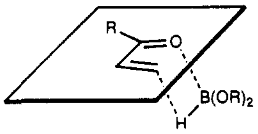
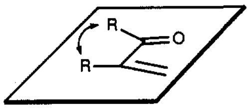
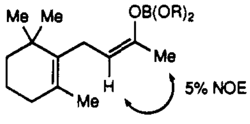


3

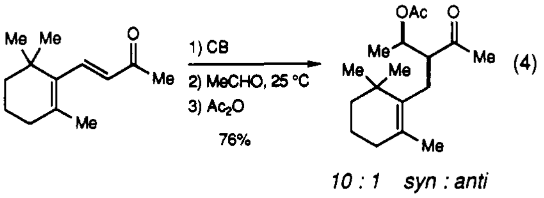


4

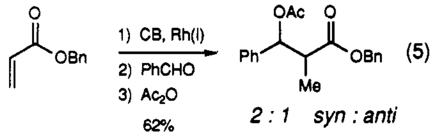
the reduction of β-ionone generates a single enolate,[15] which NOE experiments establish is indeed the Z isomer **5**. This enolate may be functionalized by reaction with electrophiles. For example, its aldol reaction with acetaldehyde affords the syn product with good selectivity (eq 4).



5



The boron enolate derived from the rhodium-catalyzed conjugate reduction of benzyl acrylate also appears to be a single isomer by ^{1}H NMR analysis, presumably the Z isomer.[16] Unexpectedly, the reaction of this species with benzaldehyde affords the syn aldol adduct with low stereoselectivity (syn:anti = 2:1) (eq 5).[17]



The rhodium-catalyzed and the uncatalyzed 1,4-addition reactions of catecholborane described herein comprise a mild and convenient method for effecting the conjugate reduction of α,β-unsaturated carbonyl compounds. Studies further defining the scope and the mechanism of these reactions are in progress.

Acknowledgment. Support has been provided by the NIH and Merck. An NSF Predoctoral Fellowship to G. C.F. is gratefully recognized. The NIH BRS Shared Instrumentation Program 1 S10 RR01748-01A1 is acknowledged for providing NMR facilities.

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[15] By the limits of detection by ^{1}H NMR spectroscopy.

[16] The rhodium-catalyzed hydrosilation of α,β-unsaturated esters has been reported to provide the Z ketene acetals with high selectivity (>98:2): Slougui, N.; Rousseau, G. *Synth. Commun.* **1987**, *17*, 1–11. However, the analogous reaction of α,β-unsaturated ketones affords a mixture of isomers: Ojima, I.; Kogure, T. *Organometallics* **1982**, *1*, 1390–1399.

[17] For examples of related condensations (α,β-unsaturated esters with ketones) which employ trimethylsilane as the reducing agent and $RhCl_3 \cdot 3H_2O$ as the catalyst, see: Revis, A.; Hilty, T. K. *Tetrahedron Lett.* **1987**, *28*, 4809–4812. Others have shown that the condensation of silyl enol ethers with aldehydes is catalyzed by Rh(I) complexes: Sato, S.; Matsuda, I.; Izumi, Y. *Tetrahedron Lett.* **1986**, *27*, 5517–5520. Reetz, M. T.; Vougioukas, A. E. *Tetrahedron Lett.* **1987**, *28*, 793–796.

α-Metalated Tertiary Enol Carbamates. New Acyl Anion Equivalents

Saumitra Sengupta and Victor Snieckus*

Guelph-Waterloo Centre for Graduate Work in Chemistry, University of Waterloo, Waterloo, Ontario, Canada N2L 3G1

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Summary: α-Metalated enol carbamate **1g** constitutes a new, conveniently generated (*sec*-BuLi/TMEDA/THF/ −78 °C), and well-behaved acyl anion equivalent. The utility and scope of this synthon for the preparation of a variety of useful synthetic building blocks (**4**, **6**, **8**, **11**, **12**) has been demonstrated.

