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An explanation for the apparent *cis*-aziridine selectivity in the iron Lewis acid-catalyzed reaction of *N*-benzylidene aniline and ethyl diazoacetate

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Dedicated to Professor Myron Rosenblum on the occasion of his 75th birthday

Abstract

The reason for the apparent *cis*-aziridine selectivity in the reaction of ethyl diazoacetate with *N*-benzylidene aniline, catalyzed by $[(\eta^5\text{-}C_5H_5)(CO)_2Fe(THF)]^+[BF_4]^-$, is reported. The catalytic reaction produces both *cis* and *trans*-aziridines. Once the imine has been consumed, the *trans*-isomer is shown to undergo a catalytic decomposition, leaving *cis*-aziridine and by-products. The reaction is graphically profiled to illustrate the relative quantity of reactants and products as a function of time. A new mechanistic model is proposed in order to explain the observed selectivity. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Aziridines; Ethyl diazoacetate; Imines; Iron Lewis acid

1. Introduction

The catalytic synthesis of aziridines, specifically from imines and diazo compounds, has been significantly examined in recent years [1]. This particular catalytic approach is quite atom-economical since the only necessary by-product is low-molecular-weight, innocuous nitrogen [2]. Using this approach, several asymmetric attempts have been reported [3], and even accomplished with excellent enantioselectivities [4], but only one outstanding asymmetric report has really surfaced [5]. For these reasons, in addition to the versatile usefulness of aziridines as synthetic intermediates [6], we have been interested in this route. Previously, we have disclosed such a catalytic synthesis using an achiral iron Lewis acid–THF adduct, $[(\eta^5\text{-}C_5H_5)(CO)_2Fe(THF)]^+[BF_4]^-$ (**1**) [7]. The reaction was generally *cis*-aziridine selective. With this approach, *cis*-selectivity is now known to be typical, since most catalytic reactions mainly yield *cis*-aziridine [1a,c,e,f,i,n,3b,5]. Curiously, a couple of catalysts promote *trans*-aziridines [1h,j,k] while others produce varying mixtures of *cis* and *trans*-aziridines [1b,g,3a,c,d,e,f,4]. Recently, two reports demonstrated that no catalyst is required to achieve high *cis*-aziridine selectivity [8]. A fair amount of data has now been generated, yet, due to multiple variables, comparisons are somewhat hampered. In effect, there has generally not been much of a satisfactory explanation for the observed selectivities [9]. Actually, a quote in the most recent paper in this area [8a] states, "Reactions of diazo compounds with imines in the presence of metal catalysts usually occur with high *cis*-selectivity, but the origin of this selectivity has not been satisfactorily explained." Herein, we report the reason for the observed *cis*-aziridine selectivity (previously unreported for any Lewis acid catalyst) in the reaction of *N*-benzylidene aniline (**2**) with ethyl diazoacetate (**3**) catalyzed by **1**.

2. Results and discussion

In our previously reported procedure [7], we added **3** by a syringe pump for over 18 h to a solution of the imine (**2**) and 10 mol% of **1**. This was done in order to

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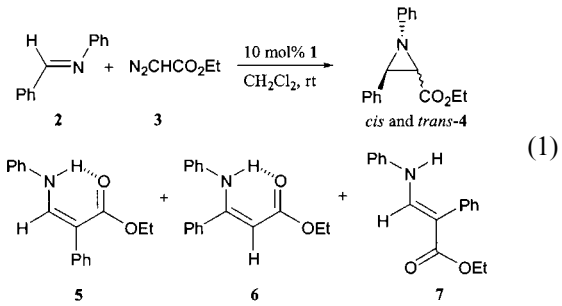
maintain a relatively low concentration of **3**, and thus reduce the formation of diethyl maleate and fumarate from iron carbenoids formed in situ [10]. Typically, the reaction was stirred for a total of 18 h and produced only the *cis*-aziridine product from most aryl imines. Since our initial report, we have observed that when this reaction is stopped immediately after the slow addition of **3**, the peaks due to the *trans*-aziridine product can also be seen in the crude ^{1}H-NMR spectrum. Furthermore, the *trans*-aziridine can be isolated. We have also discovered that the observed *cis*/*trans*-aziridine ratio is a function of time.

