THE MEASUREMENT OF NEUTRON DIFFUSION PARAMETERS IN HEAVY AND LIGHT WATER CONCENTRATIONS BY THE PULSED NEUTRON TECHNIQUE*

by

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INTRODUCTION

A study of diffusion and thermalization of neutrons in media with varied geometrical configurations is a contributing factor in the solution of the many problems in neutron physics. The data obtained through these investigations are essential for a thorough understanding of the neutron thermalization processes which take place in nuclear reactors. The diffusion and thermalization of neutrons is a function of the physical state and the temperature of the system; any change in these factors alters the magnitude of the effective cross section and the spectrum of energies transferred during the individual interactions.

For the past few years many investigations of the neutron properties of moderators have been carried out using the pulsed-neutron technique (1-5). A burst of neutrons is injected into a finite volume of moderator and the time dependence of the thermal neutron flux is measured. In general, the neutron lifetime in an ideal moderator may be considered in three separate steps: moderation, thermalization, and diffusion. The moderation process applies to the energy region greater than approximately 0.5 ev where the atoms of the moderator are considered as free. The thermalization process represents the slowing down below this energy where the neutrons interact with the molecules and crystals of the moderator rather than free atoms. The diffusion period follows after a thermal equilibrium is established. In the thermalization region, the neutron velocity spectrum reaches a constant form, but decreases in amplitude with time due to absorption and leakage from the moderating medium. Most investigations using the pulsed-neutron method examine the neutron life-

time during the diffusion period in moderator systems such as H_2O, D_2O, graphite, beryllium, etc. Excellent summaries of possible experiments utilizing this technique have been presented by von Dardel and Sjostrand (6) and Beckurts (7,8).

The experiments described measure the diffusion parameters of heavy and light water mixtures. A moderator sample in a cylindrical container receives a burst of fast neutrons during a pulse from the neutron generator. After thermalization, the neutrons are either absorbed or leak out of the system and the neutron population of the fundamental mode distribution decays exponentially with a decay constant expressed as

$$\alpha = \Sigma_a v + D_o B^2 - CB^4$$

where

v = velocity of the thermal neutron

Σ_a = macroscopic thermal absorption cross section

D_o = thermal diffusion coefficient

B^2 = geometrical buckling

C = thermal diffusion cooling coefficient

The decay constant of the neutrons in the moderator mixtures is measured with a time analyzer for each of a series of five or more cylinders. The measured decay constants are plotted versus the bucklings. Analysis of the resulting curve yields directly the diffusion coefficient, the product of the neutron velocity and macroscopic thermal absorption cross section, the thermal diffusion cooling coefficient, and indirectly

the diffusion length.

The experimental procedure is quite straightforward; accurate results depend on three important measurements:

1. Determination of the true fundamental mode decay constant, α.
2. Accurate calculation of the buckling, B^2.
3. Fitting of α versus B^2 curve.

LITERATURE REVIEW

There are at present no available data on the diffusion parameters of heavy and light water mixtures. This review, therefore, will discuss the available published data on the diffusion parameters of pure H_2O and D_2O as moderators at or near room temperature.

Since the initiation of the study of neutron parameters with the pulse technique, a number of measurements on H_2O at room temperature have been made. These results are summarized in Table I. In Table II, the diffusion parameters for H_2O obtained by other methods are summarized. Considerable variation in results is noted. In six of the more recent measurements (9-14), the range of values of the diffusion coefficient D varies from 35,400 to 37,405 cm^2/sec and the range of values of the diffusion cooling coefficient from 2,900 to 5,162 cm^4/sec. Experiments (12,15,16) to determine the dependence of C and D_o as a function of the temperature of the moderator medium have been made recently. The experiments of Dio (12) were carried out for temperatures less than 100°C while the temperatures used by Antonov et al. (15) are from 0°C to 266°C. McClure (16) measured the diffusion parameters for water and ice at 1.0°C and -19°C.

Like H_2O, a considerable number of measurements have been made on the diffusion parameters in D_2O. Also, the present published experimental data on heavy water is insufficient, since a number of these experiments were carried out over a limited range of bucklings. Measurements (17-19) of the diffusion parameters for heavy water were made as

TABLE I

Diffusion Parameters for H_2O by Pulsed Neutron Method

Reference	$D_o(cm^2sec^{-1})$	$C(cm^4sec^{-1})$	σ_a^H(mb)	L(cm)	B^2 range cm^{-2}	Temp.(°C)
Manley et al. (1)	--	--	345	--	Large Geometry	22
von Dardel and Waltner (3)	--	--	321 ± 5	--	Infinite Geometry	22
Scott et al. (4)	38500 ± 800	--	320 ± 8	2.85 ± 0.05	0.006 → 0.018	22
von Dardel and Sjostrand (5)	36340 ± 750	7300 ± 1500	333 ± 3	2.725± 0.03	0.1 → 0.7	22
Antonov et al. (24)	35000 ± 1000	4000 ± 1000	329 ±10	2.7 ± 0.1	0.09 → 0.93	22
Bracci and Coceva (25)	34850 ± 1100	3000 ± 1000	337 ±10	2.66 ± 0.11	0.09 → 0.96	22
Meads et al. (26)	--	--	335 ± 4		Infinite Geometry	22
Campbell and Stelson (27)	34800	0	327	2.69	0.08 → 1.1	22
Stooksberry and Marshall (28)	--	--	330 ± 8	--	Large Geometry	22
Dio (12)	35450 ± 600	3700 ± 700	328 ± 6	2.715± 0.06	0.093 → 0.87	22
Kuchle (9)	35400 ± 700	4200 ± 800	326 ± 6	2.72 ± 0.08	0.11 → 0.75	22
Lopez and Byster (10)	37503 ± 366	5116 ± 776	325 ± 1.6	2.828± 0.016	0.0137 → 0.591	26.7
	36892 ± 400	--	--	2.803± 0.016	0.0137 → 0.591	22
Bretscher (23)	36985 ± 1630	5021 ± 2490	322 ±17	2.82 ± 0.09	0 → 0.60	26
Beckurts (29)	34820 ± 720	3650 ± 400	--	--		22
Antonov et al. (13)	35000 ± 1000	4000 ± 1000	--	--	0.050 → 1.00	21
Dlouhy and Kvitek (14)	34600 ± 800	4000 ± 800	--	--		20
Kovel (30)	37405 ± 422	5162 ± 2144	--	--	0.1085 → 0.248	24.4
McClure (16)	29670 ± 2640	92 ± 3550	--	--	0.150 → 0.790	0

TABLE II

Neutron Diffusion Parameters for H_2O by Other Methods

Reference	$D_o(cm^2sec^{-1})$	$C(cm^4sec^{-1})$	$\sigma_a^H(mb)$	L(cm)	Method	Temp.(°C)
DeJuren et al. (35)	--	--	--	2.763 ± .015	DLM*	22
Hammermesh et al. (36)	--	--	329 ±4	--	BPE**	22
Harris et al. (37)	--	--	332 ±7	--	BPE	22
Barkow et al. (38)	--	--	--	2.71 ± .02	DLM	22
Baker (39)	--	--	327 ±4	--	BPE	22
Reier et al. (40)	--	--	--	2.72 ± .007	DLM	22
Reier (41)	37618 ± 205	--	328 ±6	--	BPE	22
Rockey et al. (42)	--	--	--	2.838 ± .018	DLM	22
Starr et al. (11)	35850 ± 100	2900 ± 350	326.9±1.6	2.754 ± .008	DLM	21

