl imine of α-methylcinnamaldehyde resulted in the formation of the (4 + 2) cycloaddition products, the 3,4-dihydro-2-pyridones **2b** and **2c** (Scheme III). The structures of these cycloadducts were assigned on the basis of the carbonyl band in the infrared at 1680–1700 cm^{-1} and the proton and ^{13}C NMR spectra as well as the elemental analysis. The presence of substituents such as chlorine atoms in the α-position of δ-lactams results in a shifting of the carbonyl absorption in the infrared to the 1690-cm^{-1} range.[7,8] The ^{13}C NMR data for these compounds may be found in Table II.

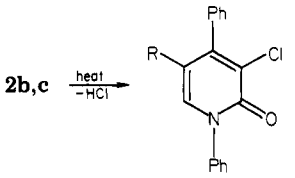
The in situ reaction of dichloroketene with the *N*-phenyl imine of β-phenylcinnamaldehyde resulted in the formation of the (2 + 2) cycloaddition product, the 2-azetidinone

Scheme IV



11

Scheme V



3a, R = H
b, R = CH_3

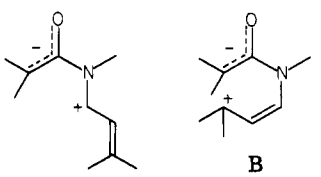
11 (Scheme IV). This compound revealed a carbonyl band in the infrared at 1785 cm^{-1}.

In all of the above-described cycloadditions, an aliquot of the crude reaction mixture was removed and analyzed by infrared spectroscopy. We did not find any examples of bands in the infrared spectrum in *both* the 1740 and 1660 cm^{-1} ranges, indicating the presence of both β- and δ-lactams. Hence, the cycloadditions occurred to give either the (2 + 2) or (4 + 2) cycloadducts as far as we could determine. The yields of the dichloroketene cycloadditions were significantly lower (49–66%) than the diphenylketene cycloadditions (62–84%), and these cycloadditions were accompanied by the formation of dichloroketene polymer. Reactions of dichloroketene with other α,β-unsaturated imines did not result in isolable products.

To determine if the (4 + 2) cycloaddition products might be formed from the (2 + 2) cycloadducts, several of the (2 + 2) cycloadducts were refluxed in *o*-dichlorobenzene overnight with no change. However, refluxing the δ-lactams derived from dichloroketene in *o*-dichlorobenzene resulted in dehydrochlorination to yield the 2-pyridones **3a** and **3b** (Scheme V).

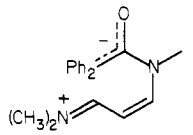
The hydrogenation of the 3,4-dihydro-2-pyridone with palladium black resulted in the reductive removal of only one of the α-chlorine atoms to yield compound **2d** which revealed a carbonyl band in the infrared at 1685 cm^{-1}. The ^{13}C NMR data for this compound are found in Table II.

The above-described results are quite consistent with a two-step process involving a dipolar intermediate. Conformations of a dipolar intermediate that would be expected to lead to the (2 + 2) and the (4 + 2) cycloaddition products are shown as A and B. Apparently, in



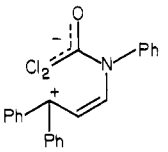
A B

the cycloaddition of diphenylketene with α,β-unsaturated imines the large phenyl groups provide significant steric hindrance in B, and cycloaddition occurs from A rather than B, leading to the (2 + 2) cycloaddition products. However, in the case of dichloroketene, the chlorine atoms do not present the steric problems that the phenyl groups do, and the cycloaddition generally yields the more stable (4 + 2) cycloadducts. There are exceptions to both of these general trends, but they are not surprising when considered in view of the dipolar intermediate. The cycloaddition of the *N*-phenyl imine of 3-(dimethylamino)-2-propenal with diphenylketene yields the (4 + 2) cycloadducts, the δ-lactam. The strong electron-releasing ability of the dimethylamino group would be expected to cause resonance structure C to make a strong contribution to the structure of the dipolar intermediate, thus leading to the (4 + 2) cycloadduct.