First enunciated by Corey and Seebach in their seminal studies on metalated 1,3-dithianes,[1] the *umpolung* (polarity reversal) principle has become a powerful concept in synthetic design.[2] The subsequent discovery of α-lithiation of alkyl vinyl ethers[3] and alkyl vinyl sulfides[4] has allowed the recent emergence of a distinct group of α-metalated α-heteroatom substituted olefins **1** as valuable *umpolung* synthons.[5-7] Stripped to the simplest form, these constitute readily available acyl anion equivalents **2**, which are generally useful in direct functionalization

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[1] Corey, E. J.; Seebach, D. *Angew. Chem., Int. Ed. Engl.* **1965**, *4*, 1075.

[2] *Umpoled Synthons*; Hase, T. A., Ed.; Wiley-Interscience: New York, 1987.

[3] Baldwin, J. E.; Hofle, G. A.; Lever, O. W., Jr. *J. Am. Chem. Soc.* **1974**, *96*, 7125. Hartmann, J.; Stähle, M.; Schlosser, M. *Synthesis* **1974**, 888. Schöllkopf, U.; Haenssie, P. *Liebigs Ann. Chem.* **1972**, *763*, 208.

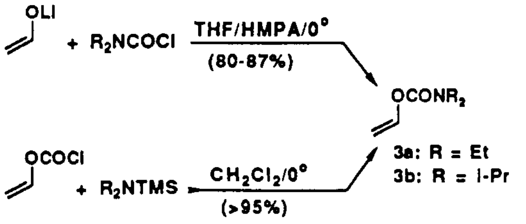
[4] Oshima, K.; Shimoji, K.; Takahashi, H.; Yamamoto, H.; Nozaki, H. *J. Am. Chem. Soc.* **1973**, *95*, 2694.

[5] Gschwend, H. W.; Rodriguez, H. R. *Org. React.* **1979**, *26*, 1.

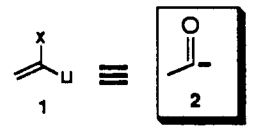
[6] (a) For a review, see: Ager, D. J. in ref 2. Recent work: (b) Soderquist, J. A.; Hsu, G. J.-H. *Organometallics* **1982**, *1*, 830. McDougal, P. G.; Rico, J. G. *J. Org. Chem.* **1987**, *52*, 4817. Smith, A. B., III; Fukui, M. *J. Am. Chem. Soc.* **1987**, *109*, 1269. Tamao, K.; Nakagawa, Y.; Arai, H.; Higuchi, N.; Ito, Y. *Ibid.* **1988**, *110*, 3712. Smith, A. B., III; Empfield, J. R.; Vaccaro, H. A. *Tetrahedron Lett.* **1989**, *30*, 7325. For a chiral version, see: McDougal, P. G.; Dicesare, J.; Kerrigan, J.; Starks, L. J. *Abstracts of Papers*; 199th National Meeting of the ACS, Boston, MA; American Chemical Society: Washington, DC, 1990; abstract no. ORGN 270. For an in situ generation of (2,2-difluoro-1-(tosyloxy)vinyl)lithium, see: Ichikawa, J.; Sonoda, T.; Kobayashi, H. *Tetrahedron Lett.* **1989**, *30*, 5437. (c) Trost, B. M.; Lavoie, A. C. *J. Am. Chem. Soc.* **1983**, *105*, 5075 and references cited therein. Fitt, J. J.; Gschwend, H. W. *J. Org. Chem.* **1979**, *44*, 303. McDougal, P. G.; Oh, Y.-I. *Tetrahedron Lett.* **1986**, *27*, 139. (d) McCombie, S. W.; Shankar, B. B.; Ganguly, A. K.; Padwa, A.; Bullock, W. H.; Dyszlewski, A. D. *Ibid.* **1987**, *28*, 4127 and references cited therein. For a formal 1-(tolylsulfinyl)vinyl α-anion, see: Cheng, H.-C.; Yan, T.-H. *Tetrahedron Lett.* **1990**, *31*, 673. (e) Krief, A. *Tetrahedron* **1980**, *36*, 2531. (f) Bates, R. B.; Beavers, W. A.; Blacksber, I. R. *Abstracts of Papers*; 169th National Meeting of the CS, Philadelphia, PA; American Chemical Society: Washington, DC, 1975. Boeckman, R. K., Jr.; Walters, M. A.; Koyano, H. *Tetrahedron Lett.* **1989**, *30*, 4787. Schmidt, R. R.; Speer, H. *Ibid.* **1981**, *22*, 4259. α-Metalated 1,2-dihydropyridines may be viewed as α-metalated enamine synthons, see: Comins, D. L.; Weglarz, M. A.; O'Connor, S. *Tetrahedron Lett.* **1988**, *29*, 1751. (g) Lau, K. S. Y.; Schlosser, M. *J. Org. Chem.* **1978**, *43*, 1595. Meyers, A. I.; Spohn, R. F. *Ibid.* **1985**, *50*, 4872.