*Diffusion Length Measurement

**Boron Poisoning Experiments

TABLE III

Diffusion Parameters of Thermal Neutrons in D_2O

Reference	$D_o \times 10^{-5}$ (cm^2/sec)	L(cm)	$C \times 10^{-5}$ (cm^4/sec)	Temp.(°C)	λ_{tr}(cm)
Auger et al. (17)	--	--	--	22	2.4 ± 0.1
Sargent et al.(19)	--	171 ± 20	--	22	--
Kash and Woods (20)	--	--	--	20	2.52 ± 0.04
Raievski and Horowitz (31)	2.00 ± 0.05	--	--	13	2.45 ± 0.07
Sjostrand (32)	--	--	3.5 ± 0.8	20	--
Bauman (33)	--	--	--	20	2.52 ± 0.03
Starr and de Villiers (34)	--	--	5		--
Ganguly et al.(21)	2.00 ± 0.04 2.08 ± 0.05	-- --	2.95 ± 0.43 3.72 ± 0.50	10 20	-- --
Kussmaul and Meister (22)	2.0 ± 0.009	--	5.25 ± 0.25	22	--
Present Investigation	1.966 ± 0.013	--	3.56 ± 1.08	21	--

early as 1946, using the extrapolated end-point method. In 1953, the diffusion parameters were measured by Kash and Wood (20) using the boron poisoning method.

The pulsed-source method has been used by a number of investigators (21,22) to measure the diffusion parameters for D_2O moderators of finite geometry. The results of these measurements are summarized in Table III.

SOME COMMENTS ON TRANSPORT THEORY

Use of the pulsed neutron technique for measuring thermal diffusion parameters has become widespread since its introduction. There are, however, considerable theoretical and experimental uncertainties remaining in its application. Nelkin (43) has given a theoretical basis for the interpretation of a measured decay constant in a non-multiplying medium.

After the injection of a burst of neutrons into a finite moderator, the thermal neutron density in the moderator decays exponentially in time. If the absorption cross section for the moderator varies as the inverse of the velocity and the dimensions of the moderator are greater than several mean free paths, the decay constant, α, may be approximated by an equation involving the mean lifetime α_o; the diffusion constant, D_o; the diffusion cooling constant, C; and the moderator buckling, B^2. The equation for α is:

$$\alpha = \alpha_o + D_o B^2 - CB^4 + \text{higher terms}, \qquad (1)$$

where, for cylinders, $B^2 = [2.405/(r + d)]^2 + [\pi/(H + 2d)]^2$. H and r are the dimensions of the system and d is the extrapolation distance.

Nelkin (43) outlined an improved theoretical derivation and justification for the use of Eq. (1), starting from the multivelocity transport equation. The approach solves for α, after a pulse, by obtaining an exact solution to the Fourier-transformed transport equation and then expanding α and the neutron spectrum in terms of a Fourier transform variable, B. In his derivation, as in others, the absorption cross

section is assumed to be 1/v, resulting in a constant α_o.

For the isotropic scattering case, a one dimensional spatial variation with no regeneration of neutrons in the medium is assumed. Also assumed is a source of neutrons only at t = 0 with isotropic scattering. The exact solution to the transport equation was obtained by taking the Fourier transform in terms of a transform variable, B, and expanding the decay constant and the flux in terms of B^2.

The energy distribution in the moderator is considered an equilibrium Maxwellian distribution at a moderator temperature T of the form

$$M(E) = E/T^2 \exp(-E/T) . \qquad (2)$$

T is measured in energy units.

When the transport equation is solved for various orders, it is found that the constants in the α expansion have physical significance. For absorption, varying as the inverse neutron velocity, v, the solution in the case of zero order yields

$$\alpha_o = \Sigma_a v \quad \text{and} \quad \phi_{o,o} = M(E) . \qquad (3)$$

α_o represents the inverse of the mean life for the thermal neutrons in an infinite system ($B^2 = 0$) and Σ_a is the macroscopic absorption cross section for the thermal neutrons. α_o is generally obtained by extrapolating the plot of the decay constants as a function of geometrical dimensions (bucklings) to infinite dimensions ($B^2 = 0$).

The first order solution to the transport equation gives the coefficient of the second term in α as

$$D_o = \frac{\int_0^\infty [3\Sigma_s(E)]^{-1} M(E)dE}{\int_0^\infty v^{-1}M(E)dE} \qquad (4)$$

for $\Sigma_a = \alpha_o/v$. Thus, the second term in the expansion of α, D_oB^2, represents the neutron leakage from the moderator based on the fact that the neutrons have reached a Maxwellian distribution. A Maxwellian distribution is considered to exist in the moderator if absorption is very small; the absorption cross section varies as the inverse velocity; and the dimensions of the system are sufficiently large to contain the neutrons long enough so that an asymptotic solution is obtained.

For the second order solution to the transport equation, the coefficient, C, of the third term in the α expansion consists of two physically distinct parts, $C = C_D + C_T$.

$$C_D = \frac{\int_0^\infty [1/3\Sigma_s(E) - D_o/v]\ \phi_{2,o}(E)dE}{\int_0^\infty v^{-1}M(E)dE} \qquad (5)$$

and

$$C_T = \frac{\int_0^\infty [1/3\Sigma_s^2(E)]\ [4/15\Sigma_s(E) - D_o/v]\ M(E)dE}{\int_0^\infty v^{-1}M(E)dE}. \qquad (6)$$

In C_D, $\phi_{2,o}$ represents the shift of the neutron spectrum to lower energies due to the increase in leakage rate with neutron velocity.

This phenomenon is called "diffusion cooling" by von Dardel (5) and represents the dominant contribution to the B^4 term of the decay constant. This value of C_D has been given in an exact solution by multi-velocity diffusion theory, and an exact solution using the model of a heavy monatomic gas for the energy transfer has been given by Hurwitz and Nelkin (44). Also, variational solutions have been given by Nelkin (45) and by Singwi and Kothari (46).

The term C_T, Eq. (6), represents the deviation from diffusion theory; e.g., a non-diffusion correction to the order B^4.

The extrapolation distance, d, is the distance from the edge to a point outside the moderator system where the asymptotic neutron flux would go to zero if the flux, both inside and outside of the moderator, were represented by the same equation. The values of d can be calculated explicitly for the one velocity Milne problem (47,48) where thermal neutrons are diffusing from an infinite plane interface into a vacuum. The vacuum has the property that any neutron which enters never returns to the moderator medium. Thus, the vacuum is equivalent to a perfect absorbing medium. In an actual experiment this condition is approximated by covering the surface of the diffusing medium with either cadmium or cadmium covered with borated paraffin. For anisotropic scattering in a medium with weak absorption

$$\Sigma_{tr} d = 0.710 \quad \text{or} \quad d = 0.710\, \lambda_{tr} , \qquad (7)$$

where Σ_{tr} and λ_{tr} are the macroscopic transport cross section and transport mean free path respectively. Although Eq. (7) does not strictly

apply for small systems, it has been used in lieu of something better. In the case of water, where the cross section is energy dependent, Sjostrand (49) has found for slab geometries, even as small as two to three centimeters thick, that the deviation from Eq. (7) was very small. His calculations for the extrapolation length were based on an infinite slab using the P_3 approximation of the spherical harmonics method.

Multi-group calculations of d for infinite slab geometries have been made by Gelbard and Davis (50) and by Nelkin (51). Their calculations indicate, in the limit as the buckling approaches zero, that d approximately equals 0.76 λ_{tr}. Nelkin states that the method of Gelbard and Davis (50) is quite satisfactory for slab geometry, but the extension to three dimensional systems such as a right circular cylinder is not very satisfactory due to the difficulty in calculating the corrections to the extrapolation distance.