Now, we have found that **3** can be added all at once to an imine-catalyst solution, resulting in a desired reaction that is over within minutes as determined by the visual cessation of nitrogen evolution and by ^{1}H-NMR. This experimental improvement eliminates the need for excess **3** and reduces the time (18 h) and equipment (syringe pump) constraint. The reaction was worked up after stirring for 15 min and found to contain no starting reagents, but only the *cis* and *trans*-aziridines (**4**) in a 3:1 ratio plus the β-amino-α,β-unsaturated ester (AUE) by-products (**5**, **6**, and **7**; Eq. (1)). Surprisingly, this quick reaction produced no diethyl maleate or fumarate. This finding further weakens a hypothesis favoring a mechanism involving an iron–carbenoid intermediate in this aziridine formation [7,10,11].



In another reaction under the same conditions, the crude was worked up after stirring for 2 h and found to contain *cis*, but no *trans*-aziridine. The only significant difference in the crude ^{1}H-NMR spectrum was that the peaks for *trans*-aziridine were remarkably absent. Therefore, using NMR we attempted to monitor a reaction over 2 h, sampling the reaction every 5 min [12]. It was determined that a shorter monitoring window and smaller sampling intervals would provide more useful information.

A reaction was stirred for 30 min and sampled every minute for the first 15 min and then at increased intervals (Fig. 1). Both *cis* and *trans*-aziridines were formed in addition to the AUE by-products. The final yield of *cis*-aziridine ($\sim$40%) matches that of a standard reaction, regardless of whether it is run in 30 min or over 18 h using a slow addition of **3**. Until 10 min,

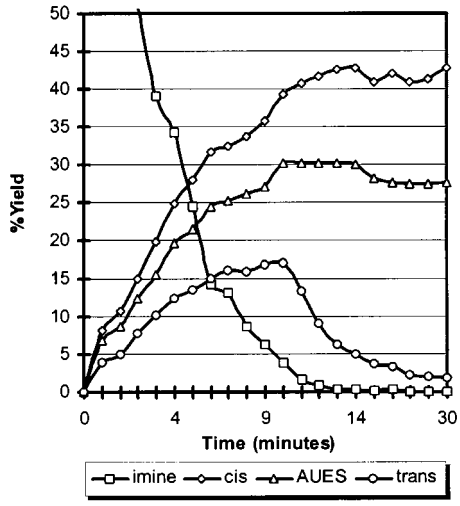
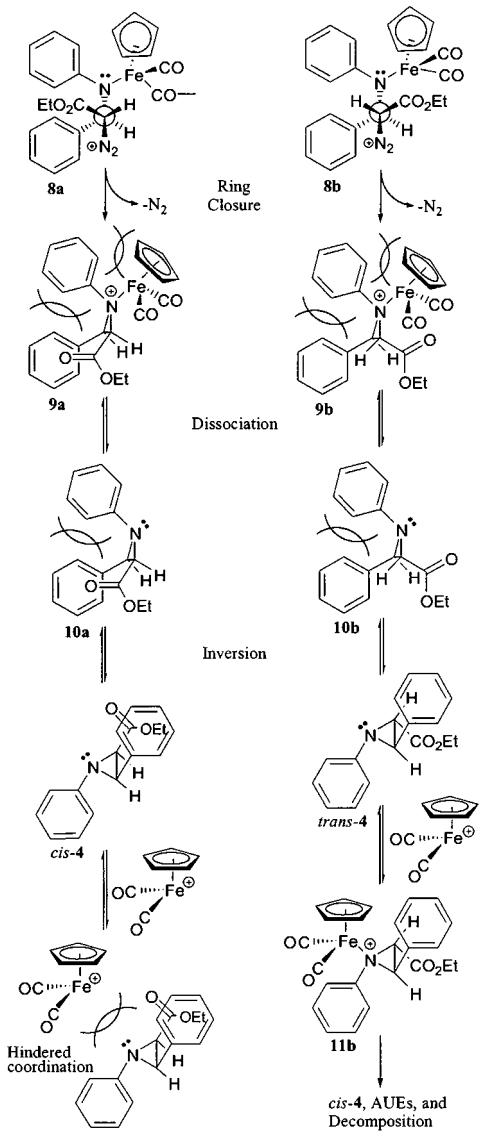


