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Contract N5-onr-86101, T. O. I.

Project NR 010 101

THE THERMAL ORTHO-PARA CONVERSION OF HYDROGEN

A Summary Report

By

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HYDROCARBON RESEARCH, INC.

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The Thermal Ortho-Para Conversion of Hydrogen

A Summary Report

Introduction

The main activity pursued under Contract NR-861 with the Office of Naval Research has been devoted to a calculation of the rate of conversion of para-hydrogen to ortho-hydrogen in the gas phase at relatively high temperatures. The present report is intended to summarize the results which have been obtained to date as well as to indicate possible directions of future investigations.

It was intended, as stated in the original proposal which was made by H. R. I. to the Office of Naval Research, to calculate the rate of the thermal para-ortho conversion of hydrogen as a subsidiary problem in a more extensive consideration of chemical reaction kinetics. As it developed, however, this particular reaction soon posed difficulties that have persisted until the present. These difficulties are partly conceptual and partly mechanical (i.e., numerical complexity).

The essential conceptual difficulty has to do with the notion of chemical species and how to formulate the expression for the probability of chemical change occurring in a reacting system. In the present program each molecular species has been identified by suitable quasi-stationary wave functions. The probability that transitions

will occur from the reactants to the products then is determined by the usual time-dependent perturbation theory of quantum mechanics. This formulation is not especially rigorous and additional investigation along this line seems desirable. Nevertheless, the formulation seems to be reasonable and can be justified in an approximate manner.

The mechanical difficulties are associated with the lack of precise information regarding the (adiabatic) interaction between atoms and molecules, especially for small distances of approach to one another. The way in which these quantities enter into the expression for the rate of chemical reaction makes for extremely complicated sums and integrals. Even if the interactions between atoms and molecules were known with great precision, it is doubtful if they could be handled in the present stage of development of the theory.

The Wave Functions

In order to obtain a physically meaningful result for the time rate of change of the probability of chemical transition, both the initial and final wave functions associated with the transition must correspond to physically accurate descriptions of the system. To facilitate arriving at such wave functions we may imagine a reacting system as being prepared in some state which corresponds to a stationary solution of a Schroedinger's equation in which an asymptotic or other

approximation is made to the Hamiltonian of the system. Under the influence of the interaction (represented by the difference between the accurate and approximate Hamiltonians for the system), the wave functions for the system will no longer remain stationary. The changes that occur may be interpreted as consisting of two kinds: (1) non-chemical changes and (2) chemical changes. The non-chemical changes represent the usual kind of scattering in which the configurational structure of the system remains unaltered. The chemical changes, which are of interest here, are those in which the configurational structure of the system is altered.

In attempting to describe in this way the changes that occur in the system, it is convenient to assume that the perturbation interaction consists of two different kinds of terms, each contributing to the kinds of change mentioned. To provide a description that will emphasize the chemical changes, it seems clear that the description of the system, when as products or reactants, should already contain the effects of non-chemical changes. This may be accomplished by selecting as "zero-order" wave functions either stationary or non-stationary solutions of Schroedinger's equation with Hamiltonians containing the scattering, but not the chemical, potentials. We have adopted the latter course, following a procedure first introduced by Weisskopf and Wigner[1] and employed subsequently by Wilson[2]. For the elastic scattering by a central field of force, we have obtained

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(1) V. Weisskopf and E. Wigner, Zeits. F. Physik., 63, 54, (1930), 65, 18, (1930).

(2) A. H. Wilson, Proc. Camb. Phil. Soc., 37, 301, (1941).

the wave function describing the relative motion between the center of mass of a molecule and an atom in the form

$$F(\vec{k},\vec{r}) = (1/V^{\frac{1}{2}}) \sum_{n} (2n+1)\, i^n A_n(k)\, j_{n+\frac{1}{2}}(kr/\hbar)\, P_n(\vec{k}\cdot\vec{r}/kr), \qquad (1)$$

where

$\vec{k}$ is the momentum vector for relative motion of the centers of mass of the molecule and the atom,

$\vec{r}$ is the vector distance between the centers of mass.