In view of the difficulty of calculating the proper extrapolation for right circular cylinders, $d = 0.710\ \lambda_{tr}$ is used for the calculation of B^2 in this experiment.

To summarize, the basic assumptions made in analyzing a pulse of fast neutrons are:

1. The system is large enough that after a sufficiently long time an asymptotic solution is obtained. The thermalized neutrons decay exponentially in time with a decay constant of the form $\alpha = \alpha_o + D_o B^2 - CB^4 + \ldots$ higher terms.

2. The asymptotic solution has a buckling, B^2, corresponding to decay of a particular mode of an infinite system.
3. The spatial mode is characterized by $\Delta\phi + B^2\phi = 0$, where $\phi \to 0$ for some extrapolated boundary.
4. The extrapolation distance, d, equals some constant, c, times the transport mean path, λ_{tr}, that may be adjusted to the diffusion constant, D_o, by a least squares fit with $\lambda_{tr} = 3D_o/v$, neglecting other corrections. This gives an approximation to B^2 that would correspond to a particular mode of an infinite system.

The conventional interpretation of pulsed measurements is obtained by assigning a geometric buckling, B^2, to each moderating sample, and plotting the decay constant as a function of B^2. A least squares fit of Eq. (1) yields the diffusion coefficient and the diffusion cooling coefficient as well as the capture cross section. The buckling is defined as the lowest eigenvalue of $\Delta\phi + B^2\phi = 0$, subject to the boundary condition that ϕ goes to zero at an extrapolated boundary at 0.71 λ_{tr} outside the actual boundary. The comparison of infinite medium theory to the present experiments depends on the validity of assigning an equivalent infinite medium buckling to a finite system. Nelkin (43) states that the usual interpretation of the pulse decay measurements in terms of infinite medium parameters is quite accurate if care is taken in assigning values to the extrapolation distance when the assignment of a buckling is made to a particular sample.

EXPERIMENTAL ARRANGEMENTS

The fast neutron source used in present investigations is a 250-kv Cockcroft-Walton accelerator. The accelerator was constructed at Virginia Polytechnic Institute and modified by the author for use in present experiments (Appendix I). The moderator assembly was located directly beneath the target and at right angles to the accelerator beam. The thermal neutron leakage for all the moderator bucklings was detected with an enriched Li^6I detector located beneath the center of the moderator. The experiments were carried out at 21°C with a temperature fluctuation of less than ±0.5°C for any one particular decay measurement. A schematic diagram of the important features of the system is given in Figure 1. Details of the system are given below.

A. Accelerator

The Cockcroft-Walton used is a positive ion accelerator with a deuterium source. The target was a zirconium "drive-in" target 20 mils thick and one and one-eighth inches in diameter. A radio frequency ion source of the Moak type (52) supplies the deuterium ions for the accelerator. By modulating the deuteron beam, a burst of fast neutrons having approximately 2.5 mev energy (53) is produced by the $_1H^2(d,n)_2He^3$ reaction. The modulating system consists of a pair of electrostatic deflection plates located in the terminal end of the accelerator tube. The operating range of the accelerator was in the range 125-150 kv throughout the experiments.

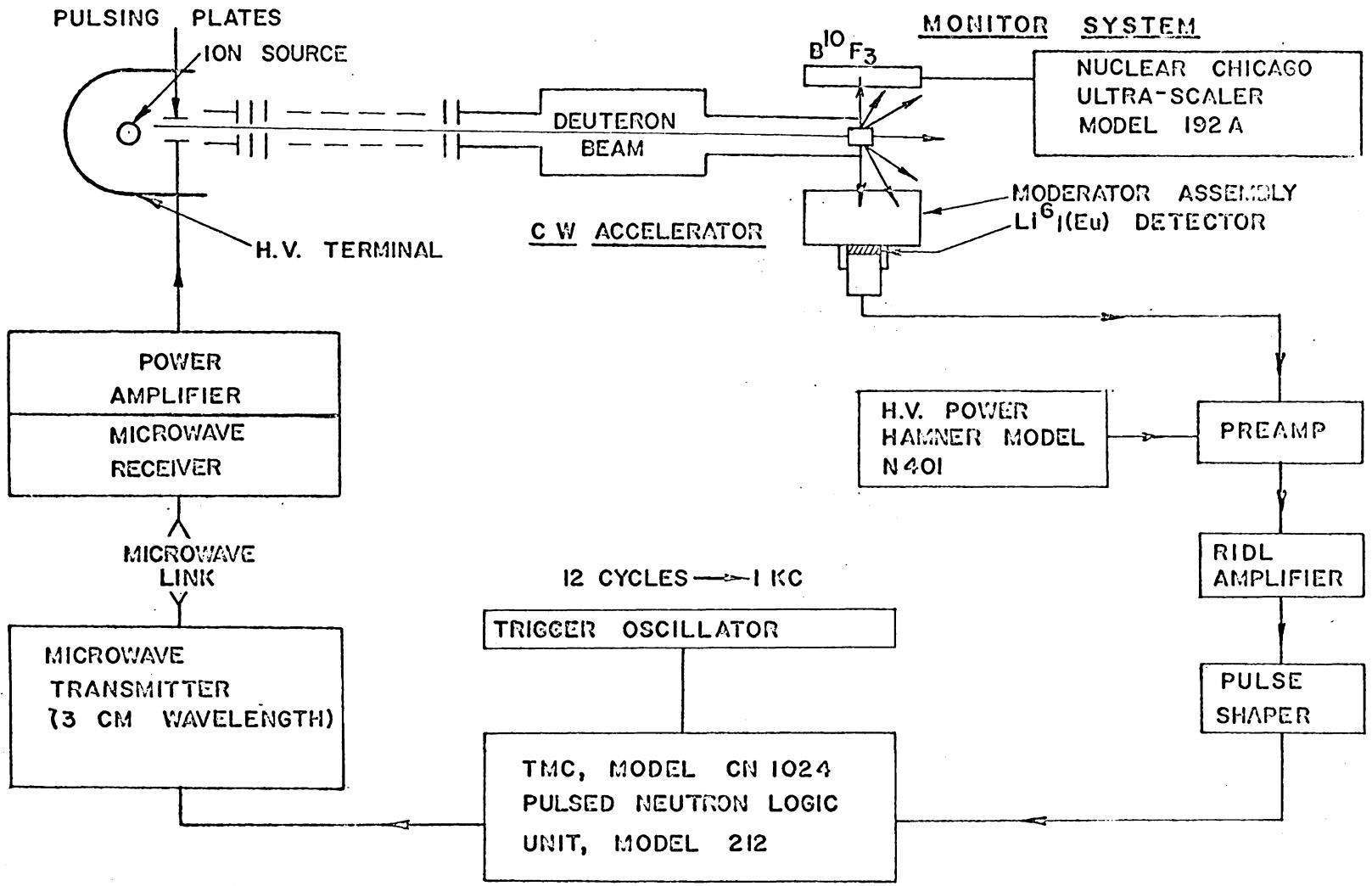

Figure 1. Block diagram of the equipment.

A pressure of 10^{-6} torr was maintained within the accelerator during operation. This pressure is monitored with a VG1A ionization gauge and the system is protected by a "drop-out" relay which turns off the diffusion pump should the pressure in the system become greater than 10^{-4} torr. The system remains off until the relay is manually reset.

B. Detectors

Two neutron detectors were employed: a monitor giving a number proportional to the total number of counts and a thermal neutron detector which measures the thermalized neutrons that leak from the moderator. The monitor system consists of a $B^{10}F_3$ counter surrounded by three inches of paraffin in an aluminum can. The aluminum was subsequently surrounded by a layer of 30 mil cadmium. The $B^{10}F_3$ counter was one inch in diameter with an active length of ten inches (manufactured by the Reuter-Stokes Company, Model RSN-7A).

