

Figure 3. Eyring plot for combined CLS and MT data.

between the phenyls. Simple molecular mechanics calculations¹⁴ suggest that the effect should be worth about 4 kcal/mol, in good agreement with observation. The value of ΔS^\ddagger is also surprisingly low compared to that of the parent system (**1**). We originally thought that this could indicate the triplet **4** (³**4**) is involved. Such reasoning appeared to be supported by the unusually long lifetimes reported for ³**4** under both cryogenic, matrix-isolation,⁸ and ambient temperature, fluid-media⁹ conditions. However, a survey of the literature reveals a *general* phenomenon wherein diphenyl substitution greatly lowers ΔS^\ddagger values for bond cleavages.¹⁵ There is thus no reason to invoke ³**4**, and we assume that the bridge-flip process is spin conservative.

It seems likely that ¹**4** does lie in a potential energy well, and we would estimate its depth at ≤ 5 kcal/mol.¹⁶ Thus, the BDE of **3** (the enthalpy difference between **3** and ¹**4**) is 7–12 kcal/mol, in excellent agreement with the 10.9-kcal/mol value determined by Adam and Wirz.¹⁸

Typically, highly strained molecules are expected to be relatively unstable. Notable exceptions include structures such as cubane¹⁹ and [1.1.1]propellane,²⁰ for which no one bond homolysis can produce a substantial release of strain. These structures contain no exceptionally weak bonds, by a BDE criterion. Diphenylbicyclopentane is very strained, and the C1–C4 bond is very weak. At room temperature the bond is breaking at a rate greater than 1 s^{-1} , yet **3** is “stable”. This is because homolysis produces biradical **4**, which rapidly recloses to **3** before any “detectable” process such as dimerization or 1,2-hydrogen shift can occur.

Molecular oxygen does react with **3** in solution at room temperature. Qualitative kinetic studies show that bond cleavage alone is not rate limiting, i.e., that every ring opening does not lead to trapping. A more detailed analysis of the O₂-trapping reaction

(14) Steric repulsions between the phenyls were estimated by a homodesmotic reaction, in which **1** plus **3** were converted to two molecules of 1-phenylbicyclo[2.1.0]pentane. ΔH° for this process was calculated to be -4.1 kcal/mol by using MACROMODEL and the standard MM2 force field.

(15) For example, placement of phenyl groups on each end of the cleaving bond of cyclopropane or cyclobutane decreases ΔS^\ddagger of stereomutation by 24 (Rodewald, L. B.; DePuy, C. H. *Tetrahedron Lett.* **1964**, 5, 2951–2953) and 12 eu (Jones, G., II; Chow, V. L. *J. Org. Chem.* **1974**, 39, 1447–1448), respectively. For other homolytic bond cleavage reactions that exhibit similar behavior, see: Engel, P. S. *Chem. Rev.* **1980**, 80, 99–150; and ref 11.

(16) We have previously presented arguments¹⁷ that singlet 1,3-biradicals with delocalizing substituents will most likely lie in a well. The 5 kcal/mol upper limit derives from two observations. First, our qualitative observations⁸ on the matrix decay of ³**4** suggest a barrier on the order of 4 kcal/mol, and since the singlet–triplet gap is certainly small,¹⁷ the ¹**4** barrier cannot be much larger. Second, if the barrier is > 5 kcal/mol, ¹**4** should be observable by flash photolysis, but it is not.⁹

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is presented in the accompanying manuscript by Adam and Wirz.¹⁸ Their data, combined with that presented here, result in a remarkably detailed model of the kinetic and thermodynamic relationships among **3**, ¹**4**, and ³**4**.

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Oxygen Trapping and Thermochemistry of a Hydrocarbon Singlet Biradical: 1,3-Diphenylcyclopentane-1,3-diyl

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Our knowledge of absolute rate constants for the reactions of triplet biradical intermediates in solution is rapidly increasing and, as a result, useful structure–reactivity relationships are emerging.¹ In contrast, very little is known about singlet biradicals; recent ab initio calculations² for trimethylene and tetramethylene have substantiated the twixtyl hypothesis of a shallow, entropy-dominated free-energy minimum proposed 20 years ago by Hoffmann and co-workers.³ In particular, the question whether or not singlet 1,3-cyclopentadienyl represents a secondary minimum on the energy surface has been the subject of continuous debate.⁴ We now present evidence for diffusional trapping of singlet 1,3-diphenylcyclopentane-1,3-diyl (**S**).

