

Ultrasonic-Assisted Synthesis of 1,3-Diaryl-2,2-dichloroaziridine Derivatives in the Presence of Phase-Transfer Catalyst under Low-Concentration Alkaline Conditions

Hossein Naeimi* and Khadijeh Rabiei

Department of Organic Chemistry, Faculty of Chemistry, University of Kashan, Kashan 87317, Iran

Received April 12, 2011; E-mail: naeimi@kashanu.ac.ir

In this research, rapid and efficient preparation of 1,3-diaryl-2,2-dichloroaziridines through the reaction of Schiff base compounds with dichlorocarbene yielded in situ in the presence of cetyltrimethylammonium bromide (CTAB) as phase-transfer catalyst under ultrasonic irradiation is described. The advantages of this reaction are very short reaction times, excellent product yields, simplicity of the method, and high purity of products.

The use of ultrasonic waves in organic synthesis has been boosted in recent years.[1–12] Their specificity has been demonstrated in homogeneous as well as in heterogeneous reactions.[13–16] Sonification of multiphase systems accelerates the reaction by ensuring a better contact between the different phases.[10,11,16] Ultrasound is known to accelerate diverse types of organic reactions and it is established as an important technique in organic synthesis.[8,17,18] Sonication also increases the reaction rate and avoids the use of high reaction temperatures.[10,19] A number of organic reactions have been revisited by means of ultrasonic waves.[8,17,18] Tao et al. have reported the reaction of amines, amides, imines, alcohols, aldehyde, and ketones with dichlorocarbene under ultrasound.[20] The advantages of ultrasound procedures, such as good yields, short reaction times, and mild reaction conditions, are well documented.[21]

Aziridines, which are extremely important synthetic building blocks, are nitrogen equivalents of epoxides, and can be similarly opened in a stereocontrolled manner with various nucleophiles, providing access to a wide range of important nitrogen-containing products.[22] These compounds are among the most fascinating intermediates in organic synthesis, acting as precursors of many complex molecules due to the strain incorporated in their skeletons.[23] Since the first synthesis of an aziridine reported by Gabriel in 1888,[24] the synthetic scope of aziridine chemistry has blossomed in recent years. Thus, obtaining aziridines, has become of great importance in organic chemistry. In particular, the antitumor and antibiotic properties of a great number of aziridine-containing compounds are of high significance among other biological properties, which make them attractive synthetic targets in their own right.[25]

With attention to the importance of *gem*-dihaloaziridines, several methods for their synthesis have been reported. The preparation has been accomplished by the addition of dichlorocarbene generated from chloroform, hexachloroacetone, or ethyl trichloroacetate with the appropriate base[26–28] and chloroform/NaOH/TEBAC (triethylbenzylammonium chloride) as phase-transfer catayst[29–31] to imines.

Ever since Laurent and Gerhard[32] synthesized the first organic imine by condensation of benzaldehyde with aniline, numerous investigations have dealt with this class of compounds, also known as Schiff bases. These compounds have been extensively studied in chemistry due to their facile syntheses, easily tunable steric and electronic properties and good solubility in common solvents.[33–36]

Phase-transfer catalysis (PTC) technology has been widely applied to synthesize specialty chemicals from organic reactions.[37–39] Recently many chemists have investigated phase-transfer catalysis in numerous reactions. As a result, PTC is considered to have great potential for industrial-scale application.[40]

In conjunction with ongoing work in our laboratory on the preparation of Schiff base derivatives,[41–44] here we decided to report the synthesis of various dichloroaziridines through the reaction of various Schiff base compounds and chloroform in the presence of cetyltrimethylammonium bromide (CTAB) as phase-transfer catalyst under ultrasonic irradiation.

Results and Discussion

In this research, the phase-transfer-catalyzed reaction of dichloroaziridination of Schiff base compounds has been studied under ultrasonic irradiation. When 0.028 mol of Schiff base compound was reacted with dichlorocarbene intermediate obtained in situ from the reaction of chloroform and base in the presence of CTAB as a phase-transfer catalyst under ultrasonic irradiation, corresponding products, 1,3-diaryl-2,2-dichloroaziridine compounds were obtained under low concentration alkaline conditions (9.3% NaOH) (Scheme 1).