$j_{n+\frac{1}{2}}$ is a spherical Bessel function of order n,

P_n is a Legendre function of the first kind of order n,

$A_n(k) = \left[1 + 4\pi^2 i\, \rho(E_k) \langle j_{n+\frac{1}{2}}(kr/\hbar) | V(r) | j_{n+\frac{1}{2}}(kr/\hbar) \rangle \right]^{-1},$

$\rho(E_k)$ is the density of free particle states per unit energy,

$V(r)$ is the central field potential between the molecule and atom, to account for the elastic scattering,

V is the region of normalization of the wave function.

The initial (reactant) wave function is taken to be a product of a molecular wave function and that expressed in Eq. (1). A similar form is adopted for the final (product) wave function. Explicitly, omitting the motion of the center of mass of the triatomic system of interest here, the initial wave function for a reactant state is taken as (Refer to Fig. 1),

$$\psi_i = H_n\left[a_1^{1/2} (\xi - \rho_1) \right] Y_K^M (\vec{\xi}/\xi)\, F(\vec{x}, \vec{r}_1), \qquad (2)$$

where[3]

H_n is a normalized hermite orthogonal function,

Y_K^M is a normalized spherical harmonic,

$\vec{x}$ is the momentum of relative motion between the centers of mass of the initial diatomic molecule and the initial atom,

ρ_1 is the equilibrium distance between the atoms of the initial molecule.

For the final (product) configuration a similar result obtains by replacing $\vec{\xi}$ by $\vec{\eta}$, $\vec{x}$ by $\vec{y}$, a_1 by a_2, etc., and making the appropriate identifications.

<u>The Reaction Probability</u>

The average time rate of change of probability of chemical change is taken to be the average over all initial (reactant) states and the sum over all final (product) states of the time rate of change of probability of transition. Explicitly, we have employed the expression

$$\dot{R} = \frac{(2\pi/\hbar)\sum_{\substack{K,n\\L,m}} e^{-(\sigma_1/kT)}\, K(K+1)\, e^{-(nh\nu_1/kT)} \int d\vec{x}\, e^{-(x^2/2\mu_1 kT)}\, S}{\sum_{K,n} (2K+1)\, e^{-(\sigma_1/kT)} K(K+1)\, e^{-(nh\nu_1/kT)} \int d\vec{x}\, e^{-(x^2/2\mu_1 kT)}} \qquad (3)$$

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(3) See S. Golden and A. M. Peiser, J. Chem. Phys. 17, 630, (1949) for a detailed definition of many of the quantities that appear in Eq. (2).

where

$$S = S(\vec{x},K,L,m,n) = \int d\vec{y} \quad \delta(E_i - E_f) \quad \sum_{M=-K}^{+K} \sum_{N=-L}^{+L} |\langle \psi_f | \mathcal{H}' | \psi_i \rangle|^2,$$

which may be recognized as being related to the transition probability summed over all degenerate energy states. Here $\mathcal{H}'$ denotes the perturbation potential that is assumed to be responsible for the chemical transition

<u>The Matrix Elements</u>

To evaluate Eq. (3) we have been obliged to make rather drastic approximations to the matrix elements implicit in $S(\vec{x},K,L,m,n)$. These approximations are twofold: (1) the perturbation potential $\mathcal{H}'$ is assumed to be singular at some specific configuration of the reacting system; (2) the molecular wave functions are sufficiently localized in configuration space to permit an additional singular approximation to be made when integrations are performed over the pertinent coordinates. (One may note that in the cases of symmetry such as obtains in ortho-para hydrogen transitions, these approximations exclude transitions in which the vibrational state of the products is different from that of the reactants. This approximation was examined in some numerical detail and appeared to be quite reasonable.)