Triplet 1,3-diphenylcyclopentane-1,3-diyl (**T**) is remarkably persistent. Its lifetime amounts to ca. 25 μs at room temperature in solution.⁵ Coms and Dougherty have observed the ESR spectrum of **T** with a half-life of hours at 77 K in 2-methyltetrahydrofuran glass; the ESR signal did not disappear upon cooling to 3.8 K, indicating that the biradical has a triplet ground state.⁶

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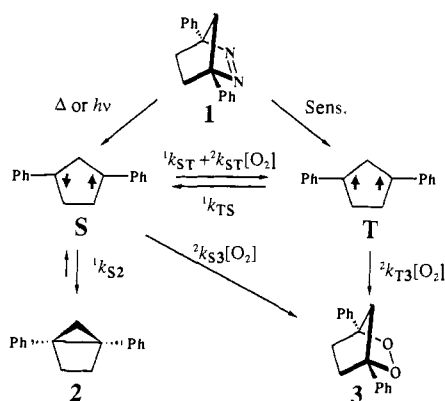
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Scheme I



Thermal or photochemical decomposition of 1,4-diphenyl-2,3-diazabicyclo[2.2.1]hept-2-ene (**1**) gave the hydrocarbon 1,4-diphenylbicyclo[2.1.0]pentane (**2**) which was stable, if acid and oxygen were rigorously excluded. Thermochemical estimates based on Benson's heat of formation increments had suggested that fission of the central bond of **2** might be almost thermoneutral.⁵ In fact, the concentration of paramagnetic **T** in equilibrium with **2** must be quite low, as an 80-MHz H NMR spectrum of **2** did not exhibit any line broadening. At temperatures above 60 °C, however, the endo and exo methylene signals broadened due to a degenerate ring-flip isomerization of **2**. From the coalescence temperature, $T_c \approx 354$ K, and the chemical shift difference, $\Delta\nu = 24$ s⁻¹, we estimate a free energy of activation $\Delta G^\ddagger(T_c) = (18.0 \pm 0.4)$ kcal mol⁻¹ for this process. This result is in good agreement with, but less precise than, the activation parameters reported in the accompanying paper by Coms and Dougherty.⁷ Due to the geometrical constraints of **2**, the degenerate ring-flip reaction must proceed via the biradical **S**, either as intermediate or as transition state.

When air was admitted, **2** was rapidly converted to the endoperoxide **3**.⁵ Attempts to determine the rate law for the autooxidation of **2** by monitoring H NMR signal intensities as a function of time gave only crude results and it was likely that the rate was partly limited by the access of air to the NMR tube. Therefore, we decided to monitor the rate of oxygen consumption in a sealed vessel containing excess **2**. The lifetime of triplet tetracene, τ_{Te1} , was used as an indicator for the concentration of triplet oxygen, [³O₂]. A degassed solution of **2** (1.1×10^{-2} M) and tetracene (ca. 2×10^{-4} M) in acetonitrile was briefly shaken under air and sealed, and τ_{Te1} was repeatedly determined by flash photolysis at intervals of 25 s ($\lambda_{exc} = 351$ nm, XeF excimer laser, $\lambda_{obs} = 460$ nm).⁸ During the first few minutes after preparation of the sample, the lifetime τ_{Te1} increased from 300 ns to >100 μ s. Subsequent analysis showed that τ_{Te1} increased exponentially with observation time t , as long as oxygen quenching was the predominant decay process of triplet tetracene (up to $\tau_{Te1} \approx 10$ μ s), i.e., $\ln(\tau_{Te1}/s) = k_{obs}t + \text{constant}$. Linear regression of $\ln(\tau_{Te1}/s)$ vs t gave the slope $k_{obs} = (2.1 \pm 0.2) \times 10^{-2}$ s⁻¹ as an average from two series of experiments at 24 ± 1 °C. A second series of experiments at lower concentrations of both **2** (3.3×10^{-3} M) and ³O₂ (ca. 4×10^{-4} M) gave $k_{obs} = (6.7 \pm 0.7) \times 10^{-3}$ s⁻¹. It follows that the depletion of [³O₂] via the reaction with **2** obeys the second-order rate law (eq 1), with [**2**] \approx constant under the

$$-d[{}^3\text{O}_2]/dt = {}^2k_{ox}[\mathbf{2}][{}^3\text{O}_2] \quad (1)$$

experimental conditions used. Analysis of the combined data according to eq 1 gave the second-order rate constant ${}^2k_{ox} = k_{obs}/[\mathbf{2}] = (2.1 \pm 0.2)$ M⁻¹ s⁻¹ as an average of four measurements

(7) Coms, F. D.; Dougherty, D. A. *J. Am. Chem. Soc.*, preceding paper in this issue.