First, we carried out this reaction in the presence of various amounts of NaOH under two phase-transfer catalysts, namely CTAB and CPB (1-cetylpyridinium bromide), and ultrasonic irradiation (55 W). The corresponding results are indicated in Table 1.

As can be seen in this table, the desired product was obtained with high yield in the presence of CTAB rather than CPB as phase-transfer catalyst using NaOH (9.3%). In order to

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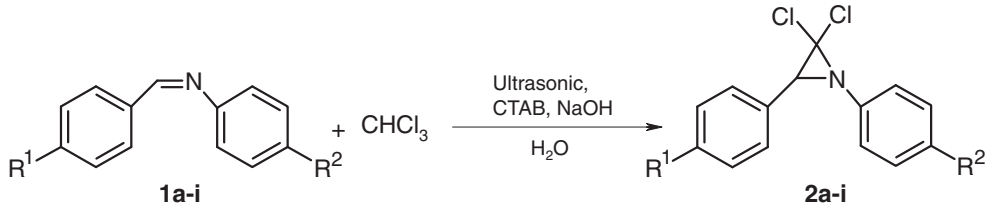


R^1 = H, CH_3, NO_2, Cl

R^2 = H, CH_3, Cl, Br

Scheme 1. Preparation of 1,3-diaryl-2,2-dichloroaziridine compounds from Schiff bases.

Table 1. Enhancement of Phase-Transfer Catalyst (PTC) and Ultrasonic Irradiation (55 W) on the Formation of 1-(4-Bromophenyl)-2,2-dichloro-3-(4-chlorophenyl)aziridine in the Presence of Various Amounts of NaOH

Entry	PTC	Amount of NaOH/%	Time /min	Yield[a)] /%
1	CTAB	5	90	50
2	CTAB	5.6	65	60
3	CTAB	6.7	50	72
4	CTAB	8.3	40	80
5	CTAB	9.3	25	90
6	CPB	9.3	45	78
7	CPB	12	30	84
8	None[b)]	9.3	720	40

a) Isolated yields. b) The reaction without PTC under magnetic stirring.

Table 2. Survey of the Effect of Ultrasonic Irradiation on the Formation of 1-(4-Bromophenyl)-2,2-dichloro-3-(4-chlorophenyl)aziridine under CTAB Catalysis Conditions

Entry	Power/W	Time/min	Yield[a)]/%
1	35	100	80
2	40	90	85
3	45	75	88
4	50	55	90
5	55	25	90
6	60	15	94
7	65	8	98
8	70	8	98
9	None[b)]	160	55

a) Isolated yields in 9.3% NaOH. b) The reaction without ultrasonic irradiation.

determine enhancement ultrasound irradiation and phase-transfer catalyst, with performance of this reaction without phase-transfer catalyst under magnetic stirring, the products were obtained in low yield and long reaction times (Table 1, Entry 8).

In continuation of this research, the effect of various powers of ultrasonic irradiation has been surveyed. Initially we carried out aziridination of 4-bromo-*N*-(4-chlorobenzylidene)aniline as model reaction in order to optimize the best suited reaction conditions. It was observed that the reaction in the presence of NaOH 9.3% and ultrasonic irradiation of 65 W afforded the best result as product was obtained in 98% isolated yield after 8 min (Table 2, Entry 7), while the reaction in the presence of PTC without ultrasonic irradiation was carried out in low yield and long reaction time (Table 2, Entry 9). Sonification of multiphasic systems accelerates the reaction by ensuring a better contact between the different phases. By executing the reaction under sonication conditions dichlorocarbene reagent was produced very quickly and thus corresponding products were obtained in excellent yields and very short reaction times.