C

The cycloaddition of the *N*-phenyl imine of β-phenylcinnamaldehyde and dichloroketene yields the (2 + 2) cycloaddition product rather than the (4 + 2) cycloadduct (see D). Again, this result is expected when an examination of the conformation of the dipolar intermediate that would lead to the (4 + 2) cycloadduct is made. The steric problems for ring closure parallel the above-described case for the diphenylketene cycloadditions.



D

In summary, the results described in this study suggest that the cycloaddition of ketenes with α,β-unsaturated imines are primarily controlled by steric effects in the proposed dipolar intermediate.

Experimental Section

Proton NMR spectra were recorded on a Perkin-Elmer R-24B nuclear magnetic resonance spectrometer, employing deuteriochloroform as the solvent with tetramethylsilane as the internal standard. ^{13}C NMR spectra were obtained on a JEOL FX-90Q FT nuclear magnetic resonance spectrometer.

Ether and triethylamine were dried and purified by distillation from sodium–potassium alloy prior to use. Diphenylketene was prepared from diphenylacetyl chloride as previously described.

α,β-Unsaturated Imines. General Procedure. (a) From α,β-Unsaturated Aldehydes. A solution of 0.2 mol of freshly distilled amine was added over 10 min to a stirred solution of 0.2 mol of α,β-unsaturated aldehydes in 50 mL of dry ether. The resulting mixture was allowed to stand at ambient temperature for about 30 min, and then the ether was evaporated under reduced pressure. The imines were vacuum distilled or recrystallized from 95% ethanol.

(b) From α,β-Unsaturated Ketones. A solution of 0.2 mol of the amine, 0.2 mol of the α,β-unsaturated ketone, and a catalytic amount of zinc chloride in 80 mL of benzene was refluxed under a water separator until water ceased to be formed. The zinc chloride was removed by filtration, the benzene was removed under reduced pressure, and the imines were recrystallized from 95% ethanol.

Typical Procedure for Diphenylketene Cycloadditions with α,β-Unsaturated Imines. To a stirred solution of 0.021 mol of the α,β-unsaturated imine in 100 mL of dry ether was added 0.020 mol of freshly distilled diphenylketene in 30 mL of dry ether under a nitrogen atmosphere at ambient temperature. When the ketene band in the IR at 2100 cm^{-1} had disappeared (usually about 1 h), the solution was concentrated on a rotatory evaporator and the residue recrystallized from 95% ethanol and benzene.

Typical Procedure for Dichloroketene Cycloadditions with α,β-Unsaturated Imines. A solution of 0.020 mol of freshly distilled dichloracetyl chloride in 50 mL of dry ether was added over a 1-h period to a stirred solution of 0.021 mol of α,β-unsaturated imine and 0.022 mol of triethylamine in 200 mL of dry ether at ambient temperature under a nitrogen atmosphere. The

resulting mixture was stirred an additional 30 min and the amine salt removed by filtration. The filtrate was concentrated on a rotatory evaporator, and the residue was recrystallized from 95% ethanol and benzene.

***N*,3,3-Triphenyl-4-styryl-2-azetidinone (1a).** There was obtained from diphenylketene and the *N*-phenyl imine of cinnamaldehyde: 5.9 g (74%); mp 172–173 °C; IR ($CDCl_3$) 1732 cm^{-1}; ^1H NMR δ 4.95 (d, 1 H), 5.35 (dd, 1 H), 6.45 (d, 1 H), 6.8 (m, 20 H). Anal. Calcd for $C_{29}H_{23}NO$: C, 86.78; H, 5.73. Found: C, 86.69; H, 5.61.

***N*,3,3-Triphenyl-4-(1-methylstyryl)-2-azetidinone (1b).** There was obtained from diphenylketene and the *N*-phenyl imine of α-methylcinnamaldehyde: 5.9 g (72%); mp 131.5–133.5 °C; IR ($CDCl_3$) 1738 cm^{-1}; ^1H NMR δ 1.2 (s 3 H), 5.0 (s, 1 H), 6.4 (s, 1 H), 6.9 (m, 20 H). Anal. Calcd for $C_{30}H_{25}NO$: C, 86.74; H, 6.02. Found: C, 86.70; H, 5.80.