Fig. 1. The reactant and product profile as a function of time. The reaction was performed as usual in CH_2Cl_2 at room temperature. The graph illustrates raw data. The % yields are based upon the integration of a peak from each compound against the $-CH_3$ peak of the internal standard acetophenone in the ^{1}H-NMR spectra of the chronological reaction extracts. The % yields for **3** are not shown due to inconsistencies owing to its volatility during work-up. The curve for the AUEs represents the combined % yields of the three AUE by-products (**5**, **6**, and **7**) shown in Eq. (1).

the combined yield of the imine, aziridines, and the AUEs was roughly equal to 90%. The remaining 10%, being nitrogenous compounds, was assumed to be coordinated to the iron Lewis acid and removed during work-up before quantification by ^{1}H-NMR. The imine was mainly consumed after 10 min when the yield of *trans*-aziridine was non-coincidentally at a maximum. Once the imine was nearly consumed, it appeared that the *trans*-aziridine was catalytically isomerized or decomposed.

Pure *cis* and *trans*-aziridines were prepared and isolated for use in control reactions. The pure *cis*-aziridine (**4**) was stirred with 10 mol% of **1** and sampled over 48 h. Insignificant decomposition occurred after 12 h. After 24 h, the baseline in the ^{1}H-NMR spectrum showed some minor disturbances but *cis*-aziridine was mainly unaffected. After 48 h, most of the *cis*-aziridine still remained but a considerable amount had decomposed to a variety of unidentified compounds. These results demonstrate a fair compatibility of the *cis*-aziridine with the iron Lewis acid. They also show that, under the reaction conditions, *cis*-aziridine is not in equilibrium with any of the previously mentioned isomers (*trans*-**4**, **5**, **6**, and **7**) [13].

In a separate reaction, the pure *trans*-aziridine (**4**) was stirred with 10 mol% of **1** and sampled over 90 min. During this time period, *trans*-aziridine was found to undergo nearly complete reaction. The crude included unreacted *trans* and *cis*-aziridine, AUE by-products, and other unidentified by-products most likely



Scheme 1.

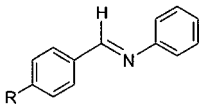
including polymer. This result could have important ramifications in an asymmetric reaction because there are at least two pathways to *cis*-aziridine and they may not involve a common intermediate. Thus, we have found that the catalytic reaction is not *cis*-aziridine selective. Rather, it produces both *cis* and *trans*-aziridines, and the *trans*-isomer undergoes a catalytic decomposition to form *cis*-aziridine and by-products.

In order to see if this phenomenon was unique to the iron Lewis acid, we attempted the same reaction employing the commonly used Lewis acid, $BF_3{\cdot}OEt_2$. One drop of $BF_3{\cdot}OEt_2$ was added to a CH_2Cl_2 solution of the pure *trans*-aziridine, instantly turning the solution dark-yellow from very pale-yellow. The solution was worked up and found to contain no aziridines; instead, a variety of unidentified by-products were formed. However, a trace of the AUE by-products was formed as deduced from the small characteristic doublet at 10.4 ppm in the baseline of the ^{1}H-NMR spectrum. From this one example, it appears that selective Lewis acid-catalyzed decomposition may be at work in other Lewis acid-catalyzed aziridine formations.