0022-3263/90/1955-5680$02.50/0

Scheme I



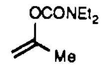
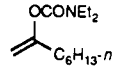
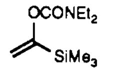
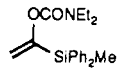
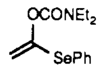
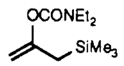
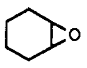
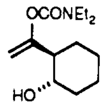
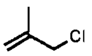
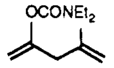
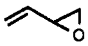
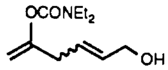
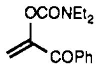
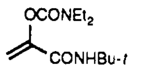
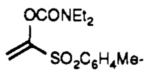
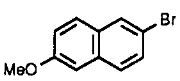
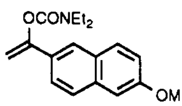
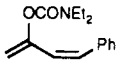
operations. The current synthetic prominence of the heteroatom-directed metalation principle in both aliphatic[5,8] and aromatic[9] chemistry and our continuing interest in aryl *O*-carbamate metalation[9a,10] stimulated the examination of α-metalated enol carbamates **1g**.[11] Herein we report our preliminary findings which demonstrate that synthon **1g** is an important addition to the armamentarium of acyl anion equivalents, offering complementarity and new versatility when compared to the conventional synthons **1a–b**.[12]



	a	b	c	d	e	f	g
X	$OR^{6a,b}$	$SR^{6a,c}$	SO_2R^{6d}	SeR^{6e}	NR_2^{6f}	$Hal^{5,6g}$	$OCONR_2^{\text{this work}}$

The prior art indicated that the preparation of enol carbamates is not a trivial task, especially by enolate chemistry.[13] In order to prepare the bare enol carbamate **3**, two different approaches were pursued (Scheme I). In the more direct approach, acetaldehyde lithium enolate[14] was O-carbamoylated to give the enol carbamates **3a** and **3b** in 80–87% yields on a multigram scale. In the alternate, potentially general approach, commercial vinyl chloroformate was treated with appropriate trimethylsilylamides to give the same enol carbamates in nearly quantitative yields.

