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COLUMBIA UNIV NEW YORK DEPT OF CHEMISTRY  
STRUCTURAL AND DYNAMIC STUDIES OF MATERIALS POSSESSING HIGH ENER--ETC(U)  
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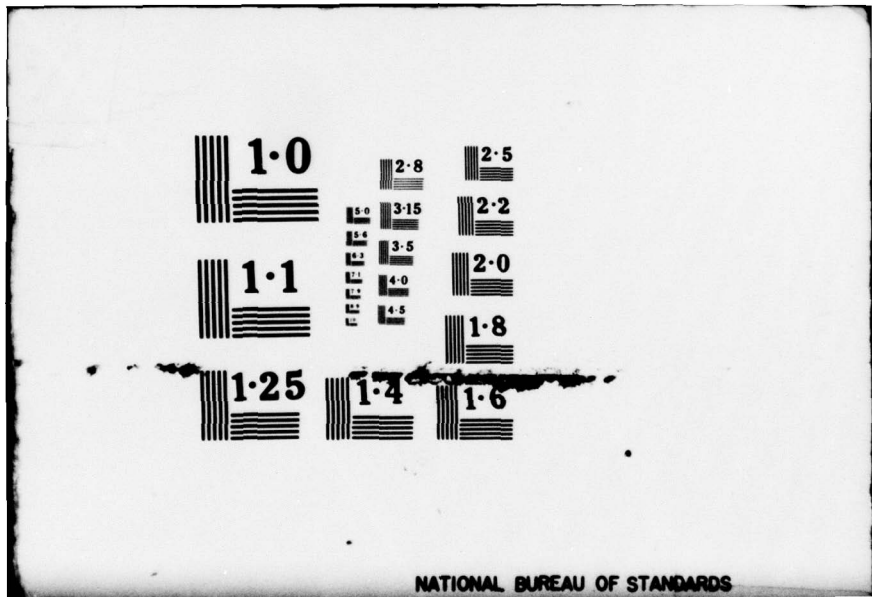
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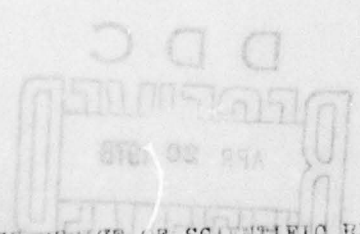
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N.J. Turro\* and V. Ramamurthy, "Source of Inefficiency in Photochemical Triplet Cycloaddition and Hydrogen Abstraction Reactions," Mol. Photochem., 8(3), 239-253 (1977).

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## 9. ABSTRACT OF OBJECTIVES AND ACCOMPLISHMENTS

The technology related to reconnaissance, information retrieval, communications, radiation detection and other areas require and will advance as our knowledge of excited molecules and their emission properties expands. Novel laser uses, novel laser design and advancement of the state of the art of photochemistry depends heavily on both technical, theoretical and experimental developments. The goal of this research is to provide a theoretical and experimental basis for the understanding of the properties of excited molecules and their emission characteristics. We have sought to elucidate the available mechanisms for thermal and photochemistry activation of selected high energy content materials which are chemiluminescent. Several novel chemiluminescent organic reactions have been discovered and thoroughly investigated. A particular emphasis has been directed toward the identification of chemiluminescent systems which might have the highest likelihood of providing a framework for the development of operating chemical lasers which emit in the visible spectral range. The thermal chemistry and photochemistry of dioxetanes and of valence isomers of benzene have been investigated. The chemistry of singlet oxygen is being explored to develop simple and convenient means of producing this species. An important development has been the discovery that the catalytic thermal generation of singlet oxygen from ground state (triplet) oxygen may be feasible. The possibility of using singlet oxygen in developing chemical lasers is also being explored. Finally, the mechanisms of electronic energy transfer and oxidation in polymers is being investigated with the goal of providing a fundamental understanding of these processes that will allow the systematic and rational development of strategies for stabilization of polymer systems.

### Research in Chemiexcitation

As a result of our investigations of the decomposition of dioxetanes and dioxetanones an unexpected but reliable selection rule for chemiexcitation has evolved: Triplet carbonyl fragments are formed directly and selectively. For example, in the case of tetramethyl-1,2-dioxetane acetone triplets are formed in 50% yield whereas acetone singlets are formed in 0.5% yield. We proposed a simple orbital basis for the spin-orbit coupling required to rationalize these results. The key idea in our model is that a  $p_x \rightarrow p_y$  orbital jump on an atomic center is required for efficient spin-orbit coupling. We have combined this concept with that of a topological description of energy surfaces in order to classify chemiexcitation processes. The "surface topology" classification allows a qualitative understanding of the vast difference in efficiencies of chemiexcitation of dioxetanes relative to benzene valence isomers.