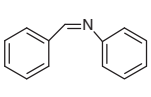
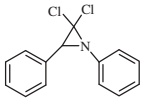
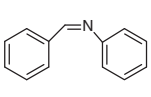
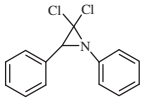
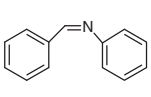
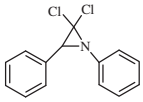
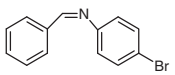
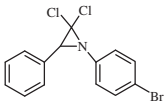
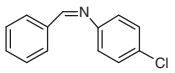
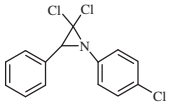
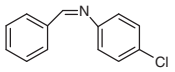
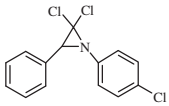
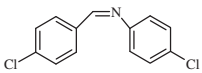
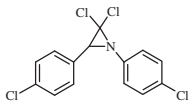
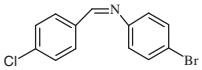
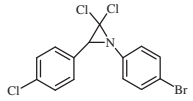
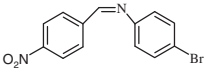
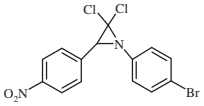
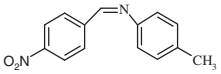
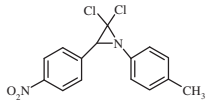
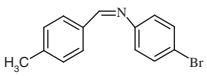
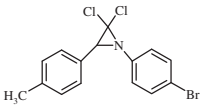
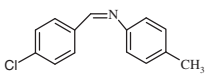
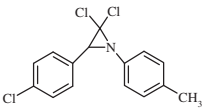
The effect of ultrasonic irradiation observed during organic reactions is due to cavitation. In the case of volatile molecules cavities are believed to act as a microreactor: as the volatile molecules enter the microbubbles the high temperature and pressure produced during cavitation break their chemical bonds thus reacting with other species.[3,45,46]

In the PTC reactions rate enhancements are typically due to mechanical effects, mainly through an enhancement in mass transfer. In the presence of ultrasonic irradiation in liquid–liquid–PTC systems, cavitational coallapse near the liquid–liquid interface disrupts the interface and impels jets of one liquid in to the other forming fine emulsion, leading to a dramatic increase in the interfacial contact area across which transfer of species can take place. In the case of PTC reaction under ultrasonic irradiation, the PTC catalyst initials' the reaction by transfer of species across the interface and ultrasound merely facilitates this transfer, possibly by increasing the interfacial area across which this transfer occurs. Also, the ultrasonic irradiation increases the production of $:CCl_2$ intermediate and causes the enhancement of addition reaction rate and thus corresponding products are obtained in excellent yield and very short reaction times.

To then ascertain the scope and limitation of the present reaction, several Schiff base compounds were reacted with $CHCl_3$ in the presence of a mixture of CTAB and the optimum amount of NaOH (9.3%) under ultrasonic irradiation and the desired dichloroaziridine derivatives were prepared. The results are summarized in Table 3. As shown in this table, the reaction of the various Schiff bases with chloroform and NaOH (9.3%), were sonicated and catalyzed by CTAB as two-phase transfer. The corresponding products were obtained in excellent yields and appropriate reaction times in the presence of PTC under ultrasonic irradiation.

The synthesis of *gem*-dichloroaziridines in the presence of PTC under ultrasonic irradiation was compared with previously reported methods. In this method, $CHCl_3$/NaOH/Schiff base mol ratio used was 2.5/2.5/1 and the desired product **2a** was obtained in excellent yield (94%) after short reaction time