The following line of reasoning is proposed in order to explain this phenomenon (Scheme 1). Most evidence points to the reactant, *N*-benzylidene aniline (**2**), existing with the phenyl groups in an *anti* conformation in solution [14]. However, recent evidence demonstrates that *N*-benzylidene anilines are found to produce adducts with Lewis acids where the aryl groups exist in a *syn* conformation, in order to accommodate the steric demand of the acceptor moiety [15]. Assuming this is the case, then nucleophilic attack by either face of the diazo compound onto the activated imine results in the formation of both transient intermediates, **8a** and **8b**. Attack by these two faces of the diazo compound onto the opposite face of the activated imine leads to the enantiomers of the intermediates **8a** and **8b**. **8a** leads to the *cis*-aziridinium ion (**9a**), while **8b** leads to the *trans*-aziridinium ion (**9b**); **8a** will not lead to **9b** by rotation about the newly formed carbon–carbon bond. On the other hand, both **8a** and **8b** lead to the by-products, **5** and **6**, by 1,2 migration of the Ph or H moiety. Due to several unfavorable steric interactions, dissociation of the iron Lewis acid–aziridine complexes (**9a** and **9b**) is able to provide free aziridines (**10a** and **10b**). In the presence of excess competing base, like the remaining unreacted imine (**2**), aziridine re-coordination would not be anticipated. After dissociation of the iron Lewis acid–aziridine complexes (**9a** and **9b**), the resulting *cis* and *trans*-aziridines (**10a** and **10b**) would undergo inversion of the configuration at the nitrogen atom in the ring. Inversion alleviates the strain due to both phenyl groups occupying the same side of the aziridine ring, and thereby, produces the favored invertomers, *cis* and *trans*-**4** [16]. Simple molecular models show that these resulting *cis* and *trans*-aziridines (**4**) are both bulky. However, the *cis*-isomer has a more crowded face on the side of the aziridine ring possessing the N-nonbonding pair of electrons. The greater reactivity of *trans*-aziridine to the iron Lewis acid is likely due, partly, to a less hindered access to the N-lone pair of electrons. When the iron Lewis acid is coordinated, the fate of the aziridinium ion (**11b**) heavily relies on the electronic nature of the aryl group on the aziridine ring carbon [17]. It is helpful here to cite the information in Table 1 to illustrate this point [7].

The results in Table 1 are similar to the results of other Lewis acid-catalyzed reactions of aryl imines and **3**, but there are notable exceptions to this trend. Entry 2 of this table shows typical results for the unsubstituted aryl imine, where the *trans*-aziridine catalytically decomposed. The reaction of the imine listed in entry 1 led to a very complex product mixture where even

Table 1
Effect of the aryl group on *cis*/*trans* selectivity and yield of aziridine



Entry	Imine	*cis*/*trans*	Yield (%) [a]
1	R = OCH_3	–	0
2	R = H	All *cis*	40
3	R = NO_2	4:1	78

[a] Isolated total aziridine yield from the reaction of **3** with the indicated imine catalyzed by the iron Lewis acid over 18 h in CH_2Cl_2 at room temperature.

cis-aziridine was not obtained [18]. Fig. 2 illustrates how the electron-donating methoxy group can stabilize an intermediate from which a variety of isomerizations and decompositions can take place. From this intermediate aziridine formation can re-occur; either *cis* or the *trans*-aziridine may be formed. In contrast, entry 3 shows that even *trans*-aziridine is stable enough to be isolated from an 18 h reaction catalyzed by the iron Lewis acid. The electron-withdrawing nitro group has the opposite effect, destabilizing such an analogous intermediate. This is despite the *trans*-aziridine being less sterically hindered from re-coordination to the iron Lewis acid than *cis*-aziridine.

3. Conclusions

In conclusion, we have shown that the catalytic reaction produces both *cis* and *trans*-aziridines. The *trans*-isomer simply undergoes a catalytic decomposition, after the imine has been consumed, to form *cis*-aziridine and various by-products. In other reactions of this type where different sets of reagents are being utilized to produce predominantly *cis*-aziridines in moderate yields, analogous phenomena may be underlying the observed results. These observations may need to be considered in the development of asymmetric variations of this reaction.

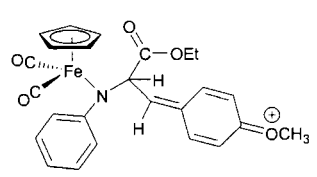


Fig. 2.

4. Experimental [19]

4.1. General considerations

All reactions were performed under a dry nitrogen atmosphere using standard Schlenk techniques. All reaction vessels were flamed under vacuum and filled with nitrogen prior to use. HPLC-grade CH_2Cl_2 (EM Science) was distilled under nitrogen from P_2O_5. Reagent grade-THF (EM Science) was distilled under nitrogen from the sodium–benzophenone ketyl. Technical-grade pentane (Phillips) was mixed with concentrated H_2SO_4, washed first with $NaHCO_3$, and then with distilled water, dried over Na_2SO_4, and distilled from sodium. HPLC-grade EtOAc (Spectrum) was dried over 4A molecular sieves prior to use. Untreated reagent-grade hexanes (EM Science) were also used for column chromatography. Flash chromatography was performed using EM Science silica gel 60, 70–230 mesh. ^{1}H-NMR spectrometry was performed using a 250 MHz or a 300 MHz Bruker spectrometer. The iron Lewis acid–THF adduct, $[(\eta^5\text{-}C_5H_5)Fe(CO)_2(THF)]^+[BF_4]^-$ (**1**), was synthesized as reported by our group [20]. All other reagents were commercially available and used as received.