***N*,3,3-Triphenyl-4-methyl-4-styryl-2-azetidinone (1c).** From diphenylketene and the *N*-phenyl imine of 4-phenyl-3-buten-2-one was obtained: 5.2 g (63%); mp 146.5–148 °C; IR ($CDCl_3$) 1739 cm^{-1}; ^1H NMR δ 1.7 (d, 3 H), 5.87 (d, 1 H), 6.4 (d, 1 H), 7.2 (m, 20 H). Anal. Calcd for $C_{30}H_{25}NO$: C, 86.74; N, 3.37; H, 5.2. Found: C, 87.01; N, 3.50; H, 6.04.

***N*,3,3-Triphenyl-4-(2-furfuryl)-2-azetidinone (1d).** From diphenylketene and the *N*-phenyl imine of furfural was obtained **1d**: 4.5 g (62%); mp 145.5–146.5 °C; IR ($CDCl_3$) 1742 cm^{-1}; ^1H NMR δ 5.5 (s, 1 H), 5.75 (d, 2 H), 6.9 (m, 16 H). Anal. Calcd for $C_{25}H_{19}NO_2$: C, 82.19; N, 3.84; H, 5.2. Found: C, 82.45; N, 3.87; H, 4.99.

***N*,3,3-Triphenyl-5,5,7-trimethyl-1-aza-2-oxospiro[5.3]-non-7-ene (1e).** From diphenylketene and the *N*-phenyl imine of isophorone was obtained **1e**: 6.2 g (76%); mp 130–131.5 °C; IR ($CDCl_3$) 1722 cm^{-1}; ^1H NMR δ 0.65 (s, 3 H), 0.7 (s, 3 H), 1.68 (s, 3 H), 1.79 (s, 4 H), 5.18 (s, 1 H), 7.2 (m, 15 H). Anal. Calcd for $C_{29}H_{29}NO$: C, 84.79; N, 3.27; H, 7.13. Found: C, 85.18; N, 3.22; H. 6.87.

***N*-Isopropyl-3,3-diphenyl-4-styryl-2-azetidinone (1f).** There was obtained from diphenylketene and the *N*-isopropyl imine of cinnamaldehyde **1f**: 5.3 g (77%); mp 110–111 °C; IR ($CDCl_3$) 1730 cm^{-1}; ^1H NMR δ 1.1 (d, 3 H), 1.22 (d, 3 H), 3.75 (m, 1 H), 4.15 (d, 1 H), 5.4 (d, 1 H), 6.4 (d, 1 H), 6.9 (m, 15 H). Anal. Calcd for $C_{26}H_{25}NO$: C, 85.01; N, 3.81; H, 6.81. Found: C, 84.75; N, 3.81; H, 6.87.

***N*,3,3-Triphenyl-4-methyl-4-(2-methylpropenyl)-2-azetidinone (1g).** From diphenylketene and the *N*-phenyl imine of 4-methyl-3-penten-2-one there was obtained **1g**: 5.1 g (70%); mp 135.5–136 °C; IR ($CDCl_3$) 1720 cm^{-1}; ^1H NMR δ 1.05 (s, 3 H), 1.4 (s, 3 H), 1.55 (s, 3 H), 5.2 (s, 1 H), 7.1 (m, 15 H). Anal. Calcd for $C_{26}H_{25}NO$: C, 85.01; N, 7.61; H, 6.52. Found: C, 85.14; N, 7.46; H, 6.52.

***N*,3,3-Triphenyl-4-(5,6-dihydro-2*H*-pyran-3-yl)-2-azetidinone (1h).** From diphenylketene and the *N*-phenyl imine of 5,6-dihydro-2*H*-pyran-3-carboxaldehyde there was obtained **1h**: 6.4 g (84%); mp 144–146 °C; IR ($CDCl_3$) 1740 cm^{-1}; ^1H NMR δ 1.95 (s, 3 H), 3.18 (t, 3 H), 3.42 (s, 3 H) 4.9 (s, 1 H), 7.0 (m, 15 H). Anal. Calcd for $C_{26}H_{25}NO_2$: C, 81.69; N, 3.67; H, 6.04. Found: C, 81.38; N, 3.51; H, 5.83.

