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HIGH TEMPERATURE GAS ENERGY TRANSFER.(U)  
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ANNUAL SUMMARY REPORT, 1 August 1979 - 31 July 1980

High Temperature Gas Energy Transfer

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OFFICE OF NAVAL RESEARCH  
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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) Work under this contract is summarized. Results are described on collisional transfer of vibrational energy by a variety of gases and on the temperature dependence of transfer efficiency. A novel and simple Variable Encounter Method has been applied to the study of energy transfer between gas molecules and a wall, at high temperatures and high levels of activation, in the transient region. Results by the Diffusion Cloud Method are also described.			

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I. Summary of Progress. This report covers the period 1 August 1979 - 31 July 1980. The studies described below demonstrate the following important results and conclusions.

- a) The collisional efficiencies for transfer of vibrational energy between a wall and various polyatomic molecules over a wide range of molecule energies have been determined for the first time. Thus, the proper magnitude of the efficiency relative to gas-phase processes is now known quantitatively for some systems; qualitatively, the wall is more efficient than homogeneous collisions.
- b) The collisional efficiency of a wall for energy transfer has been studied over a wide range of temperatures and found to decrease with rise of temperature. It approaches strong collider behavior at lower temperatures.
- c) Homogeneous studies on energy transfer between a polyatomic molecule and various bath gases at high temperatures also corroborate a decrease in efficiency with rise of temperature.
- d) Correlations have been found between collision efficiency and molecular structure parameters. In general, efficiency declines with molecule heat bath size.
- e) The transient region of energy accommodation has been studied for the first time for polyatomic molecules; mean first passage times have been measured.

These results are novel and are very important for many types of "practical" systems involving piston and jet walls, turbine blades, missile surfaces, etc., as well as for "pure" applications in shock tube, combustion and laser systems, and for theoretical understanding.

II. Complete listing of Technical Reports:

1. Technical Report TR01, 7 January 1976.

Vibrational Energy Transfer in Reaction Systems at Elevated Temperatures  
by B. S. Rabinovitch, D. G. Keil, J. F. Burkhalter and G. B. Skinner.

In this report a theoretical analysis was given of shock tube data for high temperature gas reactions. The data were analyzed to show that collisional energy transfer efficiency appears to decrease at high temperatures.

2. Technical Report TR02, 20 April 1976

Temperature Dependence of the Arrhenius Activation Energy. High Temperature Limit, J. F. Burkhalter and B. S. Rabinovitch.

In this report a conventional approximation to the high temperature vibrational partition function is shown to be defective, and the usual high temperature expression for the Arrhenius activation is in error. The variation of the activation energy for unimolecular reactions as a function of temperature is calculated for various representative systems.

3. Technical Report TR03, 15 October 1976

Intermolecular Vibrational Energy Transfer in Thermal Unimolecular Systems, D. C. Tardy and B. S. Rabinovitch.

This report gives a comprehensive and detailed survey and analysis of energy transfer in thermal gaseous systems. The present status of the field is delineated and future areas of work indicated.

4. Technical Report TR04, 15 April 1977

On the Use of Exact Vibrational State Counting Methods in RRKM Rate Calculations, S. E. Stein and B. S. Rabinovitch.

This report describes an algorithm for accurate state sum and density calculations and refutes an error in the literature.

5. Technical Report TR05, 15 August 1977

Vibrational Translational Energy Transfer in Atom-Polyatomic Molecule Collisions in Thermal Reaction Systems by I. Oref and B. S. Rabinovitch.

In this report a simple theoretical model describing energy transfer probabilities is given. Conservation of angular momentum was imposed. The importance of completeness and detailed balance conditions and correction of an earlier model in the literature are illustrated. The model is applied to experimental data.

6. Technical Report TR06, 1 September 1977

Problems of Diffusion in a Low Pressure Gas Stream as Related to the Polanyi Diffusion Flame (Cloud) Method by D. G. Keil, J. F. Burkhalter and B. S. Rabinovitch.

A theoretical and experimental study was made of flow and diffusion of a reactant in a low pressure gas stream. Connection is made with earlier theoretical analysis.

## 7. Technical Report TR07, 10 October 1978

Do Highly Excited Reactive Polyatomic Molecules Behave Ergodically?  
I. Oref and B. S. Rabinovitch.