The first approximation amounts to evaluating each wave function in the expression $3(\vec{x},K,L,m,n)$ at some configuration indicated by the nature of the perturbation potential $\mathcal{H}'(\vec{\xi},\vec{\eta})$ and assuming that

$$\int d\vec{\xi}\,\mathcal{H}'(\vec{\xi},\vec{\eta}) = \int d\vec{\eta}\,\mathcal{H}'(\vec{\xi},\vec{\eta}) = \mathcal{V}_0 . \tag{4}$$

For simplicity, we omit the details and present the result obtained after an integration over the orientations of the initial relative momentum vector is carried out:

$$\int d\Omega_x \; S = \frac{16\pi^2 V \mathcal{V}_0^2 \mu_2 y}{h^3} \quad (2K+1)^{\frac{1}{2}}(2L+1)^{\frac{1}{2}} \quad G_{KL}(x,y), \tag{5}$$

where

$G_{KL}(x,y) =$

$$\sum_j \left\langle (2K+1)^{\frac{1}{2}} P_K(z) \middle| \sum_k (k+\tfrac{1}{2}) \; |A_k(x)|^2 \, j^2_{k+\frac{1}{2}} \; (xR_1/\hbar) \; P_k(z) \middle| \; (2j+1)^{\frac{1}{2}} \; P_j(z) \right\rangle$$

$$\cdot \left\langle (2j+1)^{\frac{1}{2}} P \; (z) \middle| \sum_m (m+\tfrac{1}{2}) \; |B_m(y)|^2 \; j^2_{m+\frac{1}{2}}(yR_2/\hbar) \; P_m(z) \middle| \; (2L+1)^{\frac{1}{2}} P_L(z) \right\rangle$$

and $B(y)$ is a function analogous to $A(x)$, except that the general triatomic system will require possibly different elastic scattering potentials for the reactants and the products; R_1, R_2 are the values

of the distances between the centers of mass of the atom and the molecule, viewed as reactants or products respectively, for which the perturbation potential is assumed to have its singular value. Note that y is related to x through the conservation of energy in the transition.

To carry out the computation of G_{KL} it is necessary to specify R_1 and R_2 as well as the elastic scattering potentials of the reactants and the products (which determine $A(x)$ and $B(y)$, respectively). To determine the elastic scattering potential between hydrogen atoms and hydrogen molecules, we have estimated the electronic energy of interaction at various configurations of the system by means of the Heitler-London approximation.(4) Those configurations were selected that resulted in expressions for the interaction energy in terms of the $^1\Sigma$ and $^3\Sigma$ states for molecular hydrogen. It was found that a considerable portion of those configurations which were examined could be fitted to the form

$$V(r) = 2.33 \times 10^{-11} e^{-2.11 r^2} \text{ ergs}$$

where r, the distance between the center of mass of the molecule and the atom, is expressed in Angstrom units. This result is quite similar to that which has been found by Amdur, as a result of

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(4) F. London, Probleme der modernen Physik (Sommerfeld Festschrift), (S. Herzel, Leipzig, 1928), p. 104; Zeits. F. Elektrochemie, 35 552(1929).

scattering experiments.[5]

In the computations which we have made, R_1 and R_2 were taken to be one-half the equilibrium distance between the molecular hydrogen atoms. (We now feel that this choice, which corresponds to a coincidence of the free hydrogen atom and one of those associated with the molecule, is not a very good one and that more important contributions to chemical transition may come from regions farther removed, where the wave functions are appreciably larger in magnitude.)