Table I. Reaction of α-Lithio Enol Carbamate **1g** with Electrophiles

entry	electrophile	product (4)	yield, %
1	MeI		82
2	n-$C_6H_{13}I$		70
3	Me_3SiCl		85
4	$Ph_2MeSiCl$		76
5	PhSeSePh		54
6	Me_3SiCH_2OTf		56
7[a]			66
8[b]			62
9[c]			72[d]
10	PhCOCl		40
11	t-BuNCO		55
12	p-$MeC_6H_4SO_2F$		75
13[e]			71
14[f]			80

[a] 2 equiv of $BF_3 \cdot Et_2O$ was added prior to addition of the epoxide. [b] 1 equiv of CuI·2LiCl complex was used and alkylation carried out at 0 °C. [c] 1 equiv of CuCN·2LiCl complex was used and the alkylation carried out at 0 °C. [d] 75:25 mixture (by NMR) of *E*:*Z* isomers. [e] $ZnBr_2$ (1 equiv), $Pd(PPh_3)_4$ (5 mol %), THF, reflux, 4 h. [f] $ZnBr_2$ (1 equiv), $Pd(PPh_3)_4$ (5 mol %), THF, room temperature, 4 h.

Under standard metalation conditions (*sec*-BuLi/TMEDA/THF/–78 °C) following inverse addition protocol, **3a** undergoes smooth α-lithiation and the resulting α-lithio species **1g** may be trapped with a variety of electrophiles to give products **4** in good yields (Scheme II, Table I).[15] Primary alkyl iodides serve as effective reaction partners (entries 1, 2) and silicon and selenium electrophiles may be introduced cleanly (entries 3–5). The 2-oxyallylsilane (entry 6) constitutes a particularly uncommon product with promise as an annulation reagent.[16] Cyclohexene oxide, an otherwise stubborn electrophile,

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(7) In α-blocked α-heteroatom substituted vinyl derivatives, β-metalation and, if available, γ-metalation (allylic) takes place, see: (a) Vinyl ethers: McDougal, P. G.; Rico, J. G.; VanDerveer, D. *J. Org. Chem.* **1986**, *51*, 4492 and references therein; Cox, P.; Mahon, M. F.; Molloy, K. C.; Lister, S.; Gallagher, T. *Tetrahedron Lett.* **1988**, *29*, 1993. (b) Enamines: Stork, G.; Shiner, C. S.; Cheng, C.-W.; Polt, R. L. *J. Am. Chem. Soc.* **1986**, *108*, 304. Stork, G.; Polt, R. L.; Li, Y.; Houk, K. N. *Ibid.* **1988**, *110*, 8360.

(8) Klumpp, G. W. *Recl. Trav. Chim. Pays-Bas* **1986**, *105*, 1. Beak, P.; Meyers, A. I. *Acc. Chem. Res.* **1986**, *19*, 356.

(9) (a) Snieckus, V. *Chem. Rev.*, in press; (b) Snieckus, V. *Bull. Soc. Chim. Fr.* **1988**, 67. (c) Narasimhan, N. S.; Mali, R. S. *Top. Curr. Chem.* **1987**, *138*, 63. (d) Beak, P.; Snieckus, V. *Acc. Chem. Res.* **1982**, *15*, 306.

(10) Sibi, M. P.; Snieckus, V. *J. Org. Chem.* **1983**, *48*, 1935. Miah, M. A. J.; Snieckus, V. *J. Org. Chem.* **1985**, *50*, 5436.

(11) As comprehensively demonstrated by Hoppe and co-workers, α-anions of *allyl* carbamates are diversely useful synthetic intermediates, see: Kramer, T.; Schwark, J.-R.; Hoppe, D. *Tetrahedron Lett.* **1989**, *30*, 7037. Review: Hoppe, D. *Angew. Chem., Int. Ed. Engl.* **1984**, *23*, 932.

(12) (a) While this work was in progress, an isolated example of α-lithiation of an enol carbamate appeared: Kocienski, P.; Dixon, N. J. *Synlett* **1989**, 52. (b) A solitary case of α-lithiation of a thioenol carbamate is known: Hoppe, D.; Beckmann, L.; Follmann, R. *Angew. Chem., Int. Ed. Engl.* **1980**, *19*, 303.