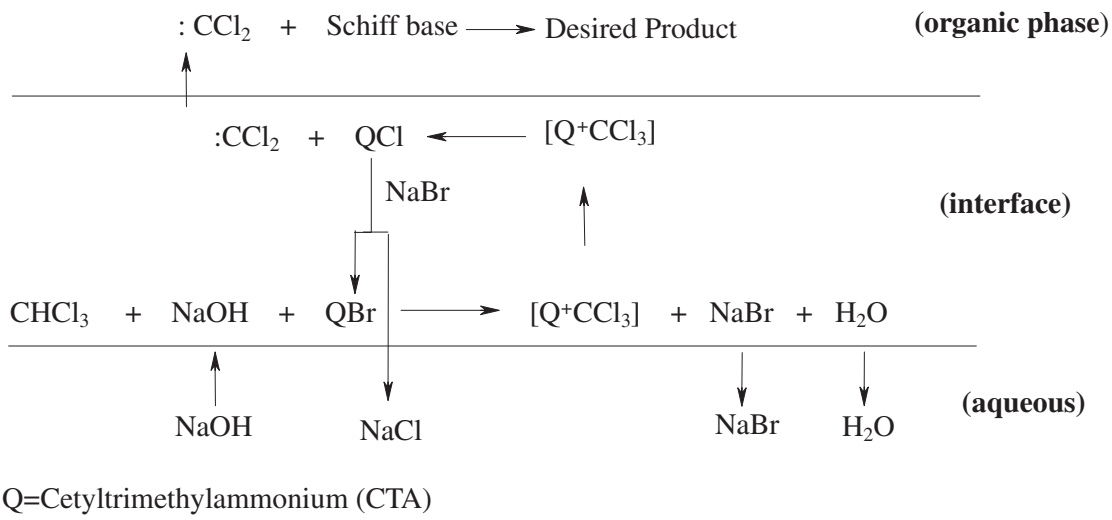
Table 3. Synthesis of 1,3-Diaryl-2,2-dichloroaziridine in the Presence of CTAB Catalyst and 9.3% NaOH under Ultrasonic Irradiation with 65 W Power

Entry	Substrate	Product		Mp/°C	Time/min	Yield[a]/%
1			**2a**	100–102	15	94
2			**2a**	98–99	360	61[b]
3			**2a**	98–99	400	55[c]
4			**2b**	143–145	12	94
5			**2c**	72–74	12	95
6			**2c**	71–72	960	68[d]
7			**2d**	139–141	8	97
8			**2e**	134–136	8	98
9			**2f**	141–143	13	92
10			**2g**	140–142	14	92
11			**2h**	146–148	11	96
12			**2i**	128–130	10	97

a) Isolated yields. b) By hexachloroacetone and sodium methoxide, Ref. 26. c) By sodium methoxide and chloroform, Ref. 27. d) By potassium *t*-butoxide and chloroform, Ref. 28.

(15 min) (Table 3, Entry 1), while, in previously reported methods, ($CHCl_3$ or Cl_3COCl_3)/$NaOCH_3$/Schiff base mol ratio used was 4/4/1 and corresponding product was obtained in 55 and 61% yields after several hours in the absence of PTC (Table 3, Entries 2 and 3).[26,27] On the other hand, synthesis of this compound **2a** was reported in the presence of TEBAC as a PTC without ultrasonic irradiation in which $CHCl_3$/NaOH/Schiff base mol ratio used was 10/25/1 and the desired product was obtained in 74 and 88% yields.[29,30] Also preparation of product **2c** by using this method was carried out in excellent yield and very short reaction time that is comparable with the reported method (Table 3, Entry 5 vs. 6).[28] It seems this method for preparation of *gem*-dichloroaziridines has some advantages such as high efficiency, and mild conditions compared to other protocols.

The structure of products has been confirmed by physical and spectroscopic data such as IR, ^{1}H NMR, ^{13}C NMR, mass spectroscopy, and C. H. N. elemental analyses. In the IR



Scheme 2. The proposed reaction mechanism for preparation of *gem*-dihaloaziridines.

spectra, the stretching frequency of aromatic C=C is formed in the region between $\nu = 1490$–$1600\,cm^{-1}$. The stretching vibration of C–H in the alkyl groups appeared at a region between $\nu = 2898$–$2930\,cm^{-1}$. In the ^1H NMR spectra, one proton of CH–N has a chemical shift at δ 3.65–4.40. The signals around δ 6.59–8.55 are assigned to protons of CH=CH of aromatic rings. In the ^{13}C NMR spectra, one carbon of C–N has a chemical shift at δ 52.1–55.1 and the signal around δ 74.1–77.1 is assigned to one carbon of the CCl_2 of aziridine ring.

The Proposed Reaction Mechanism. The proposed reaction mechanism for synthesis of diarylaziridines is shown in Scheme 2. In this reaction, first, the dichlorocarbene ($:CCl_2$) as reactive intermediate is generated from the interfacial reaction of chloroform, sodium hydroxide, and quaternary ammonium salt to form an active complex ($QCCl_3$) of the dichlorocarbene ($:CCl_2$) precursor.