The thermal neutron detector consists of an Li^6I scintillating crystal (Harshaw Chemical Company) approximately four centimeters in diameter and two millimeters thick. This crystal was optically connected to a two-inch Dumont 6292 photomultiplier tube. The Li^6I scintillation crystal is black to thermal neutrons. It responds to neutrons by the reaction $Li^6(n,\alpha)He^3$ where the disintegration products lose their energy in the crystal and cause a large, well-defined scintillation pulse which can be discriminated from pulses caused by γ rays (54). The detector used had approximately 15 percent resolution.

C. Electronic Instrumentation

To initiate a deuteron pulse on the target, a signal from a pulse generator is fed to the input of the Model 212 pulsed neutron logic unit of the Model CN-1024 analyzer (TMC). The signal triggers the analyzer and after a preset time delay, a signal is transmitted by the pulsed neutron logic unit to the input of the microwave pulsing system (Appendix I). During this time delay, the background is counted and stored in the first channel of the analyzer. A signal of a given width, determined by the microwave transmitter, is transmitted to the high voltage terminal. In the dome, the signal is received by a microwave receiver, amplified, and fed to the beam pulsing plates. A pulse of deuterons strikes the target for a length of time (100 microseconds) designated by the width of the received pulse. The time delay between the storage of the background data in the first analyzer channel and the gating of the second channel to start the analysis of the neutron burst was set for 160 microseconds. In the present measurements, 64 of the 1,024 analysis channels were used, each having an analysis time of ten microseconds. The total time elapse between channels was 20 microseconds, ten microseconds being used for data storage. The time delay between the end of the neutron burst and the gating of the first analysis channel was 60 microseconds.

The thermal neutrons leaking from the system were detected by a cadmium enclosed Li^6I detector. Only the portion of the detector in contact with the moderator container was exposed. The signal from the detector was fed through a cathode follower preamplifier into the input of an RIDL Model 30-16 amplifier. The amplifier signal was taken from

the positive pulse height selector, fed through a pulse shaper to the signal input of the time analyzer. The data was visually displayed at all times on a Tektronix-Type RN 503 oscilloscope and a permanent record of this data was printed on paper tape by a Hewlett-Packard Model 544 561B digital recorder.

The electronic instrumentation used with the $B^{10}F_3$ neutron monitor was a Model 192A Ultrascaler (Nuclear-Chicago). The Dumont 6292 photomultiplier tube, used with the Li^6I detector, was operated at 1,000 volts supplied by a current regulated Hamner, Model N401, power supply.

D. Moderator Geometry

The moderator materials were contained in aluminum right circular cylinders with dimensions such that their height to diameter ratio was approximately equal to one. The wall thickness of the cylinders was 0.159 centimeters. The cylinders were surrounded by 30 mil cadmium covers. A circular hole was cut through the center of one end of the cadmium shield so that the cap of the neutron detector could be placed directly in contact with the end of the aluminum container. The remaining portion of the detector and the photomultiplier tube were shielded with a 30 mil sheet of cadmium, and the base containing the photomultiplier was shielded with a double thickness of 30 mil cadmium.

The moderator cylinder was centered symmetrically below the target, and the top of the moderator surface was approximately three centimeters below the center of the target. Care was taken to insure that the geometric arrangement was the same for each experimental determination.

The temperature of the moderator was taken with a mercury thermometer before and after each run.

EXPERIMENTAL PROCEDURE

Measurements of the time-dependent neutron decay constant for various heavy and light water concentrations as a function of buckling (B^2) were carried out at 21°C. The decay constants for mixtures of 20, 50, and 80 percent D_2O and pure H_2O and D_2O were measured.

The measurements of the D_2O and H_2O decay constants were first run and a comparison made with other published data. This provided a check on the capability of the accelerator modifications and the reliability of the instrumentation. During all the runs, a Tektronix-Type 545 oscilloscope was used to monitor the pulsing system and a $B^{10}F_3$ neutron detector was used to monitor the neutron yield.

The D_2O samples were prepared in a dry box to eliminate, as much as possible, the contamination by H_2O vapor. The aluminum container, after being filled with D_2O, was covered with a double layer of thin plastic (Saran Wrap) and enclosed in a 30 mil cadmium shield. The height of the liquid in the container was measured to an accuracy of 0.05 centimeters. This accuracy was verified by weighing the D_2O in the sample cylinder of known radius and calculating the height using 1.104 gm/cm^3 for the density. The H_2O, D_2O mixtures were mixed percent by volume and the various samples were prepared as previously indicated.

DATA ANALYSIS

The data obtained directly from the experiment were the neutron leakage per unit time following a neutron burst. Values for the decay constant, α, were determined from the accumulated counts in the time analyzer as printed out by the digital recorder. The numerical values for α were determined by a least squares fit using an IBM 7040 electronic computer and Peierls' Method (55).

Typical neutron density decay and α versus B^2 curves are shown in Figures 2 and 3. Standard deviations for the values of the decay constant were obtained from the computer program (Appendix II).

The diffusion parameters were determined from the α versus B^2 curves using the IBM 7040 computer and a computer program that calculates D_o, C, λ_{tr}, the standard deviation of D_o, the standard deviation of C, and a corrected B^2 for a given α_o. The input for the program requires the geometric dimensions of the moderator, a fixed α_o, a trial λ_{tr}, and an average neutron velocity. Since the buckling of a moderator system depends on λ_{tr} through the extrapolation distance, the program uses an iterative procedure (21) to perform a least squares fit of the α versus B^2 data to the equation,

$$\alpha = \alpha_o + D_o B^2 - CB^4 + \text{higher terms.} \qquad (1)$$

Iteration is required since λ_{tr} is included in D_o. This can be seen directly from the buckling equation for cylinders,

$$B^2 = [2.405/(r + d)]^2 + [\pi/(H + 2d)]^2, \qquad (2)$$

where $d = 0.71\lambda_{tr} = (0.71)3D_o/v = 2.13D_o/v$.