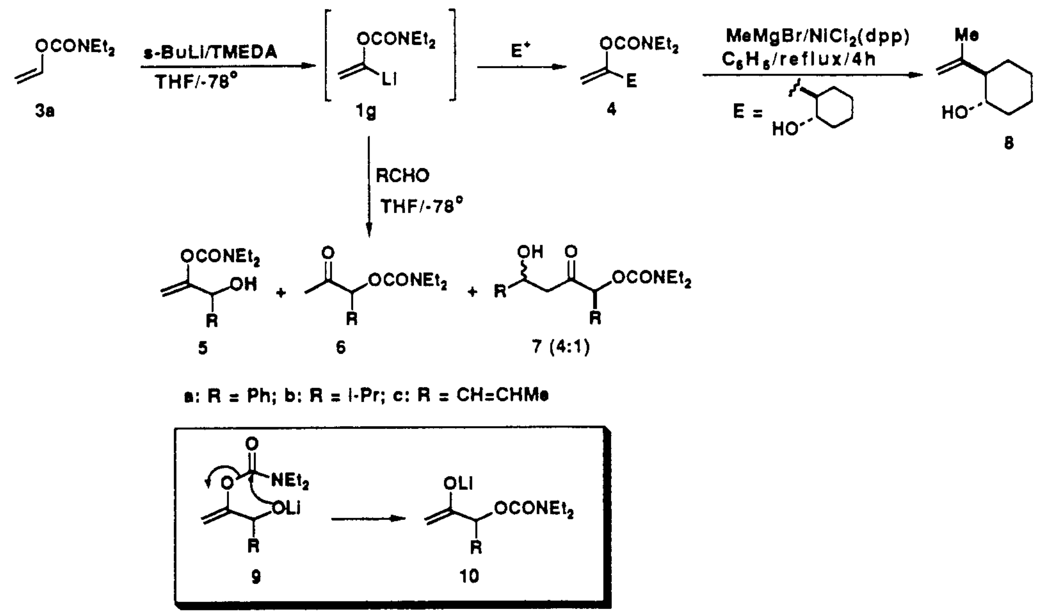
(13) Olofson, R. A.; Cuomo, J. *Tetrahedron Lett.* **1980**, *21*, 819. de Cusati, P. J.; Olofson, R. A. *Ibid.* **1990**, *31*, 1405. Mahe, R.; Sasaki, Y.; Bruneau, C.; Dixneuf, P. H. *J. Org. Chem.* **1989**, *54*, 1518.

(14) Generated by n-BuLi-mediated fragmentation of tetrahydrofuran: Jung, M. E.; Blum, R. L. *Tetrahedron Lett.* **1977**, 3791. Bates, R. B.; Kroposki, L. M.; Potter, D. E. *J. Org. Chem.* **1972**, *37*, 560. Oakes, F. T.; Yang, F.-A.; Sebastian, J. F. *J. Org. Chem.* **1982**, *47*, 3094.

(15) All new compounds show analytical and spectral (IR, NMR, MS) data in accord with the assigned structures. Typical procedure: A THF solution (3 mL) of **3a** (179 mg, 1.25 mmol) was added via cannula to a solution of s-BuLi (1.40 mL, 0.94 M, 1.31 mmol) and TMEDA (0.20 mL, 1.32 mmol) in THF (8 mL) at –78 °C. After 1 h at –78 °C, TMSCl (0.19 mL, 1.40 mmol) was added. It was quenched after 15 min with saturated NH_4Cl solution. Standard workup with ether followed by flash chromatography (20% ether in hexane) gave a clear liquid: 228 mg (85%); ^1H NMR ($CDCl_3$) δ 5.40 (d, 1 H, J = 1.2 Hz), 5.03 (d, 1 H, J = 1.2 Hz), 3.31 (q, 4 H, J = 7.1 Hz), 1.15 (t, 6 H, J = 7.1 Hz), 0.18 (s, 9 H); IR (neat) 1710, 1650 cm^{-1}.

