Reactions of Charged and Neutral Recoil Particles Following Nuclear Transformations

Final Report

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Principal Investigator:

Hans J. Ache
Professor of Chemistry
Virginia Polytechnic Institute and State University

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The various activities under this contract during the contract period and their main objectives can be summarized as follows:

STUDY OF THE REACTIONS OF ENERGETIC PARTICLES GENERATED IN NUCLEAR REACTIONS.

Chemical reactions in the gas phase and liquid phase are an essential part of basic energy related research and the ability to control them is of tremendous practical importance.

In order to control these reactions it is necessary to understand the mechanisms and kinetics involved in these processes which in turn requires the identification and structural determination of intermediates, measurement of rate constants for their formation, where intermediates could be defined as atoms, radicals, ion and ion molecules, the influence of three dimensional molecule structure on the rate constants, steric effects, etc.

These are exactly the topics of modern hot atom chemistry and they were also the subject of this work.

Gas phase hot atom chemistry has been for years an extremely successful tool in the assessment of the parameters which determine the progress of chemical reactions which occur with kinetic energies greater than thermal and in chemical dynamics. The ultimate goal of hot atom chemistry and of non-Boltzmann chemistry in general has been characterized as "the description of chemical reactions in terms of the respective cross-sections for each of the various possible reactions at each of the energies at which the reaction is possible".

Within this framework our special interest was concentrated on the evaluation of the nature of the reacting species (ions or neutral atoms), its electronic state, the kinetic energy involved, the exact mechanism, configuration and lifetime of the intermediates and the stereochemical course of the reactions.

In the condensed phase the assessment of the detailed mechanism of the hot atom reactions is further complicated by the greater multiplicity of overlapping reactions and the enhanced presence of thermal processes which obscure the hot reaction channels.

Thus in addition to the objectives, as outlined above for gas phase reactions, other major considerations had to be included in our investigation, namely, the presence of caging effects and the effects of the solvent on the course of the reactions. It was especially this latter topic which was thoroughly scrutinized during the more recent contract periods.

We feel that the evaluation of the molecular properties of solvent molecules which appear to be responsible for their caging efficiency can significantly contribute to a better understanding of the photocatalytic cage effects and/or deexcitation processes. These factors may in turn substantially affect the yields of photochemical reactions used in photochemical energy conversion processes.

The importance of the search for direct non-synthetic methods to incorporate shortlived radionuclides into molecules of biochemical interest to prepare carrier free compounds of high specific activity to be used in nuclear medical research has long been recognized as an important goal in the nuclear science program.

Under this contract methods such as the excitation labelling which have been developed as a result of our basic hot atom chemistry program have been tested as potential synthetic routes to prepare such radiopharmaceuticals.

Another aspect of hot atom chemistry most directly related to Energy Research and Development is the fact that energy generation via thermonuclear fusion processes in CTR's is strongly impacted by "hot atom phenomena". Hot ions and atoms produced in the plasma impinge with considerable kinetic energies on the vacuum walls of the CTR causing corrosion of the first wall resulting in plasma contamination.

Therefore, the fate of hot atoms and ions incorporated into deeper layers of the materials and their reactions with the

materials suggested as wall material or as protective curtain such as graphite, silicon carbide, boron carbide, etc., and their potential effect on CTR operation was investigated under this contract.

Another section of our program dealt with the study of micellar systems, which have become important components in energy related research.

Micelles have been employed to facilitate photochemical conversion of light energy into chemical energy which is potentially applicable for storage of solar energy in a useful form and as injection fluids for enhanced (tertiary) oil recovery.

We, therefore, utilized the full potential of the positron annihilation technique, a method whose chemical applications were to a great extent developed in this laboratory (in combination with conventional methods) to study the structural properties of and the dynamics involved in micellar processes including inverted micelles, microemulsion and surfactant vesicles.

Personal and Publications

During the contract period from January 1, 1967 - December 31, 1980 a total of 12 postdoctoral research associates, 21 graduate students, 3 laboratory technicians and several visiting scientists were involved in the various phases of this work. Of these 21 graduate students, 14 received their Ph.D. and 3 their M.S. degree.

The work carried out under this contract has been described in numerous publications (see attached publication list; work done entirely or partially with DOE support is indicated by an asterisk) and presented in over 230 papers, 105 by invitation, at international and national scientific meetings, seminars, etc.

Ache, Hans J.

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Appendix

Collection of Reprints Published During Contract Period Not Previously Submitted

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