(8) A priori estimates (Stevens, B.; Algar, B. E. *Chem. Phys. Lett.* **1967**, *1*, 58) as well as a blind test showed that any oxygen consumption attributable to tetracene-sensitized photoperoxidation was negligible under the experimental conditions used.

at 24 ± 1 °C. Further experiments at lower temperatures gave ${}^2k_{ox} = (7.2 \pm 0.8) \times 10^{-2}$ (-13 ± 1 °C) and $(5.0 \pm 0.8) \times 10^{-3}$ M⁻¹ s⁻¹ (-40 ± 2 °C).⁹

Two conclusions are drawn from the observation that the autooxidation rate law is first order in [³O₂]: (i) pathways involving a formation of **T** by spontaneous intersystem crossing (ISC) are eliminated from consideration and (ii) ISC is rate-determining in the spontaneous decay of **T** (this question was raised, but not answered, in our recent paper).⁵ From the direct observation of **T** by flash photolysis⁵ we know that oxygen trapping of **T** (${}^2k_{T3}[\text{}^3\text{O}_2] = 7.5 \times 10^9[\text{}^3\text{O}_2]$ s⁻¹) is faster than the intrinsic decay (4×10^4 s⁻¹) down to [³O₂] = 5×10^{-6} M. Therefore, any formation of **T** is irreversible in aerated solutions and the rate law for autooxidation by spontaneous ISC (i) would be zero-order in [³O₂]. As for (ii), the cyclization of **S** must be faster than the competing processes of ISC. If this were not so, the formation of **S** from **2** would also be irreversible in aerated solutions, the rate law for autooxidation would again be zero-order in [³O₂], and, from the rate of the ring-flip process (${}^1k \approx 2$ s⁻¹ at 297 K),⁷ one would predict that a 10⁻² M solution of **2** would consume air oxygen within <1 s. This is clearly inconsistent with the observations. Inefficient ISC from **S** also accounts for the fact that **T** was not observed by direct flash photolysis of the azo compound **1**.⁵

Although **S** appears to be too short-lived to undergo spontaneous ISC efficiently at room temperature, we nevertheless maintain that it is a real intermediate. A first indication for this comes from the fact that **T** is formed upon direct irradiation of **1** at lower temperatures; obviously, a thermally activated process (leading to **2**) competes with ISC (leading to **T**) at some stage in the photoelimination of nitrogen from **1**. We attribute this bifurcation to the singlet biradical intermediate **S**, i.e., ${}^1k_{S2}$ vs ${}^1k_{ST}$.

Second, it is hard to envision that the autooxidation of **2** takes place within minutes via direct addition of ³O₂ to **2**. However, a mechanism involving reaction of ³O₂ with the singlet biradical intermediate **S** (Scheme I) seems quite plausible from the following considerations. The rate constant for the ring-flip process amounts to about 2 s⁻¹ at room temperature, i.e., the equilibrium **2** \rightleftharpoons **S** is established rapidly on the time scale of autooxidation. A pre-equilibrium mechanism is consistent with the observed rate law (eq 1) and requires that ${}^2k_{ox} = ({}^2k_{S3} + {}^2k_{ST})K$, where $K = [\mathbf{S}]/[\mathbf{2}]$. It is known from many precedents that spin-allowed, weakly exothermic ISC is catalyzed by oxygen at an essentially diffusion-controlled rate.¹⁰ If we then assume that the reaction of oxygen with **S** is limited only by the rate of diffusion, ${}^2k_{diff} \approx 2.8 \times 10^{10}$ M⁻¹ s⁻¹,¹¹ we determine the equilibrium constant to be $K(297 \text{ K}) = {}^2k_{ox}/{}^2k_{diff} \approx 7.5 \times 10^{-11}$ and hence the Gibbs free energy for the reaction **2** \rightarrow **S** is $\Delta G^\circ(297 \text{ K}) = -RT \ln K \approx 13.7 \pm 0.1$ kcal mol⁻¹. Comparison with the Gibbs free energy of activation for the same reaction, $\Delta G^\ddagger(297 \text{ K}) = 16.7 \pm 0.1$ kcal mol⁻¹,¹² suggests that the singlet biradical is protected from cyclization by a free energy barrier of 3.0 ± 0.2 kcal mol⁻¹ and has a lifetime of ca. 20 ps. Similarly, an enthalpy difference between **S** and **2**, $\Delta H^\circ = (10.9 \pm 0.5)$ kcal mol⁻¹, is obtained from the slope of a van't Hoff plot of $\ln K$ vs $1/T$.¹¹ For comparison, the activation enthalpy ΔH^\ddagger for the ring-flip reaction amounts to 12.2 ± 0.6 kcal mol⁻¹.⁷ Note that the activation barriers for the cyclization of the singlet biradical **S**, $\Delta G^\ddagger(297 \text{ K}) = 3.0 \pm 0.2$