***N*-Cyclohexyl-3,3-diphenyl-4-styryl-2-azetidinone (1i).** From diphenylketene and the *N*-cyclohexyl imine of cinnamaldehyde there was obtained **1i**: 6.7 g (82%); mp 139–140 °C; IR ($CDCl_3$) 1730 cm^{-1}; ^1H NMR δ 1.5 (m, 10 H), 3.2 (m, 1 H), 4.65 (d, 1 H), 5.38 (dd, 1 H), 6.45 (d, 1 H), 7.0 (m, 15 H). Anal. Calcd for $C_{29}H_{29}NO$: C, 85.50; N, 3.44; H, 7.13. Found: C, 85.38; N, 3.34; H, 7.14.

***N*,3,3-Triphenyl-4-(2-phenylstyryl)-2-azetidinone (1j).** From diphenylketene and the *N*-phenyl imine of β-phenylcinnamaldehyde there was obtained **1j**: 8.0 g (84%); mp 199–200.5 °C; IR ($CDCl_3$) 1734 cm^{-1}; ^1H NMR δ 5.24 (d, 1 H), 5.49 (d, 1 H), 7.04 (m, 25 H). Anal. Calcd for $C_{35}H_{27}NO$: C, 88.05; N, 2.94; H, 5.66. Found: C, 88.10; N, 2.71: H, 5.36.

***N*-*tert*-Butyl-3,3-diphenyl-4-styryl-2-azetidinone (1k).** From diphenylketene and the *N*-*tert*-butyl imine of cinnamaldehyde there was obtained **1k**: 6.3 g (83%); mp 158–159.5 °C; IR ($CDCl_3$) 1729 cm^{-1}; ^1H NMR δ 1.3 (s, 9 H), 4.6 (d, 1 H), 5.35 (dd, 1 H), 6.4 (d, 1 H), 6.9 (m, 15 H). Anal. Calcd for $C_{27}H_{27}NO$: C, 85.04; N, 3.67; H, 7.09. Found: C, 85.15; N, 3.58; H, 6.98.

***N*,3,3-Triphenyl-4-(dimethylamino)-3,4-dihydro-2-pyridone (2a).** From diphenylketene and the *N*-phenyl imine of 3-(dimethylamino)-2-propenal there was obtained **2a**: 4.7 g (64%); mp 129.5–131 °C; IR ($CDCl_3$) 1675 cm^{-1}; ^1H NMR δ 2.15 (s, 6 H), 3.8 (d, 1 H), 5.2 (dd, 1 H), 6.0 (d, 1 H), 6.9 (m, 15 H). Anal. Calcd for $C_{25}H_{24}N_2O$: C, 81.52; N, 7.61; H, 6.52. Found: C, 81.34; N, 7.46; H, 6.52.

3,3-Dichloro-3,4-dihydro-*N*,4-diphenyl-2-pyridone (2b). From dichloroketene and the *N*-phenyl imine of cinnamaldehyde there was obtained **2b**: 3.1 g (49%); mp 147–148 °C; IR ($CDCl_3$) 1680 cm^{-1}; ^1H NMR δ 4.1 (dd, 1 H), 5.15 (dd, 1 H), 6.1 (dd, 1 H), 6.95 (m, 10 H). Anal. Calcd for $C_{17}H_{13}Cl_2NO$: C, 64.16; N, 4.40; H, 3.87. Found: C, 64.42; N, 4.12; H, 3.87.

3,3-Dichloro-3,4-dihydro-5-methyl-*N*,4-diphenyl-2-pyridone (2c). From dichloroketene and the *N*-phenyl imine of α-methylcinnamaldehyde there was obtained **2c**: 4.4 g (66%); mp 113–114.5 °C; IR ($CDCl_3$) 1700 cm^{-1}; ^1H NMR δ 1.55 (s, 3 H), 3.7 (s, 1 H), 5.85 (s, 1 H), 6.9 (m, 10 H). Anal. Calcd for $C_{18}H_{15}Cl_2NO$: C, 65.08; N, 4.22; H, 4.55. Found: C, 65.28; N, 4.35; H, 4.37.