We have unfortunately been unable to obtain anything more than the crudest estimate of $\mathcal{V}_0$. From Eq. (4), it can be seen that $\mathcal{V}_0$ is essentially the integrated interaction energy in the region assumed to be important for chemical transitions. For the region which we have assumed to be important, we have estimated this quantity from the repulsive energy between two hydrogen atoms. Using the repulsive portion of the Morse curve, we have been able to obtain the value $\mathcal{V}_0 = 2.13 \times 10^{-36}$ ergs cm^3. Taking the $^3\Sigma$ repulsive state of hydrogen, and restricting the region of importance to about 0.2 Angstrom units, $\mathcal{V}_0 = 13.1 \times 10^{-36}$ ergs cm^3. Clearly, $\mathcal{V}_0$ depends strongly upon the form assumed for the interaction between hydrogen atoms at small distances of approach. As indicated by the two aforementioned values, the rate calculated will change by a factor of about fifty, depending on which of these values is employed.

The foregoing results may be incorporated into the expression for the rate of chemical transition and a result is obtained for the matrix elements apart from factors due to the nuclear wave functions.

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(5) I. Amdur, J. Chem. Phys., 11, 157 (1943).

The Nuclear Weights

Because the hydrogen atoms possess nuclear spin, it is necessary to take into account the matrix elements that involve the nuclear wave functions. The nuclear wave functions that must be used depend upon the symmetry or anti-symmetry of the rotational states of the reactants and products, respectively. Inasmuch as the energy states of the system are degenerate in the nuclear states, it is necessary to sum over all such degenerate states, in accordance with the weight determined from the nuclear matrix elements.

We carried out the counting operation involved but omit the details. For the transitions of para-hydrogen to ortho-hydrogen we find that the rate expression should be multiplied by (3/4). Similarly, a computation for transitions of ortho-deuterium to para-deuterium gives the factor (1/4).

Results

We have combined the foregoing considerations and have computed the rate of conversion of para-hydrogen to ortho-hydrogen at temperatures in the vicinity of 1000°K.

With the approximations stated above, Eq. (3) for the average reaction probability can be reduced to (numerical) quadratures. For ortho-para transitions of any of the isotopes of hydrogen, we obtain for the rate constant associated with the rotational states

K and L,

$$k_{KL} = \frac{32\ V_0^2\ m}{9\pi^{3/2}\ \hbar^3 \rho}\ N_0\ (3\sigma/kT)^{5/2}$$

$$\cdot \int_0^\infty u\ du\ (2K+1)^{\frac{1}{2}}(2L+1)^{\frac{1}{2}}\ G_{KL}\left(\frac{2\hbar u}{\rho}, \frac{2\hbar v}{\rho}\right) \times e^{-(3\sigma/kT)\ u^2} \qquad (6)$$

where N_0 = Avogadro's number,

$u = x\rho/2\hbar$,

$v = y\rho/2\hbar$, and is related to u by conservation of energy,

σ = the rotational constant for the molecule.

The overall rate constant is given by

$$k\ (\text{cc.g.mol}^{-1}\text{sec}^{-1}) = \sum_{K,L} k_{KL}\ e^{-\sigma K(K+1)/kT} \qquad (7)$$

Eq. (7) must be multiplied by a factor of 2 to take into account the symmetry of the molecule, and by the nuclear factor discussed above.

At 1000°K we obtain for para-ortho hydrogen,

$$k = 0.018 \times 10^{+12}\ \text{cc. g.mol}^{-1}\ \text{sec}^{-1},$$

with the larger of the two values of V_0 given above. This result is approximately one-hundredth of the observed value.[6] The rate of conversion at other temperatures, relative to that at 1000°K, is