Second, the active complex $QCCl_3$ (or dichlorocarbene $:CCl_2$) transfers from the aqueous phase to the organic phase. Then, dichlorocarbene reacts with Schiff base to produce 1,3-diaryl-2,2-dichloroaziridine in the organic phase.

Conclusion

In this research, we have synthesized dichloroaziridine derivatives through the reaction of various Schiff base compounds with chloroform in the presence of cetyltrimethylammonium bromide (CTAB) under ultrasonic irradiation. The corresponding products have been obtained in excellent yields, high purity, and short reaction times. The products have been confirmed by physical and spectroscopic data such as; IR, ^1H NMR, ^{13}C NMR, MS spectroscopy, and C. H. N. elemental analyses.

Experimental

Materials. All the materials were of commercial reagent grade. All the Schiff bases were prepared according to previously reported procedures.[41,42]

Apparatus. IR spectra were recorded as KBr pellets on a Perkin-Elmer 781 spectrophotometer and an Impact 400 Nicolet FTIR spectrophotometer. ^1H NMR and ^{13}C NMR were recorded in DMSO/$CDCl_3$ solvents on a Bruker DRX-400 spectrometer with tetramethylsilane as internal reference. Mass spectra were recorded on a Finnigan MAT 44S by Electron Ionization (EI) mode with an ionization voltage of 70 eV. The elemental analyses (C. H. N.) were obtained from a Carlo ERBA Model EA 1108 analyzer. A BANDELIN ultrasonic HD 3200 with probe model KE 76, 6 mm diameter, was used to produce ultrasonic irradiation and homogenize the reaction mixture. Piezoelectric crystals of this kind of probe normally work in the range of 700 kHz, but using through proper clamps the output frequency of piezoelectric crystals have been controlled and reduced to 20 kHz. Therefore, the induced frequency of probe to the reaction mixture is equal to 20 kHz. By changing the power of Tip the cavitations rate is displaced so that the Tip frequency under various amount of power is constant. A thermal method was used for calibration of ultrasonic power. Melting points obtained with a Yanagimoto micro melting point apparatus are uncorrected. The purity determination of the substrates and reaction monitoring were accomplished by TLC on silica gel polygram SILG/UV 254 plates (from Merck Co.).

Typical Procedure for the Synthesis of 1,3-Diphenyl-2,2-dichloroaziridine. Measured quantities of NaOH (0.07 mol, 2.8 g) and CTAB (0.3 g) were dissolved in 30 mL of water. The mixed solution was introduced to a 100 mL flask. The ultrasonic probe was immersed directly in the reactor. The ultrasonic generator (HD 3200) emitted the 65 W sound vibration into the reaction mixture for two minutes. Then, Schiff base (organic reactant; 0.028 mol, 8.2 g) dissolved in chloroform (0.07 mol, 8.3 mL) was gradually added dropwise to the mixed solution. The progress of the reaction was monitored by TLC. After the completion of the reaction in 15 min, the solution was separated and the aqueous solution was extracted twice with ether. Magnesium sulfate was also added to adsorb the residual water. The organic solvent (chloroform) and other residues were stripped in a vacuum evaporator. The pale yellow solid, 1,3-diphenyl-2,2-dichloroaziridine, was obtained in 94% yield, mp 100–102 °C. All of the diarylaziridine products were identified by physical and spectroscopic data as follows.

1,3-Diphenyl-2,2-dichloroaziridine (2a): Pale yellow solid; mp 100–102 °C (mp 98–99 °C);[26,27] IR (KBr): ν/cm^{-1} 3090, 2930, 1600, 1520 (C=C, Ar); ^{1}H NMR ($CDCl_3$): δ 4.20 (s, 1H, HCN), 7.01–7.80 (m, 10H, Ar); ^{13}C NMR ($CDCl_3$): δ 52.1, 74.1, 119.0, 120.1, 125.1, 126.1, 127.1, 128.3, 134.2, 140.1; MS: m/z 268 (M + 4, 7), 266 (M + 2, 29), 264 (M^+, 45), 233 (34), 231 (54), 229 (100), 154 (55), 152 (86), 77 (95); Anal. Calcd for $C_{14}H_{11}Cl_2N$: C, 63.65; H, 4.19; N, 5.31%. Found: C, 63.67; H, 4.19; N, 5.30%.