(16) For a comprehensive review on allyl silanes, see: Fleming, I.; Dunoguès, J.; Smithers, R. *Org. React.* **1989**, *37*, 57.

Scheme II

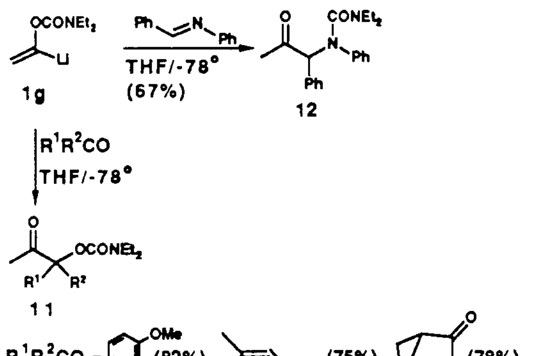


a: R = Ph; b: R = i-Pr; c: R = CH=CHMe

reacts only in the presence of $BF_3{\cdot}Et_2O$[17] whereas allylic electrophiles can be coupled with **1g** in the presence of Cu(I) salts to give S_N2' products (entries 8, 9). α-Benzoylation and α-amidation proceed in modest yields (entries 10, 11), and the products represent, together with the α-tosyl derivative (entry 12), members of push–pull olefins, an increasingly important class of compounds.[18] Furthermore, aryl and vinyl groups are efficiently introduced (entries 13, 14) by the in situ conversion of **1g** into the corresponding zinc species ($ZnBr_2$) and subsequent Pd-catalyzed cross coupling with aryl and vinyl bromides, respectively.[19] This cross coupling equivalent of the Friedel–Crafts acylation (entry 13) has potential general synthetic significance. Perhaps the most striking feature of the present methodology lies in the transformation **4** → **8**, where the *O*-carbamate function, after serving its role as a directed metalation group, now acts as a leaving group towards Ni(0)-catalyzed Grignard substitution.[20]

The reaction of α-lithio enol carbamate **1g** with aldehydes and ketones is markedly different from those observed with other α-heteroatom vinylmetallics. Thus the reaction with benzaldehyde (−78 °C, 30 min) afforded **6a** (55–60%) and **7a** (20–30%) as the major products together with minor amounts of **5a**. This product distribution can be rationalized in terms of a facile carbamoyl transfer in the initial adduct **9** leading to the enolate **10**,[21] which, upon further aldol condensation with benzaldehyde, gives rise to **7a**. This intramolecular carbamoyl transfer is extremely rapid and remains unaffected by variations in temperature and solvents as well as recourse to the bulkier enol carbamate **3b**. Transmetalation of **1g** with Et_2AlCl, $MgBr_2$, or $CeCl_3$ was not profitable since, in all these cases, the rate of the initial alkylation was greatly inhibited. However, short reaction times (5 s at −78 °C) were found to minimize the subsequent aldol condensation of **10**, and under these conditions, both aromatic and aliphatic aldehydes reacted with **1g** to give the *umpolung* adducts **6a–c** in 76, 65, and 72% yields, respectively; only minor amounts (10–12%) of the corresponding aldols (**7a–c**) were detected. The products **7a–c**, although an undesirable complication in the present context, were obtained uniformly with good diastereoselectivity (4:1), a feature which has not been previously observed in aldols derived from α′-oxymethyl ketones.[22]

Scheme III



Analogous carbamoyl transfer was also observed in reactions of **1g** with ketones leading to the acyloin carbamates **11** in 70–82% isolated yields (Scheme III). In these cases, an excess (1.5 equiv) of ketone was required to obtain good yields. As a further indication of the scope of species

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[17] Eis, M. J.; Wrobel, J. E.; Ganem, B. *J. Am. Chem. Soc.* **1984**, *106*, 3693. Compare: Alexakis, A.; Hanaizi, J.; Jachiet, D.; Normant, J.-F.; Toupet, L. *Tetrahedron Lett.* **1990**, *31*, 1271.