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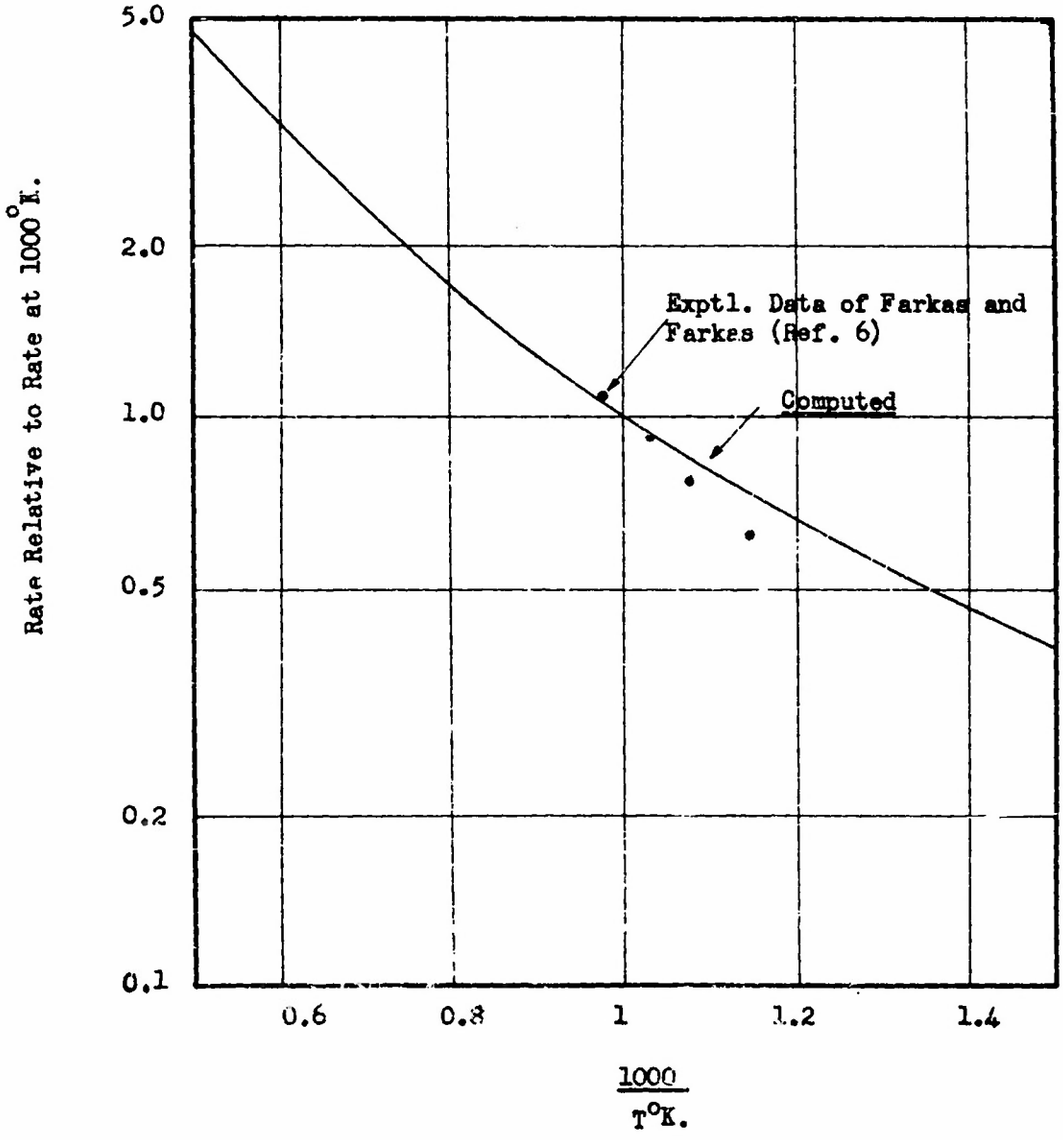
(6) A. Farkas and L. Farkas, Proc. Royal Soc., 152A, 124 (1935).