(9) Small corrections were applied to the concentrations in order to account for density changes of acetonitrile with temperature: *International Critical Tables*, Washburn, E. W., Ed.; McGraw Hill: New York, 1928; Vol. 3.

(10) Saltiel, J.; Atwater, B. W. In *Advances in Photochemistry*; Volman, D. H., Hammond, G. S., Gollnick, K., Eds.; Wiley: New York, 1988; Vol. 14, pp 1-90.

(11) Rate constants for the diffusion of oxygen in acetonitrile were calculated by the equation ${}^2k_{diff} = 8RT/(2000\eta)$ ¹⁴ using available data for the viscosity η of acetonitrile.¹⁵ These rate constants obeyed a linear relation $\log({}^2k_{diff}/\text{M}^{-1} \text{s}^{-1}) = 12.08 - 483/(T/\text{K})$ which was used to determine $K = {}^2k_{ox}/{}^2k_{diff}$ as a function of temperature.

(12) Extrapolation of the activation parameters for the ring inversion⁷ gives $\Delta G^\ddagger(297 \text{ K}) = 17.1 \pm 0.1$ kcal mol⁻¹. If the biradical **S** is a true intermediate, a transmission coefficient of 0.5 must be attributed to the ring-flip process. Hence the free energy of activation for the formation of **S** is reduced by $RT \ln 2 = 0.4$ kcal mol⁻¹ to $\Delta G^\ddagger(297 \text{ K}) = 16.7$ kcal mol⁻¹.

kcal mol⁻¹ and $\Delta H^\ddagger = 1.3 \pm 1.0$ kcal mol⁻¹, should be considered as lower limits inasmuch as the value assumed for oxygen trapping of **S** represents the upper limit of diffusion control.¹³

In conclusion, we rationalize our observations in terms of a metastable singlet 1,3-biradical intermediate **S** which is accom-

modated in a potential energy trough of several kcal mol⁻¹. At room temperature, ISC of the singlet biradical **S** is inefficient but is mediated through diffusional encounters with oxygen in aerated solution. At 77 K, spontaneous ISC of **S** (formed by direct photolysis of **1**) to **T** is predominant.

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(13) A direct addition of ³O₂ to **2** would of course also follow a rate law of the form given by eq 1 and thus obviate the necessity to postulate trapping of **S**, but the high value of the rate constant would be quite unprecedented, and its consistency with the ring-flip kinetics would represent an amazing coincidence.

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Book Reviews*

Basic Solid State Chemistry. By Anthony R. West (University of Aberdeen). John Wiley and Sons: New York, 1984, 1988. x + 415 pp. \$32.95. ISBN 0471-91798-2.

The explosive growth in the discipline of materials science is substantially chemistry-driven and underscores the importance of including solid-state chemistry in the chemistry curriculum. Conventional treatments of this topic in physical and inorganic textbooks have often of necessity been superficial, reflecting the many competing subjects to be covered. As the undergraduate and graduate curricula evolve, however, solid-state chemistry is likely to assume a larger role in existing courses and to acquire status as a separate course. *Basic Solid State Chemistry* is an early entry in this potential market.

Much of this text by West appeared earlier under the title, *Solid State Chemistry and its Applications* and was reviewed previously in this journal (Honig, J. M. *J. Am. Chem. Soc.*, **1985**, *107*, 4806). West has condensed his 1984 work into a more pedagogically useful form.