1-(4-Bromophenyl)-2,2-dichloro-3-phenylaziridine (2b): White solid; mp 143–145 °C; IR (KBr): ν/cm^{-1} 3100, 2914, 1600, 1524 (C=C, Ar); ^{1}H NMR (DMSO): δ 4.40 (s, 1H, HCN), 7.10–7.80 (m, 9H, Ar); ^{13}C NMR (DMSO): δ 53.4, 77.1, 120.9, 121.3, 127.8, 128.1, 130.1, 131.2, 135.2, 142.1; MS: m/z 348 (M + 6, 10), 346 (M + 4, 25), 344 (M + 2, 45), 342 (M^+, 30), 309 (85), 307 (100), 233 (65), 230 (45), 152 (80), 77 (85); Anal. Calcd for $C_{14}H_{10}BrCl_2N$: C, 49.02; H, 2.94; N, 4.08%. Found: C, 49.15; H, 2.95; N, 4.12%.

1-(4-Chlorophenyl)-2,2-dichloro-3-phenylaziridine (2c): Pale yellow solid; mp 72–74 °C (mp 71–72 °C);[28] IR (KBr): ν/cm^{-1} 3090, 2900, 1598, 1499 (C=C, Ar); ^{1}H NMR ($CDCl_3$): δ 4.21 (s, 1H, HCN), 7.03–7.90 (m, 9H, Ar); ^{13}C NMR ($CDCl_3$): δ 53.1, 76.9, 119.1, 120.8, 121.1, 127.9, 128.1, 130.1, 131.2, 135.1, 142.2; MS: m/z 304.5 (M + 6, 7), 302.5 (M + 4, 15), 300.5 (M + 2, 32), 298 (M^+, 35), 263 (100), 261 (85), 174 (75), 172 (80), 91 (95), 77 (45); Anal. Calcd for $C_{14}H_{10}Cl_3N$: C, 56.31; H, 3.37; N, 4.69%. Found: C, 56.31; H, 3.38; N, 4.70%.

1,3-Bis(4-chlorophenyl)-2,2-dichloroaziridine (2d): White solid; mp 139–141 °C; IR (KBr): ν/cm^{-1} 3085, 2910, 1600, 1504 (C=C, Ar); ^{1}H NMR (DMSO): δ 4.34 (s, 1H, HCN), 7.13–7.55 (m, 8H, Ar); ^{13}C NMR (DMSO): δ 53.2, 75.9, 122.3, 128.9, 129.3, 129.7, 130.1, 132.2, 134.1, 143.6; MS: m/z 341 (M + 8, 4), 339 (M + 6, <3), 337 (M + 4, 4), 335 (M + 2, 10), 333 (M^+, 20), 298 (70), 296 (65), 174 (60), 172 (95), 161 (80), 159 (100), 89 (50), 77 (55); Anal. Calcd for $C_{14}H_9Cl_4N$: C, 50.49; H, 2.72; N, 4.20%. Found: C, 50.48; H, 2.74; N, 4.21%.

1-(4-Bromophenyl)-2,2-dichloro-3-(4-chlorophenyl)aziridine (2e): White solid; mp 134–136 °C; IR (KBr): ν/cm^{-1} 3100, 2920, 1598, 1509 (C=C, Ar); ^{1}H NMR (DMSO): δ 4.33 (s, 1H, HCN), 7.07–7.59 (m, 8H, Ar); ^{13}C NMR (DMSO): δ 53.1, 75.9, 122.7, 128.9, 129.7, 130.1, 132.2, 132.6, 134.1, 144.1; MS: m/z 384.5 (M + 8, 6), 382.5 (M + 6, <2), 380.5 (M + 4, 10), 378.5 (M + 2, 35), 376.5 (M^+, 25), 341.5 (75), 339.5 (67), 217.5 (89), 205.5 (78), 204.5 (100), 202.5 (90), 89 (60); Anal. Calcd for $C_{14}H_9BrCl_3N$: C, 44.54; H, 2.40; N, 3.71%. Found: C, 44.64; H, 2.42; N, 3.73%.