### Adiabatic Photoreactions

The conversion of an electronically excited reactant,  $R^*$ , directly into an electronically excited product,  $P^*$ , is termed an adiabatic photoreaction. Such processes have great potential for developing novel laser systems since a population inversion in  $P^*$  is achieved as soon as the first product molecules are formed, if the process is efficient.

We have discovered the first examples of a number of novel adiabatic organic photoreactions including (a) the photoconversion of naphthalene to triplet naphthalene; (b) the photoconversion of dioxetanes to triplet ketones; (c) the photoconversion of Dewar benzene to triplet benzene.



### Quantum Chain Reactions

A chain reaction in which an electronically excited state is the propagating species is termed a "quantum chain" reaction. Such processes may be useful in modeling "run away" reactions in which a large amount of heat is released rapidly. We have discovered the first examples of efficient quantum chain organic reactions. For example, the photoinitiated decomposition of dioxetanes possess quantum chain lengths greater than 1000! The quantum chain process for Dewar benzenes has also been uncovered and in this case chain lengths are of the order of 10.

### Energy Transfer Processes in Polymers

We have employed chemiexcitation techniques as a simple and quantitative method to study singlet-singlet, triplet-singlet and triplet-triplet energy transfer processes in polymers. This procedure allows convenient investigation of systems that are not amenable to standard photoexcitation techniques, i.e., strongly absorbing acceptors may be employed.

### Photoinitiation of Polymer Degradation "in the Dark"

We showed that the chain emission of a styrene-methyl isopropenyl ketone copolymer can be induced by acetone triplets produced "in the dark" from tetramethyl-1,2-dioxetane.

### Energy Hopping Between Carbonyl Chromophores

We have shown via a direct method and an indirect method that energy hopping may occur between carbonyl chromophores. The key idea in these experiments is

to use chemiexcitation to produce an electronically excited carbonyl fragment which then transfers excitation to a molecule in a large excess of ground state carbonyl containing molecules.

#### Discovery of Novel Chemiluminescent Systems

Our investigation of high energy content materials has led to the discovery of novel chemiluminescent systems. For example, the rearrangement of Dewar benzenes to benzene has been shown to be generally chemiluminescent. This information has been used to demonstrate Dewar benzene intermediates in the decomposition of strained azo compounds and in the thermal rearrangement of bis-cyclopropenyls to benzenes.

#### Reactions of Molecular Oxygen with High Energy Ethylenes

We have shown that reaction of molecular oxygen (both  $^1\text{O}_2$  and  $^3\text{O}_2$ ) with strained acetylenes produces 1,2-diketones in a chemiluminescent reaction probably proceeding via a dioxetene. An exciting spin-off of this study was the discovery that an oxidizing species, possibly singlet oxygen, is produced when the acetylene is heated with air. As a result we have proposed a general mechanism for the catalytic thermal generation of singlet oxygen (eq. 1).



#### Singlet Oxygen in Polymer Systems

We have employed chemiluminescence methods to study the physical and chemical properties of singlet oxygen in polymers. From measurements of chemiluminescence

as a function of substrates that react with singlet oxygen, we have evaluated the lifetime and/or diffusion parameters of singlet oxygen in polymers. We have also initiated studies which will allow measurements of reactivities of different polymer substrates toward singlet oxygen and identification of the processes by which singlet oxygen causes polymer degradation.



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(Abstract, continued) We have sought to elucidate the available mechanisms for thermal and photochemistry activation of selected high energy content materials which are chemiluminescent. Several novel chemiluminescent organic reactions have been discovered and thoroughly investigated. A particular emphasis has been directed toward the identification of chemiluminescent systems which might have the highest likelihood of providing a framework for the development of operating chemical lasers which emit in the visible spectral range. The thermal chemistry and photochemistry of dioxetanes and of valence isomers of benzene have been investigated. The chemistry of singlet oxygen is being explored to develop simple and convenient means of producing this species. An important development has been the discovery that the catalytic thermal generation of singlet oxygen from ground state (triplet) oxygen may be feasible. The possibility of using singlet oxygen in developing chemical lasers is also being explored. Finally, the mechanisms of electronic energy transfer and oxidation in polymers is being investigated with the goal of providing a fundamental understanding of these processes that will allow the systematic and rational development of strategies for stabilization of polymer systems.

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