3,3-Dichloro-*N*-phenyl-4-(2-phenylstyryl)-2-azetidinone (1l). From dichloroketene and the *N*-phenyl imine of β-phenylcinnamaldehyde there was obtained **1l**: 4.8 g (61%); mp 136.5–137 °C; IR ($CDCl_3$) 1785 cm^{-1}; ^1H NMR δ 4.66 (d, 1 H), 5.24 (d, 1 H), 7.05 (m, 15 H). Anal. Calcd for $C_{23}H_{17}Cl_2NO$: C, 70.05; N, 3.55; H, 4.31. Found: 70.30; N, 3.48; H, 4.30.

3-Chloro-*N*,4-diphenyl-2-pyridone (3a). A 1.0-g portion of **2b** was refluxed in *o*-dichlorobenzene for 3 h, and there was obtained **3a**: 0.87 g (98%); mp 185.8–187 °C; IR ($CDCl_3$) 1661 cm^{-1}; ^1H NMR δ 6.0 (d, 1 H), 6.95 (d, 1 H), 7.1 (m, 10 H). Anal. Calcd for $C_{17}H_{12}ClNO$: C, 72.47; N, 4.97; H, 4.26. Found: C, 72.17; N, 5.09; H, 4.06.

3-Chloro-5-methyl-*N*,4-diphenyl-2-pyridone (3b). A 1.0-g portion of **2c** was refluxed in *o*-dichlorobenzene for 3 h, and there was obtained 0.89 g (100%) of **3b**: mp 166–167.5 °C; IR ($CDCl_3$) 1650 cm^{-1}; ^1H NMR δ 1.7 (s, 3 H), 7.0 (m, 11 H). Anal. Calcd for $C_{18}H_{14}ClNO$: C, 73.1; N, 4.74; H, 4.74. Found: C, 72.91; N, 4.48; H, 4.74.

3-Chloro-3,4-dihydro-5-methyl-*N*,4-diphenyl-2-pyridone (2d). A 1.0-g portion of **2c**, 0.2 g of palladium black, and 20 mL of benzene were agitated in a hydrogenation apparatus for 8 h, and there was obtained **2d**: 0.87 g (98%); mp 120–122 °C; IR ($CDCl_3$) 1685 cm^{-1}; ^1H NMR δ 1.5 (s, 3 H), 3.45 (d, 1 H), 4.85 (d, 1 H), 5.95 (d, 1 H), 6.95 (m, 10 H). Anal. Calcd for $C_{18}H_{16}ClNO$: C, 72.60; N, 4.71; H, 5.38. Found: C, 72.43; N, 4.63; H, 5.15.

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Registry No. **1a**, 30340-68-2; **1b**, 86129-83-1; **1c**, 86129-84-2; **1d**, 86129-85-3; **1e**, 86129-86-4; **1f**, 86129-87-5; **1g**, 86129-88-6; **1h**, 86129-89-7; **1i**, 86129-90-0; **1j**, 86129-91-1; **1k**, 86129-92-2; **1l**, 86129-93-3; **2a**, 86129-94-4; **2b**, 86129-95-5; **2c**, 86129-96-6; **2d**, 86129-97-7; **3a**, 27296-22-6; **3b**, 86129-98-8; diphenylketene, 525-06-4; dichloroketene, 4591-28-0; cinnamaldehyde *N*-phenylimine, 953-21-9; α-methylcinnamaldehyde *N*-phenylimine, 86129-99-9; 4-phenyl-3-buten-2-one *N*-phenylimine, 17424-79-2; furfural *N*-phenylimine, 3237-23-8; isophorone *N*-phenylimine, 36755-22-3; cinnamaldehyde *N*-isopropylimine, 42956-08-1; 4-methyl-3-penten-2-one *N*-phenylimine, 64723-73-5; 5,6-dihydro-2*H*-pyran-3-carboxaldehyde *N*-phenylimine, 86130-00-9; cinnamaldehyde *N*-cyclohexylimine, 15286-54-1; β-phenylcinnamaldehyde *N*-phenylimine, 86130-01-0; cinnamaldehyde *N*-*tert*-butylimine, 29940-86-1; 3-(dimethylamino)-2-propenal *N*-phenylimine, 1534-15-2.