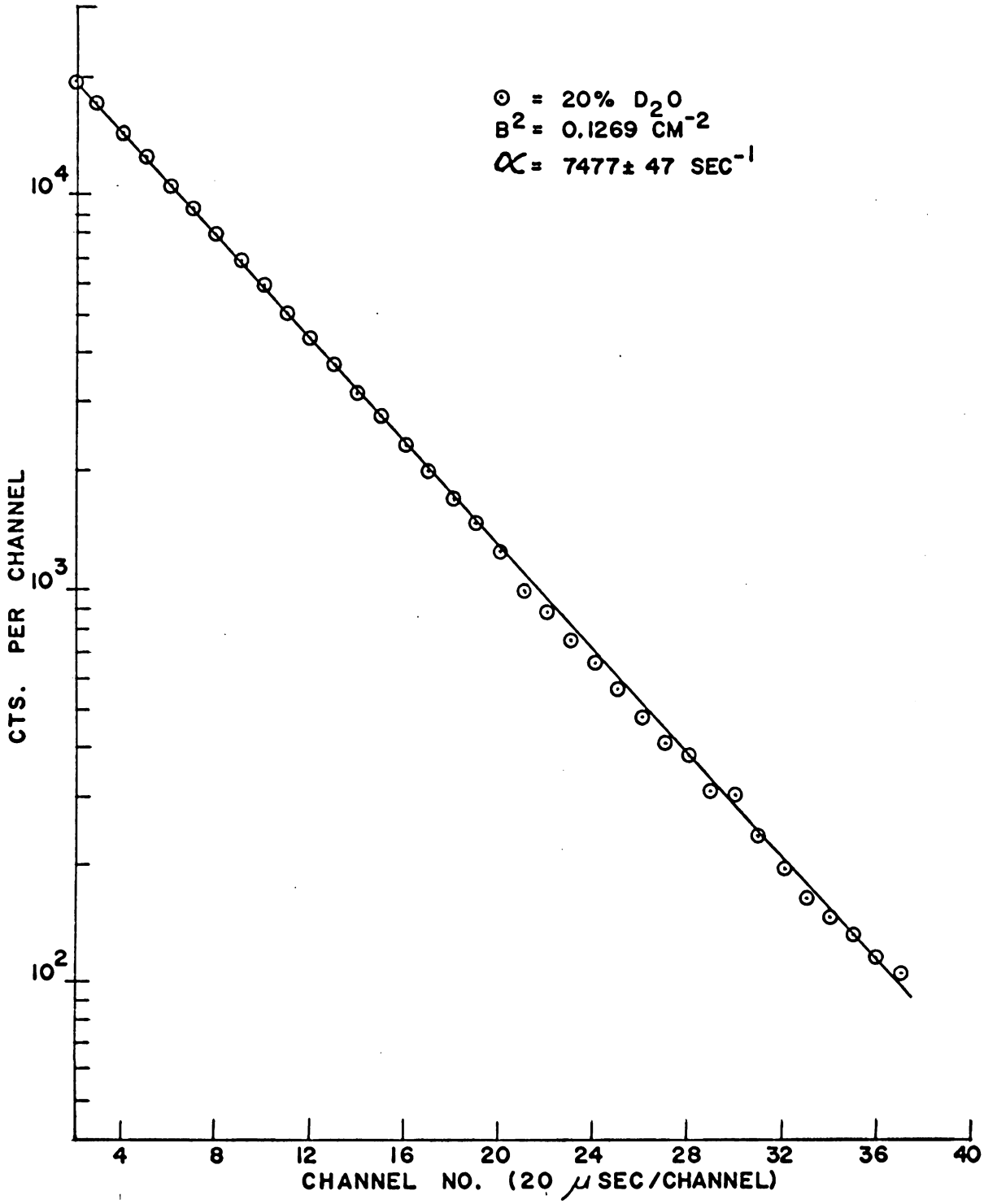

Figure 2. Neutron decay curve for 20 percent D_2O concentration (background approximately 80 counts per channel).

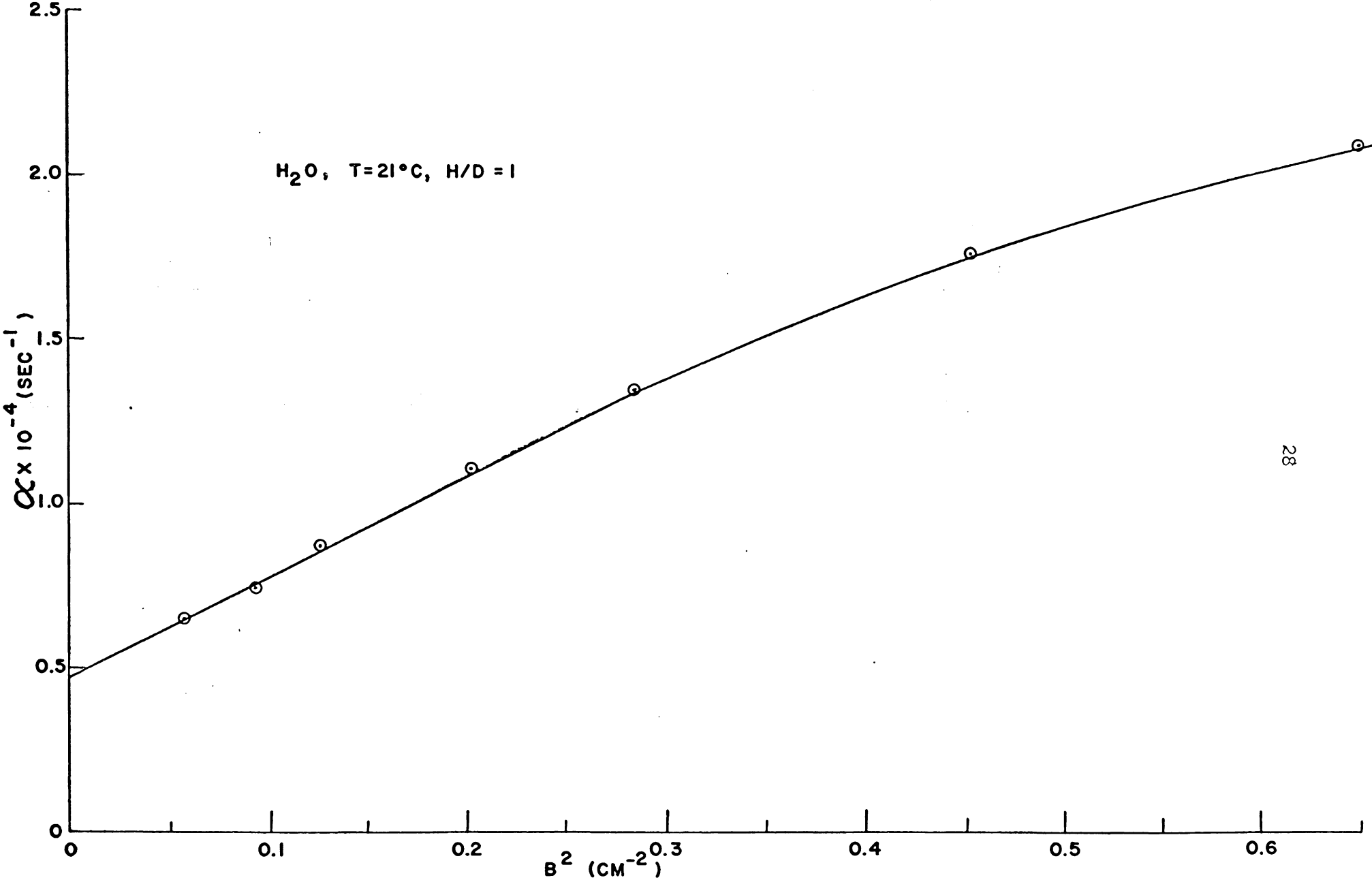

Figure 3. α versus B^2 for pure H_2O (error in α approximately equal to circle diameter).

The neutron velocity used was 2.2×10^5 cm/sec (0.025 ev) for the thermal system. The program output also gave the number of iterations necessary for a prescribed input for the initial λ_{tr}.

The initial λ_{tr} values for the H_2O - D_2O mixtures were calculated using the H^1, D^2, and O^{16} scattering cross sections. The transport mean free path

$$\lambda_{tr} = 1/\Sigma_{tr},$$

where

$$\Sigma_{tr} = \Sigma_s(1 - \mu_o) = \sum_i N_i(\sigma_s)_i(1 - \mu_o)_i.$$

Σ_s = macroscopic scattering cross section

μ_o = average value of the cosine of the scattering angle

N_i = number of atoms of i^{th} component in moderator

$(\sigma_s)_i$ = microscopic scattering cross section for i^{th} component in the moderator

The calculated values of Σ_{tr} and λ_{tr} are tabulated in Table IV. The density at 21°C, for H_2O and D_2O is 0.996 gm/cm^3 and 1.104 gm/cm^3 respectively.

