COO-3498-2

Progress Report

Contract No. AT(11-1)3498

Fundamental Studies in Isotope Chemistry

between

Division of Research

U.S. Atomic Energy Commission

and

University of Rochester

Jacob Bigeleisen
Professor of Chemistry
Principal Investigator

1 July 1971 - 30 June 1972

DISCLAIMER

Scope of Activities

During the past year our researches in "Fundamental Studies in Isotope Chemistry" have given definitive results on the relationship between the mean square forces in simple liquids and solid and the structures of the liquids and solids. The important experimental finding has been that structure is not important, rather it is just the bulk density which differentiates the mean square force in the solid, liquid or gas as well as its temperature coefficient for simple liquids like neon argon and krypton. These results were obtained by vapor pressure measurements on enriched krypton isotopes (Dr. M.W. Lee, postdoctoral fellow and D.M.Eshelman, graduate(Hooker) fellow), determination of the single stage isotope separation factor for liquid argon from the triple point to the critical point (Dr. J.T.Phillips, postdoctoral fellow, and Dr. Carl U. Linderstrom-Lang, visiting senior research associate from the Danish AEC), and our previous measurements on the vapor pressure isotope effects in solid and liquid neon and argon.

Theoretical calculations of the mean square force in fluid argon, gas and liquid, by the Percus-Yevick and Weeks, Chandler, Anderson perturbation theories give excellent agreement with the experimental fractionation factor and with more detailed calculations from molecular dynamics.

F. Mandel (postdoctoral fellow).

The reduced partition function ratios $^{238}UF_6/^{235}UF_6$ in the gas, liquid and C_7F_{16} solution have been calculated to give chemical exchange, distillation,exchange distillation, and adsorption isotope separation factors for the uranium isotopes, using UF_6 gas. Significantly $^{238}UF_6$ has been calculated to be more volatile than $^{235}UF_6$ in agreement with experiment. This project was carried out to provide technological data necessary for the work of the Benedict Committee appointed by the AEC Assistant General Manager for Production, Mr. G. F. Quinn.

Work is in progress on the following projects:

1) vapor pressure isotope effect for $^{13}CO_2$ and $^{12}C\ ^{18}O_2$ compared with $^{12}C^{16}O_2$. (Lee and Bilkadi).
2) liquid-vapor isotope ($^{15}N/^{14}N$) separation factor for NO from the triple point to the critical point. (Eshelman and Torre).
3) calculation of the mean square force in liquid argon for the Lennard-Jones 6-12, exponential -6, and Barker potentials. (Mandel).
4) correlation of isotope effects with molecular forces in polyatomic molecules (Bigeleisen with Professor T. Ishida, Brooklyn College, and Professor W. Spindel, Yeshiva University).

Mr. William Watson continues to supply the necessary technical support for the program. During the coming year we anticipate a major interruption to our experimental

program. The Department of Chemistry of the University of Rochester is moving to new quarters, Hutchison Hall.

Publications

J. Bigeleisen, T. Ishida and W. Spindel. Correlation of Isotope Effects with Molecular Forces II. Triatomic Molecules. J. Chem. Phys. 55, 5021 (1971). Removed

M. W. Lee, D. M. Eshelman, and J. Bigeleisen. Vapor Pressures of Isotopic Krypton Mixtures. Intermolecular Forces in Solid and Liquid Krypton. J. Chem. Phys. 56, 4584 (1972). Removed

J. Bigeleisen. The Calculation of Some Isotope Separation Factors for Chemical Exchange, Distillation, Exchange Distillation and Absorption using UF_6(g). J. Inorg. and Nuc. Chem. In Press. Removed

J. T. Phillips, C. U. Linderstrom-Lang, and J. Bigeleisen. Liquid-Vapor Argon Isotope Fractionation from the Triple Point to the Critical Point. Mean Laplacian of the Intermolecular Potential in Liquid Argon. J. Chem. Phys. 56, 5053 (1972). Removed

Manuscripts in Preparation

F. Mandel. Calculations of the Isotope Separation Factor between Argon Liquid and Vapor. Removed

Lectures and Talks

Los Alamos Scientific Laboratory	
National ^{13}C Symposium	June 1971
State University College-Geneseo	October 1971
University of California-Berkeley	December 1971
Hildebrand Symposium	
University of Rochester-Chemistry	December 1971
Colloquium	

Professional and College Activities

Professional Activities:

Special Evaluation Panel on Isotope Separation - AEC, Oak Ridge Operations Office

Gordon Research Conferences: Trustee and Chairman of the Board

Visiting Committee - Carnegie-Mellon University

National Academy of Sciences - Chairman, Chemistry Section

Division of Chemistry and Chemical Technology, National Research Council

College Activities:

Iota Chapter - Phi Beta Kappa, President Elect

Faculty Senate Committee on Fringe Benefits