The literature is surveyed and analyzed. A variety of excitation techniques including crossed molecular beam, laser, chemical activation, photochemical and thermal are considered. The answer to the title question is "yes." The relationship to the present work on intermolecular transfer is the following: multiphoton laser experiments frequently involve collisional heating of the gas. In order to understand the results, one must have a good basis for interpretation of data. In this survey, some suggested models for energy relaxation by molecular collisions are criticized and data reinterpreted in light of results obtained in this laboratory.

## 8. Technical Report TR08. 1 November 1978

Vibrational Energy Transfer in Thermal Unimolecular Systems by the Diffusion Cloud Method. Cyclopropane. E. Kamaratos, J. D. Burkhalter, D. G. Keil and B. S. Rabinovitch.

A study of vibrational energy transfer by the Diffusion Cloud Method has been made at temperatures from 975 K to 1175 K in the cyclopropane isomerization system.  $H_2$ , He,  $N_2$  and  $CO_2$  were studied as inert bath gases. Their relative efficiencies increase in that order. Values of  $\langle \Delta E \rangle$  vary from  $150 \text{ cm}^{-1}$  to  $1100 \text{ cm}^{-1}$  at 975 K. It was shown that these values decline, and the collisional efficiencies,  $\beta_c$ , decrease markedly with rise of temperature to 1175 K. This is only the second measurement of such a phenomenon.

## 9. Technical Report TR09. 15 March 1979

Collisional Relaxation of Non-Equilibrium Vibrational Energy Distributions in a Thermal Unimolecular System. Surface Collisions. D. F. Kelley, B. D. Barton, L. Zalotai, and B. S. Rabinovitch.

A novel, simple technique, the Variable Encounter Method, is described for obtaining information on energy transfer efficiency between a gas and a solid in the transient region, and as a function of temperature. The method is as powerful as it is simple. Results for cyclopropane are described. Decrease in collisional efficiency  $\langle \Delta E \rangle$  with rise of temperature is confirmed, and the form of the transfer probability matrix  $\underline{P}$  is discovered. This method and these results are considered to be some of the most innovative and important that have ever been made in this field.

## 10. Technical Report TR10. 15 June 1979

Transients in Vibrational Relaxation of Polyatomic Molecules at High Temperatures by VEM. M. C. Flowers, D. F. Kelley, F. C. Wolters, and B. S. Rabinovitch.

Extension of the VEM technique to cyclobutane is described. An exponential form of  $P$  is required to fit the data. The value of  $\langle \Delta E \rangle$  declines with rise of temperature.

The following were submitted in the present report period:

## 11. Technical Report TR11; 15 September 1979

Collisional Relaxation of Transient Vibrational Energy Distributions in a Thermal Unimolecular System. The Variable Encounter Method. D. F. Kelley, L. Zalotai and B. S. Rabinovitch.

The Variable Encounter Method permits the study of the transient in the relaxation of an initial vibrationally cold ensemble of molecules in a vibrationally hot distribution by a known and variable number of successive collisions with a hot wall. The system studied was the isomerization of 1,1-cyclopropane- $d_2$  with a fused quartz wall temperature of 800 K to 1175 K, and average number of collisions from 2.3 to 22.3. The detailed theory of the experiment was presented, together with the method of data deconvolution. Various modified gaussian and exponential models of energy transfer were found to give agreement with the data. The average down-step size was found to decline from  $\leq 3500 \text{ cm}^{-1}$  at the lowest temperature to  $\sim 2500 \text{ cm}^{-1}$  at the highest on the basis of a gaussian model, but heterogeneous energy transfer is more efficient than homogeneous gas-gas collisional transfer. A mathematical analysis of the relation between mean first-passage times and incubation times is given. Incubation times increase from  $\sim 7$  to  $\sim 12$  collisions with increasing temperature. Transient population distributions and the sequential reaction probabilities as a function of collision number are calculated.

## 12. Technical Report TR12. 10 October 1979

Vibrational Energy Transfer in a Diffusion-Flow Cyclopropane- $d_2$  System. J. F. Burkhalter, E. Kamaratos, and B. S. Rabinovitch.

In this study, absolute rate constants for isomerization of cyclopropane- $d_2$  and high temperature homogeneous energy transfer were determined in a diffusion-flow system. The relative rates of the competitive isotopic isomerization channels were measured for two bath gases,  $N_2$  and He, at two temperatures, 973 K and 1073 K. Values of the average energy down-jump size  $\langle \Delta E \rangle$  were computed from both the absolute rates and the isotopic relative rates by suitable modelling by a stochastic calculation. The results confirm earlier diffusion cloud measurements.