The usual way to determine the diffusion parameters is to fit the experimental points by the method of least squares to Eq. (1) which is terminated after two or three terms. The higher terms are obviously not zero but the result of the analysis will depend on the buckling range covered by the experiment. From the results, it appears that for a given

TABLE IV

Calculation of Σ_{tr} and λ_{tr} for the Moderator Mixtures

Percent D_2O	$\Sigma_{tr}(cm^{-1})$	$\lambda_{tr}(cm)$	Reference
0	--	0.424	Lopez and Byster (10)
20	1.078	0.927	Calculated
50	0.710	1.407	Calculated
80	0.516	1.940	Calculated
100	--	2.52	Nash and Woods (20)

experimental error of the measured points, there is an optimum range beyond which no further accuracy is gained in determining the diffusion parameters. A feeling for the error involved may be obtained for three terms in Eq. (1) by plotting the coefficients for a decreasing number of points; e.g., dropping progressively the point of maximum buckling (56). The data presented in this report were analyzed in this manner and the criterion for the best fit of the data was determined by calculating the variance (57)

$$\beta = \sum_i (\alpha_i - \alpha_i^*)^2/(N-n).$$

α_i = measured value of decay constant at B_i^2

α_i^* = value of decay constant at B_i^2, calculated from the functional form

N = number of measurements

n = number of parameters in function

The number of points which made β a minimum were retained.

RESULTS AND DISCUSSION

Measured decay constants and corresponding bucklings are given in Tables VI, VII, VIII, IX, and X for moderators consisting of various concentrations of heavy and light water. The measurements were taken at 21°C. Table V shows the diffusion coefficient, diffusion cooling coefficient, and λ_{tr} for each of the concentrations including pure H_2O and D_2O. The data were processed by the previously indicated procedure. Figure 4 shows a plot of α as a function of B^2 for each of the concentrations. Figures 5 and 6 show D_o and C as a function of the heavy water concentration.

The data in Table V for H_2O and D_2O are in agreement with published data shown in Tables I and II in the Literature Review.

With reference to Figure 5, D_o as a function of heavy water concentration, the leakage increases with heavy water concentration. This result is expected since the "effective" mean free path in the moderator increases with heavy water concentration. A similar conclusion is drawn for the diffusion cooling coefficient by examining Figure 6.

The value of α_o, the intercept of α versus B^2 at $B^2 = 0$, was calculated for each concentration using data of Section II, $\alpha_o(H_2O) = 4768\ sec^{-1}$ (10) and $\alpha_o(D_2O) = 19\ sec^{-1}$ (22). These calculated values for the intercept were used to secure a better fit of the data since very large geometries (small bucklings) could not be reached with the available D_2O.

The major error associated with an experiment of this type is the

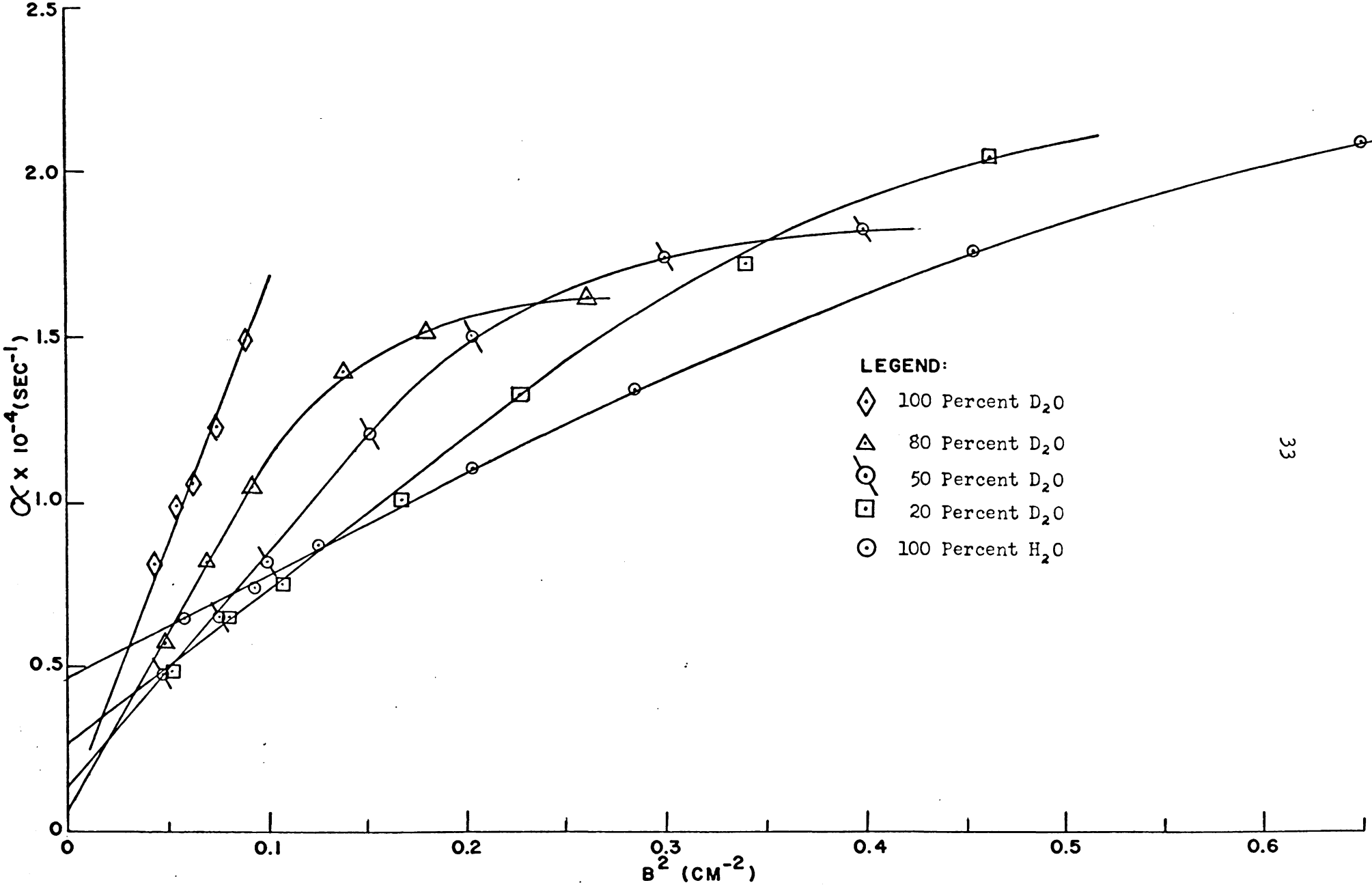

Figure 4. α versus B^2 for 20, 50, and 80 Percent D_2O, including pure H_2O and D_2O.