4.2. cis and trans-2-Ethoxycarbonyl-1,3-diphenylaziridine (4)—new fast procedure

A 167 mg (0.50 mmol) sample of iron Lewis acid (**1**) was dissolved in 20 ml CH_2Cl_2 under nitrogen. To this solution, 920 mg (5.0 mmol) of imine (**2**) was added and the mixture was stirred for 10 min. Ethyl diazoacetate (**3**) (0.58 ml, 5.0 mmol) was added all at once to the catalyst–imine solution and the reaction mixture was stirred for an additional 10 min. The reaction was stopped by the addition of 5 ml THF. The mixture was immediately passed through a silica gel plug, and the organic products were eluted with CH_2Cl_2. The solvent was removed under reduced pressure to a pale-yellow liquid. ^{1}H-NMR ($CDCl_3$) of the crude showed **4**, *cis*–*trans* ratio of roughly 2:1. The products were separated on a silica gel column using EtOAc (0–6%) in pentane in the following order: AUEs **5** and **6**, *trans*-**4** (20%, 270 mg), *cis*-**4** (40%, 540 mg), and AUE **7**.

4.3. Reaction monitored over 30 min

A 10 mol% sample of Lewis acid (**1**) (109 mg, 0.32 mmol) was dissolved in 20 ml CH_2Cl_2 under nitrogen. One equivalent of imine (**2**) (600 mg, 3.2 mmol), in 5 ml of CH_2Cl_2 was added to the catalyst solution. The solution was stirred at room temperature (r.t.) for 25 min and ethyl diazoacetate (**3**) (0.379 ml, 3.2 mmol) was added in one portion to the reaction mixture. The solution immediately began to evolve bubbles of nitro-

gen. After 30 s, 400 μl of solution was removed and passed through a silica gel plug. The organic compounds were rinsed through with CH_2Cl_2. The solvent was removed under reduced pressure to provide a pale-yellow liquid that was stored under nitrogen. The reaction was additionally sampled in this way after every minute for 15 min, and again after 17, 20, 25, and 30 min. The ^{1}H-NMR spectrum of each remaining extract was obtained. Clear peaks for **2**, *cis*-**4**, *trans*-**4**, **5**, **6**, and **7** were integrated against the peak for the internal standard acetophenone (2.6 ppm, 5.77 mM in $CDCl_3$).

4.4. *cis*-Aziridine (**4**) with **1**—control reaction

A 262 mg (0.98 mmol) sample of *cis*-**4** was dissolved in 5 ml CH_2Cl_2. To this solution, a 32 mg (0.095 mmol) sample of iron Lewis acid (**1**) was added and the solution was stirred for 1 h at r.t. A 250 μl solution was removed and passed through a silica gel plug. The organic compounds were rinsed through with CH_2Cl_2 and the solvent removed under reduced pressure. The ^{1}H-NMR spectrum was obtained from the remaining pale-yellow liquid in $CDCl_3$ to monitor the extent of reaction. The reaction was additionally sampled and monitored in this way after 14, 24, and 50 h. An additional 4 ml of CH_2Cl_2 was added over 50 h in order to make up for the solvent lost due to slow evaporation.

4.5. *trans*-Aziridine (**4**) with **1**—control reaction

A 203 mg (0.74 mmol) sample of *trans*-**4** was dissolved in 8 ml CH_2Cl_2. To this solution, 25 mg (0.074 mmol) of iron Lewis acid (**1**) was added. The reaction mixture was allowed to stir for 1 h and a 350 μl solution was removed and processed as described above. The ^{1}H-NMR spectrum showed significant decomposition, but also showed a notable amount of the remaining *trans*-**4**. The mixture was stirred for a total of 90 min, 3 ml THF was added, and the solution was worked up as previously described. The ^{1}H-NMR spectrum clearly showed *cis*-**4**, the AUE by-product (**5**), and the unreacted *trans*-**4**.

5. Supplementary material

^{1}H-NMR spectra of the progress of the control reactions and further details for the reaction by which Fig. 1 was obtained plus ^{1}H-NMR spectral data with integrals may be obtained from the authors upon request.

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