## 13. Technical Report TR13, 10 October 1979

Transients in the Vibrational Excitation of Cyclobutane Decomposition Using the Variable Encounter Method. M. C. Flowers, F. C. Wolters, B. D. Barton and B. S. Rabinovitch.

The probability of reaction of cyclobutane molecules in a fixed-time interval after experiencing a known number of collisions with a hot surface at temperatures between 749 K and 1126 K was determined using the Variable Encounter Method. Calculations utilizing exponential or gaussian models for energy transfer enabled the average amounts of energy transferred for deactivating collisions,  $\langle \Delta E' \rangle$ , to be estimated. The exponential model fits the experimental data best and, using this model,  $\langle \Delta E' \rangle$  is  $2430 \text{ cm}^{-1}$  at 748 K and decreases to  $1470 \text{ cm}^{-1}$  at 1123 K. Surface collisions are more efficient than binary gas collisions; but cyclobutane is a less efficient partner than cyclopropane- $\text{d}_2$  (TR No. 11). The results follow from a quasi-statistical model of energy accommodation.

## 14. Technical Report TR14, 1 December 1979

A Crucial Demonstration of Strong Collisional Behavior of Vibrational Energy for Gas-Surface Collisions. M. C. Flowers, F. C. Wolters, D. F. Kelley and B. S. Rabinovitch.

Application of the VEM technique was made to the thermal cyclobutene decomposition system. Vibrational energy transfer in the cyclobutene - seasoned quartz surface system was studied. Collisional efficiency again declines with rise of temperature, but the occurrence of strong collisions was demonstrated in crucial manner at temperatures below 450 K. The wall appears to behave like a theoretical strong collider below 450 K. This is a very novel finding.

## 15. Technical Report TR15, 31 March 1980

Gas-Surface Vibrational Energy Transfer in the Transient Region of a Low-Pressure Unimolecular Reaction. B. D. Barton, D. F. Kelley and B. S. Rabinovitch.

The Variable Encounter Method, for the study of gas-wall vibrational energy transfer in the transient region of a unimolecular reaction was applied to the isomerization of cyclopropane to propylene. Temperatures in the range 900 K - 1125 K were employed. The average probability of reaction per collision,  $\bar{P}_c(m)$ , was deduced from the data and compared with a theoretical stochastic calculation based on both gaussian and exponential models for the energy transfer probability. The former model is more appropriate. The efficiency of a seasoned quartz wall was greater than gas-gas collisions of substrate and the efficiency declined with increase of temperature. The steady state is closely approached (90%) in a comparatively small number of gas-wall collisions - 10-20, approximately. The efficiency of transfer by cyclopropane is comparable with that by cyclopropane- $\text{d}_2$  (T.R. No. 11).