Figure 2

Rate of Para → Ortho Conversion of Hydrogen

(Relative to Rate at 1000°K) as a

Function of Temperature



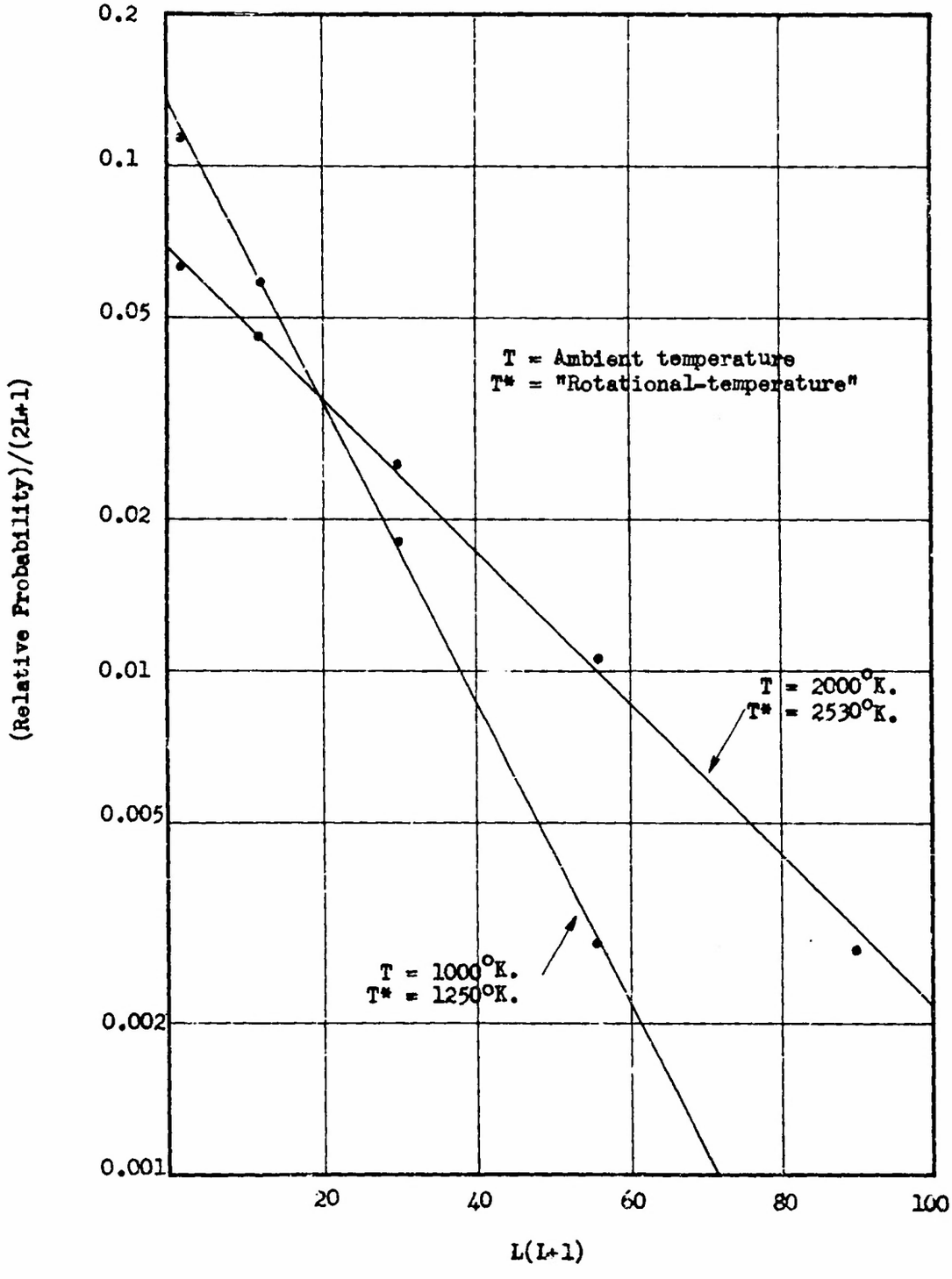
plotted in Figure 2. It will be noted that the temperature coefficient leads to an apparent activation energy of 4.6 kcal/g.mol. while the experimental data lead to a value of 6.0 kcal/g.mol.