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[21] Qualitatively, the carbamoyl transfer (**9** → **10**) is faster than the reaction of **1g** with PhCHO at −78 °C, since formation of **6a** cannot be suppressed even when less than 1 equiv of PhCHO is used with or without $BF_3{\cdot}Et_2O$. For a similar carbamoyl transfer in thioenol carbamates, see ref 12b.

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1g, reaction with an aldimine under similar conditions afforded **12** and constitutes a new, potentially general, method for the preparation of α-amino methyl ketone derivatives.[23]

In summary, α-metalated tertiary enol carbamates **1g** are new, conveniently generated species with potential general synthetic use as acyl anion equivalents. Based on the preliminary results, broad scope for the preparation of α-hydroxy and α-amino methyl ketone building blocks and application in sequential *unpolung* and normal reactivity sequences (**7**) may be anticipated.[24]

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Articles

Ab Initio and Semiempirical Calculations on the Tautomeric Equilibria of N-Unsubstituted and N-Substituted Benzotriazoles

Alan R. Katritzky* and Konstantina Yannakopoulou

Department of Chemistry, University of Florida, Gainesville, Florida 32611-2046

Ernst Anders*

Institut für Organische Chemie der Universität Erlangen-Nürnberg, Henkestrasse 42, D-8520 Erlangen, FRG

John Stevens*

3M Corporation, St. Paul, Minnesota 55144

Miroslaw Szafran*

Department of Chemistry, Adam Mickieweicz University, ul. Grundwaldzka 6, 60-780 Poznan, Poland

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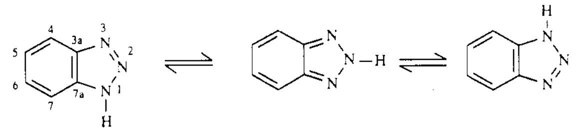
The geometries, relative stabilities, ionization potentials, and dipole moments for benzotriazole tautomers and their (dimethylamino)methyl derivatives were calculated by PM3, AM1, and MNDO semiempirical methods with full geometry optimization and with an ab initio 3-21G basis set. The geometries optimized by semiempirical methods are comparable with those obtained with partial optimization ab initio (6-31G and 3-21G levels) and available crystallographic data. Ab initio and semiempirical calculations failed to reproduce the N2–N3 bond length in 1*H*-benzotriazole. The X-ray dimensions of compound **4**, which due to its structural and electronic properties can be considered as a model compound for the 1-[(dimethylamino)methyl]benzotriazole **2a**, indicate that the "small" 3-21G basis set predicts bond lengths for this 1-substituted derivative, which are close to experimental data. The PM3 method gives ΔH_f in agreement with ab initio calculations, but both the AM1 and the MNDO methods do not. For benzotriazole, both semiempirical and ab initio calculations predict a large energy preference of the 1*H* over the 2*H* form. For the *N*-[(dimethylamino)methyl] derivatives, the ab initio results correctly predict an almost equal stability of the two forms (**2a** and **2b**) but the semiempirical methods fail. The influence of the fused benzo ring, together with the electronic properties of the N substituents, determines the relative stabilities of 1- and 2-substituted benzotriazoles.

Introduction

The tautomeric equilibrium of unsubstituted benzotriazole has been studied extensively and has been summarized in several reviews.[1,2] All indications are that the 1*H* form **1a** (**1c**) predominates strongly under all conditions studied. Thus, in the crystalline state the sole existence of form **1a** is demonstrated by X-ray studies[3] and also by ^{13}C NMR.[4] In solution, comparisons of the ultraviolet spectra of benzotriazole with those of the 1- and 2-methyl derivatives demonstrate that **1a** dominates,[5] and infrared studies agree.[6] Proton NMR comparisons with the *N*-methyl derivatives[7] and a variable-temperature

Scheme I



1a 1b 1c = 1a

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