1-(4-Bromophenyl)-2,2-dichloro-3-(4-nitrophenyl)aziridine (2f): White solid; mp 141–143 °C; IR (KBr): ν/cm^{-1} 3080, 2924, 1600, 1522 (C=C, Ar); ^{1}H NMR ($CDCl_3$): δ 4.09 (s, 1H, HCN), 7.22–8.55 (m, 8H, Ar); ^{13}C NMR ($CDCl_3$): δ 53.9, 74.5, 121.2, 121.5, 123.7, 128.9, 132.3, 139.6, 143.3, 148.4; MS: m/z 392 (M + 6, 5), 390 (M + 4, 8), 388 (M + 2, 18), 386 (M^+, 10), 353 (100), 351 (89), 307 (94), 305 (85), 153 (90), 77 (60); Anal. Calcd for $C_{14}H_9BrCl_2N_2O_2$: C, 43.33; H, 2.34; N, 7.22%. Found: C, 43.43; H, 2.35; N, 7.24%.

2,2-Dichloro-1-(4-methylphenyl)-3-(4-nitrophenyl)aziridine (2g): Yellow solid; mp 140–142 °C; IR (KBr): ν/cm^{-1} 3090, 2918, 1589, 1490 (C=C, Ar); ^{1}H NMR ($CDCl_3$): δ 2.36 (s, 3H, CH_3), 3.89 (s, 1H, HCN), 6.59–8.31 (m, 8H, Ar); ^{13}C NMR ($CDCl_3$): δ 21.2, 53.5, 75.2, 119.6, 123.7, 128.9, 129.9, 134.6, 140.3, 141.7, 148.3; MS: m/z 326 (M + 4, 20), 324 (M + 2, 29), 322 (M^+, 40), 289 (90), 287 (100), 243 (60), 241 (80), 154 (70), 152 (82), 91 (92); Anal. Calcd for $C_{15}H_{12}Cl_2N_2O_2$: C, 55.74; H, 3.74; N, 8.67%. Found: C, 55.75; H, 3.74; N, 8.67%.

1-(4-Bromophenyl)-2,2-dichloro-3-(4-methylphenyl)aziridine (2h): Yellow solid; mp 146–148 °C; IR (KBr): ν/cm^{-1} 3100, 2898, 1600, 1500 (C=C, Ar); ^{1}H NMR ($CDCl_3$): δ 2.31 (s, 3H, CH_3), 3.75 (s, 1H, HCN), 7.04–7.48 (m, 8H, Ar); ^{13}C NMR ($CDCl_3$): δ 20.1, 55.1, 74.1, 117.9, 121.9, 128.4, 129.6, 132.1, 134.1, 138.2, 141.1; MS: m/z 362 (M + 6, <2), 360 (M + 4, 10), 358 (M + 2, 27), 356 (M^+, 15), 323 (100), 321 (89), 234 (84), 232 (70), 91 (97); Anal. Calcd for $C_{15}H_{12}BrCl_2N$: C, 50.46; H, 3.39; N, 3.92%. Found: C, 50.59; H, 3.39; N, 3.94%.

2,2-Dichloro-3-(4-chlorophenyl)-1-(4-methylphenyl)aziridine (2i): White solid, mp 128–130 °C; IR (KBr): ν/cm^{-1} 3090, 2920, 1600, 1508 (C=C, Ar); ^{1}H NMR ($CDCl_3$): δ 2.35 (s, 3H, CH_3), 3.65 (s, 1H, HCN), 6.94–7.47 (m, 8H, Ar); ^{13}C NMR ($CDCl_3$): δ 22.1, 54.2, 76.1, 118.1, 121.5, 128.9, 129.6, 132.1, 134.1, 138.1, 144.1; MS: m/z 318 (M + 6, <2), 316 (M + 4, 7), 314 (M + 2, 12), 312 (M^+, 15), 279 (85), 277 (95), 242 (70), 154 (90), 152 (100), 91 (95); Anal. Calcd for $C_{15}H_{12}Cl_3N$: C, 57.62; H, 3.87; N, 4.48%. Found: C, 57.64; H, 3.88; N, 4.49%.

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