In Figure 3, we present plots of the distribution of final products according to their rotational state (obtained by summing the terms in Equation (17) over all K for a fixed L). The plot presented gives the relative probability divided by (2L+1), the latter quantity representing the statistical weight of the state. This sort of a plot permits the ready determination of the "rotational-temperature" of the products of reaction, which has been indicated. It is interesting to note that no very large difference is obtained between the "rotational-temperature" and the actual temperature of the reaction. Presumably this result is associated with the fact that the overall reaction is virtually isoenergetic.

We have been able to estimate the rate of the ortho-to-para conversion of deuterium directly in terms of the results for hydrogen. Referring to Equations (6) and (7), it will be noted that a change in the mass of the system produces a change in the expression for the average rate constant. Some of the dependence upon the mass is implicit in the function $G_{KL}(x,y)$, since the density-in-energy $\rho(E_k)$ will depend upon the mass of the system. We have examined this dependence and it appears that except for very large or very small energies of relative motion, $|A_n(k)|^2$ will vary inversely as

Figure 3

Distribution of Initially Formed Ortho-Hydrogen Molecules.



the square of the mass of the system. Accordingly, since the rotational constant σ depends inversely on the mass of the system, we find that the rate constant at temperature T for any isotope of hydrogen is, apart from nuclear factors,

$$k\ (T;\ \text{isotope}) = \left(\frac{m_h}{m}\right)^3 k\left(\frac{m}{m_h}\ T;\ \text{hydrogen}\right), \qquad (8)$$

where m and m_h are the respective masses of the isotopic atom and the hydrogen atom.

For the ratio of the rate constants for deuterium and hydrogen we have obtained at 1000°K.

$$\frac{\text{Rate (ortho-to-para deuterium)}}{\text{Rate (para-to-ortho hydrogen)}} = 0.20,$$

while Farkas and Farkas (Ref. 6) report the value 0.24.

If the approximations leading to Equation (8) are justifiable, it follows that the apparent energies of activation for the ortho-para reactions of the isotopic species of hydrogen should vary inversely as the mass of the isotope. This conclusion is at variance with the usually held opinion regarding these quantities. However, a careful examination of the existing data available for the ortho-para deuterium conversion reveals that the data are incapable either of confirming or denying this result. Indeed, there seem to be no reliable data for this reaction that will permit its apparent energy of activation

to be determined directly, with an accuracy comparable to that which is available for the corresponding hydrogen reaction.

Discussion

We have indicated throughout the body of this report the nature of the approximations which we have been obliged to make, and thereby offered an indication of the lines of investigation which might profitably be undertaken in the future. The major points are listed below.

(1) Of paramount importance to this kind of investigation is the notion of chemical species. We feel that an investigation of suitable and consistent criteria for identifying the reactants and products of a chemical change is essential. Many of the difficulties which have been encountered in the present investigation could be avoided (formally, at least) were such criteria available. The question of the proper wave functions to be used, for instance, would undoubtedly be clarified considerably.

(2) The nature of interaction potentials between atoms and molecules for small distances of approach is a necessary adjunct to the study of reaction rates. Here the problem appears to be of considerable numerical complexity, but well-defined conceptually.

(3) Special attention should be devoted to application of modern automatic computing machinery to evaluate sums and integrals developed from the theory. It seems unlikely that reasonably accurate

results from a more accurate theory will be obtained unless the numerical computations are obtained from machines. The effort is too great and time consuming when the computations are done by desk machine.

(4) Particular attention should be devoted to the design of experiments that will furnish more elementary information on chemical transitions. This would avoid the considerable difficulty of averaging over the distribution of states of the reactants in order to obtain a result comparable with conventional experimental results. The experimental techniques employed by the physicist in similar cases seems to be most appropriate from the theoretical point of view.

Inasmuch as some of the conclusions that are obtainable from the approximate theory which we have employed lead to results at some variance with conventional theories, it would be especially helpful to test these conclusions. This would indicate, at least, the directions to be undertaken in the nature of approximations which can be made.

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