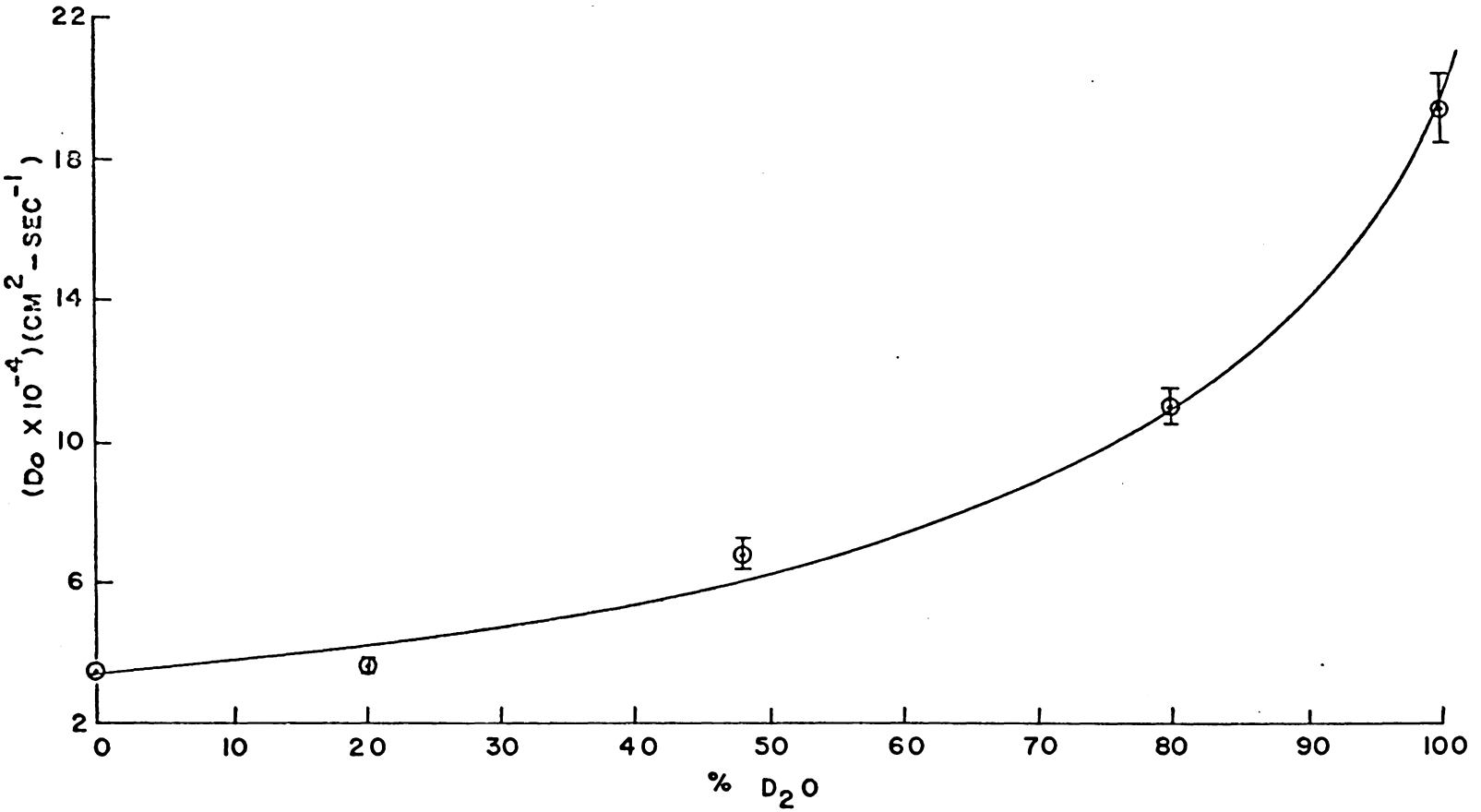

Figure 5. D_o versus D_2O concentrations.

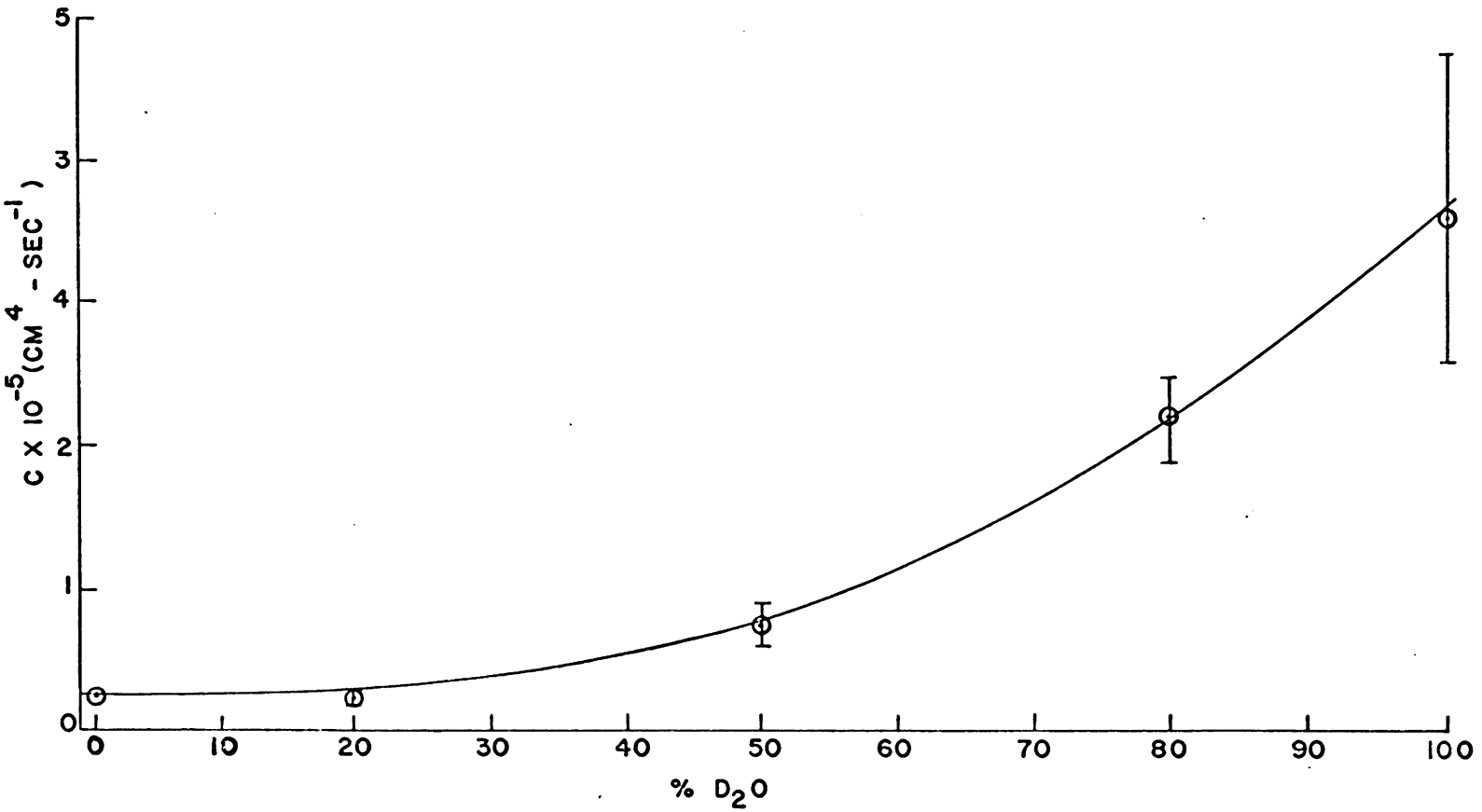

Figure 6. C versus D_2O concentrations.