### III. Listing of Publications.

1. Vibrational Energy Transfer in Reaction Systems at Elevated Temperatures, Proceed. Tenth Internat. Shock Tube Sympos. ed. G. Kamimoto, Kyoto, 1976.
2. Intermolecular Vibrational Energy Transfer in Thermal Unimolecular Systems. D. C. Tardy and B. S. Rabinovitch, Chem. Revs. 77, 369 (1977).
3. On the Use of Exact Vibrational State Counting Methods in RRKM Rate Calculations. S. E. Stein and B. S. Rabinovitch, Chem. Phys. Lett. 49, 183 (1977).
4. Vibrational Translational Energy Transfer in Atom-Polyatomic Molecule Collisions in Thermal Reaction Systems. I. Oref and B. S. Rabinovitch, Chem. Phys. 26, 385 (1977).
5. Problems of Diffusion in a Low Pressure Gas Stream as Related to the Polanyi Diffusion Flame (Cloud) Method, D. G. Keil, J. F. Burkhalter and B. S. Rabinovitch, J. Phys. Chem. 82, 355 (1978).
6. Vibrational Energy Transfer in Thermal Unimolecular Systems by the Diffusion Cloud Method. Cyclopropane. E. Kamaratos, J. F. Burkhalter, D. G. Keil and B. S. Rabinovitch, J. Phys. Chem. 83, 984 (1979).
7. Do Highly Excited Reactive Polyatomic Molecules Behave Ergodically? I. Oref and B. S. Rabinovitch, Accts. Chem. Res. 12, 166 (1979).
8. Collisional Relaxation of Non-equilibrium Vibrational Energy Distributions in a Thermal System. Surface Collisions, D. F. Kelley, B. D. Barton, L. Zalotai, and B. S. Rabinovitch, J. Chem. Phys. 71, 538 (1979).
9. Transients in Vibrational Relaxation of Polyatomic Molecules at High Temperatures by Variable Encounter Method. M. C. Flowers, B. D. Barton, F. C. Wolters and B. S. Rabinovitch. Am. Soc. Mass. Spectrom. Proceedings, ed. A. E. Harrison, 27, 661 (1979).
10. Collisional Relaxation of Transient Vibrational Energy Distributions in a Thermal Unimolecular System. The Variable Encounter Method. D. F. Kelley, L. Zalotai, and B. S. Rabinovitch. Chem. Phys. 46, 379 (1980).
11. Vibrational Energy Transfer in a Diffusion-Flow Cyclopropane-d<sub>2</sub> System. J. F. Burkhalter, E. Kamaratos, and B. S. Rabinovitch, J. Phys. Chem. 84 (1980) 476.
12. A Crucial Demonstration of Strong Collisional Behavior of Vibrational Energy for Gas-Surface Collisions. M. C. Flowers, F. C. Wolters, D. F. Kelley and B. S. Rabinovitch, Chem. Phys. Lett. 69, 543 (1980).
13. Transients in the Vibrational Excitation of Cyclobutane Decomposition Using the Variable Encounter Method. M. C. Flowers, F. C. Wolters, B. D. Barton and B. S. Rabinovitch, Chem. Phys. 47 (1980) 189.
14. Gas-Surface Vibrational Energy Transfer in the Transient Region of a Low-Pressure Unimolecular Reaction. B. D. Barton, D. F. Kelley, and B. S. Rabinovitch. J. Phys. Chem. 84, 1299 (1980).

#### IV. Current Work

The following work has been completed (but reports have not yet been circulated), or is in progress.

1. Methylcyclopropane. A study by VEM has been completed. The purpose of the study is to extend the test of the effect of molecular structure on energy transfer efficiency. The work goes over a temperature range up to 1100 K. This molecule has a looser vibrational structure than does cyclobutane and the efficiency is a littler lower. This will be described in TR16.
2. Cyclopropane-Cyclobutane. A study by VEM has been nearly completed of a mixture of the two cycloalkanes in order to resolve some earlier discrepancies. The work previously reported in TR11 is confirmed. This will be described in TR17.
3. A study of iodopropane by VEM has been initiated in order to extend the test of the relation between molecular structure and collisional transfer efficiency.

#### Future Work.

The following studies are planned in the year ahead:

1. Effect of molecular structure.
  - a) Nitroalkanes. The test of the relation between collisional efficiency and molecular structure will be extended to nitroalkanes such as nitromethane/nitroethane. These molecules have a large dipole moment and should prove to be of considerable interest. It appears that efficiency may depend (increase) markedly when strength of the molecule-wall interaction.
  - b) Isotope Effect. There is an indication that cyclopropane-d<sub>2</sub> is less efficient than the light compound. The test will be extended to cyclopropane-d<sub>6</sub> which is an even larger energy sink (lower frequencies).
2. Temperature Variation.

It is important to extend the VEM studies upward to temperatures that overlap shock and laser decomposition studies more fully. We will push these investigations to above 1300 K. This work is of extreme practical and theoretical importance for detecting the possible transition of the observed behavior from an attractive potential-dominated accommodation phenomenon to a Landau-Teller type repulsive potential-dominated impulsive phenomenon.

### 3. Surface Variation

We hope to examine the effect of variation of the nature of the wall surface and propose to investigate next a metal (gold) surface.

### 4. Initial Energy Distribution

Time permitting, we will investigate the result of changing the initial temperature (average vibrational energy) of the molecule species.

## V. Personnel

The following graduate students and fellows have been involved on this project in the period.

Barrie Barton

John Burkhalter

Dr. M. C. Flowers (Senior Lecturer, Southampton University, on leave)

Dr. Toshio Kasai

David Kelley

Dr. Fred C. Wolters

B. S. Rabinovitch  
Principal Investigator

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