The organization of the text begins with an extensive (roughly 60 pages) survey of crystal structures, including descriptions of Miller indices, *d*-spacings, close-packed structures, and other common structures. A second chapter of roughly equal length explores bonding in solids. Much of this section is standard inorganic text material, with representative topics including ionic radii, thermochemical calculations, and an introduction to band theory. Experimental techniques follow, with the third chapter covering crystallography and diffraction techniques and the fourth chapter surveying microscopy, spectroscopy, and thermal analysis. Chapter 5 provides an extensive discussion of crystal defects, nonstoichiometry, and solid solutions. Chapter 6 covers the phase diagrams of solids. The final two chapters are devoted to descriptions of electrical, magnetic, and optical properties. Concluding the text are supplemental reading lists; four appendices; a set of basic, chapter-keyed study questions (without answers); and an extensive index.

Overall, the text presents a good, nonmathematical survey of solid-state chemistry. Strengths include the chapters on crystal structures, crystallography and diffraction techniques, crystal defects, and phase diagrams. The numerous illustrations drawn from the literature include recent developments in high-temperature superconductivity and conducting polymers.

A weakness of the text is its uneven subject treatment. While it is admittedly difficult to cover everything, synthetic methods are largely ignored, despite the importance of preparative conditions in determining the physical properties of solids. Similarly, band theory, various spectroscopic techniques, and some descriptions of physical properties comprising the last two chapters are treated in a disappointingly cursory fashion. There are also some nonlinearities in topic sequence that may frustrate the reader: band gaps, for example, are used to interpret salt ionicities before their significance has been discussed.

From a student's perspective (having polled several), the book is clearly written and generally well organized. The style is a bit dry; a livelier presentation might be more effective in engaging an initially disinterested student.

Its deficiencies notwithstanding, *Basic Solid State Chemistry* can certainly be recommended as supplementary reading for a course that

includes solid-state chemistry among its subjects. For a course exclusively devoted to this area, it can serve as a primary text for some topics but needs to be augmented for others.

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The Flavonoids: Advances in Research since 1980. By J. B. Harborne (University of Reading). Chapman and Hall: London and New York, 1988. xiv + 621 pp. \$247.50. ISBN 0-412-28770-6.

Nineteen experts contributed the 16 chapters of the third volume of the *Flavonoids* series, which reviews the progress achieved from 1980 to 1985. It is, however, more than a mere description of the work done recently. The volume includes checklists of all known structures, a plant species index, and frequent references to the two previous publications in the series. It is indeed a source of information about all aspects of flavonoids.

The first 10 chapters are related to the various classes of flavonoids: anthocyanins, flavans and proanthocyanidines, C-glycosidylflavonoids, biflavonoids, isoflavonoids, neoflavonoids, flavones and flavonols, flavones and flavonolglycosides, minor flavonoids, and miscellaneous flavonoids. The critical review deals with the multiple aspects of the investigations in each class, isolation, analytical procedures, synthesis, distribution, biochemistry, and biological activities to mention only a few of them.

Chapter 11 provides an account of the biosynthesis, with emphasis on pathways to precursors, individual steps of flavonoid classes and modifications, and regulation of enzyme activities. Four chapters are then related to the distribution and evolution of flavonoids in lower plants, in gymnosperms, in the dicotyledons, and in the monocotyledons.

The 16th and last chapter deals with flavonoids and flower color.

This book will prove a valuable tool for scientists interested in botany and in the chemistry and biochemistry of plants. Specialists will appreciate the thorough covering of the field and the wealth of information contained in the numerous, neatly presented tables. The multidisciplinary approach provides a fascinating source of research ideas in the broad field of biotechnologies. This volume is a constant reminder that, in the study of nature and natural products, multidisciplinaryity is highly rewarding.

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Introduction to Quasicrystals. Aperiodicity and Order. Volume 1. Edited by Marko V. Jaric (Texas A&M University). Academic Press: San Diego, 1988. x + 285 pp. \$49.50. ISBN 0-12-040601-2.

One must take a special perspective toward this book since it represents an entirely new subject area that began with Schechtman, Blech, Gratias, and Cahn reporting in 1984 the existence of a structure generally held to be impossible to form. The newness of the area that has resulted should strongly temper the expectations one holds for the book. Understandably, about three-fourths of the book presents the results of phenomenological studies and descriptive information on icosahedral quasicrystals. The discussion of the subject is necessarily limited in its scope since, at best, there are only threads of understanding and certainly no comprehensive intellectual fabric. This dilemma can be appreciated by comparing the Preface to the actual contents of the book. The Preface correctly recognizes the importance of findings in Chaos and the attractiveness of discovering if new temporal and spatial senses of order are related in ways that are not commonly understood. However, no further

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