TABLE V

Thermal Neutron Diffusion Parameters

Percent D_2O	$D_o \times 10^{-4}(cm^2 sec^{-1})$	$C \times 10^{-5}(cm^4 sec^{-1})$	$\lambda_{tr}(cm)$
0	3.634 ± 0.048	0.161 ± 0.008	0.443
20	3.779 ± 0.017	0.170 ± 0.032	0.515
50	6.964 ± 0.044	0.74 ± 0.12	0.915
80	11.75 ± 0.68	2.19 ± 0.42	1.602
100	19.66 ± 1.37	3.56 ± 1.08	2.670

TABLE VI

Measured Decay Constants for H_2O

r(cm)	H(cm)	$B^2(cm^{-2})$	$\alpha(sec^{-1})$
11.69	24.57	0.0586	6358 ± 48
9.41	19.22	0.0920	7212 ± 48
8.07	16.07	0.1269	8515 ± 27
6.37	12.70	0.2035	10890 ± 72
5.36	10.75	0.2865	13313 ± 44
4.23	8.61	0.4550	17576 ± 114
3.55	7.14	0.6508	20987 ± 187

TABLE VII

Measured Decay Constants for 20 Percent D_2O

r(cm)	H(cm)	$B^2(cm^{-2})$	$\alpha(sec^{-1})$
11.69	24.57	0.0586	4840 ± 23
9.41	19.22	0.0920	6478 ± 34
8.07	16.07	0.1269	7477 ± 47
6.37	12.70	0.2035	10181 ± 62
5.36	10.75	0.2865	13407 ± 84
4.23	8.61	0.4550	17233 ± 89
3.55	7.14	0.6508	20590 ± 102

TABLE VIII

Measured Decay Constants for 50 Percent D_2O

r(cm)	H(cm)	$B^2(cm^{-2})$	$\alpha(sec^{-1})$
11.69	24.57	0.0525	4419 ± 32
9.41	19.22	0.0801	6491 ± 25
8.07	16.07	0.1080	7916 ± 42
6.37	12.70	0.1663	12131 ± 58
5.36	10.75	0.2260	15015 ± 110
4.23	8.61	0.3389	17492 ± 144
3.55	7.14	0.4598	18270 ± 303

TABLE IX

Measured Decay Constants for 80 Percent D_2O

r(cm)	H(cm)	$B^2(cm^{-2})$	$\alpha(sec^{-1})$
11.69	22.57	0.0523	5705 ± 37
9.41	19.22	0.0750	8109 ± 48
8.07	16.07	0.1000	10435 ± 52
6.37	12.70	0.1512	13930 ± 98
5.36	10.75	0.1900	15148 ± 210

TABLE X

Measured Decay Constants for 100 Percent D_2O

r(cm)	H(cm)	$B^2(cm^{-2})$	$\alpha(sec^{-1})$
11.69	24.57	0.0436	8161.3 ± 100
11.69	16.83	0.0545	9916.6 ± 105
9.41	19.21	0.0638	10639 ± 111
8.82	16.39	0.0745	12394 ± 141
8.82	12.29	0.0884	14870 ± 151

selection of the proper extrapolation distance for the moderator system. The problem was discussed earlier (Section III). Analyzing the data for the decay constant by Peierls' Method (55) gave the values of α to less than one percent statistical accuracy. The estimated accuracy for the diffusion coefficient and diffusion cooling coefficient was given by the standard error or standard deviation of the data for an α versus B^2 curve.

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APPENDICES

APPENDIX I

Accelerator Modifications

The accelerator used is the Virginia Polytechnic Institute Cockcroft-Walton accelerator as described by McClure (16). This accelerator was modified for the present investigation so that a pre-acceleration pulsing system is used. A three centimeter wavelength microwave transmitter (Figure 7) transmits the pulsing signal to the terminal. In the terminal, the pulsing signal is received by microwave receiver (Figure 8), amplified to 650 volts; and subsequently, applied to a set of electrostatic deflection plates. The pulsing plates are located just beyond a single stage einzel lens through which the ion beam from a radio frequency ion source passes before entering the accelerator column.

The einzel lens is an "electrostatic" lens whose operation depends on the electric field between two adjacent electrodes. A diagram is given in Figure 9. Its operation may be understood by examining what happens to a parallel beam that passes from left to right. When the protons arrive in region 1, they encounter a force which has a sidewise component and the impulse bends them toward the central axis. It appears that the charged particles would receive an equal and opposite impulse at region 2, but this is not true. When the protons reach region 2, they have gained energy and spend less time in this region. The forces are the same, but the time is shorter so the impulse is less. In going through regions 1 and 2, there is a net axial impulse and the protons are bent toward a

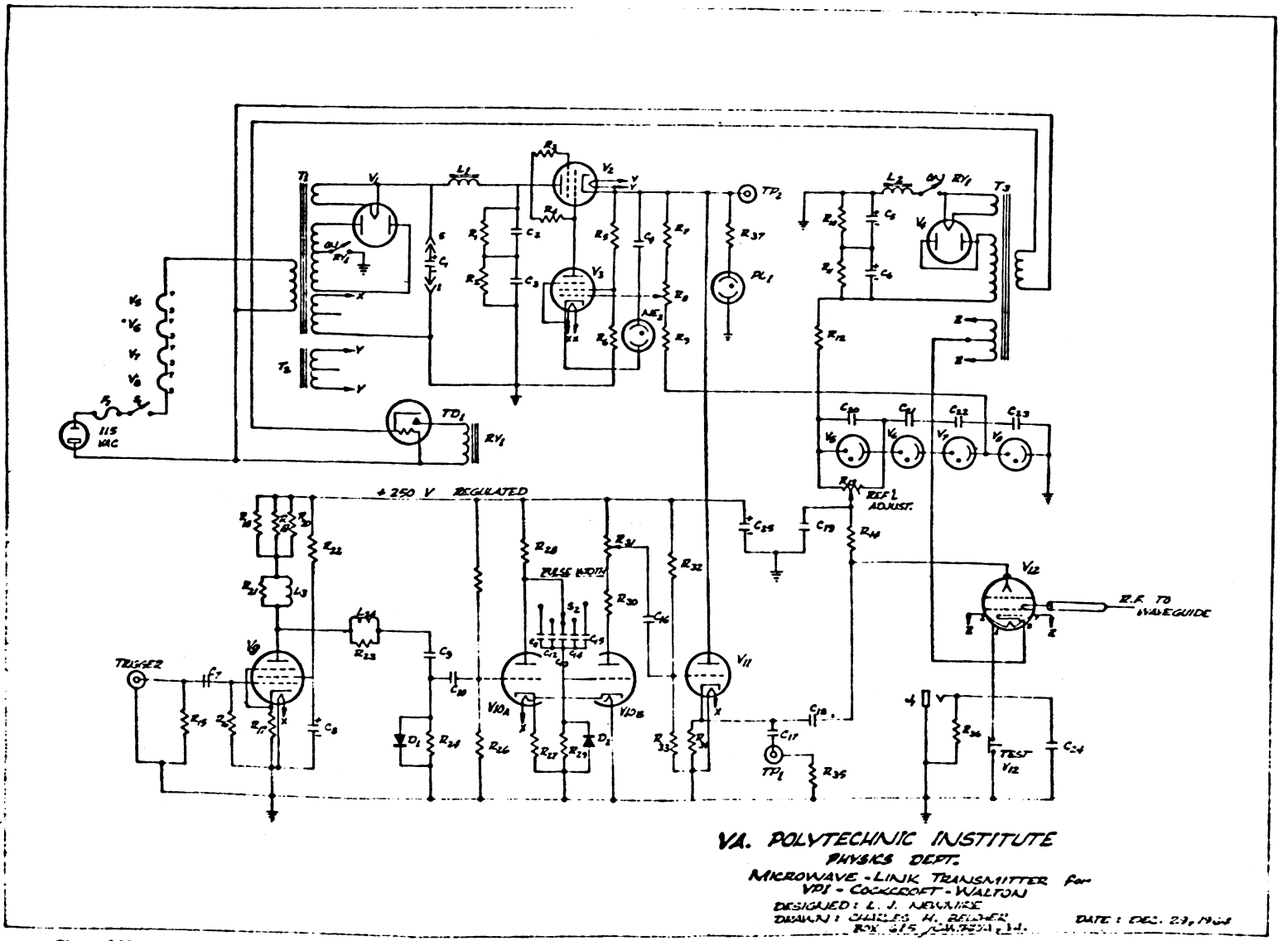

Figure 7. Microwave transmitter.

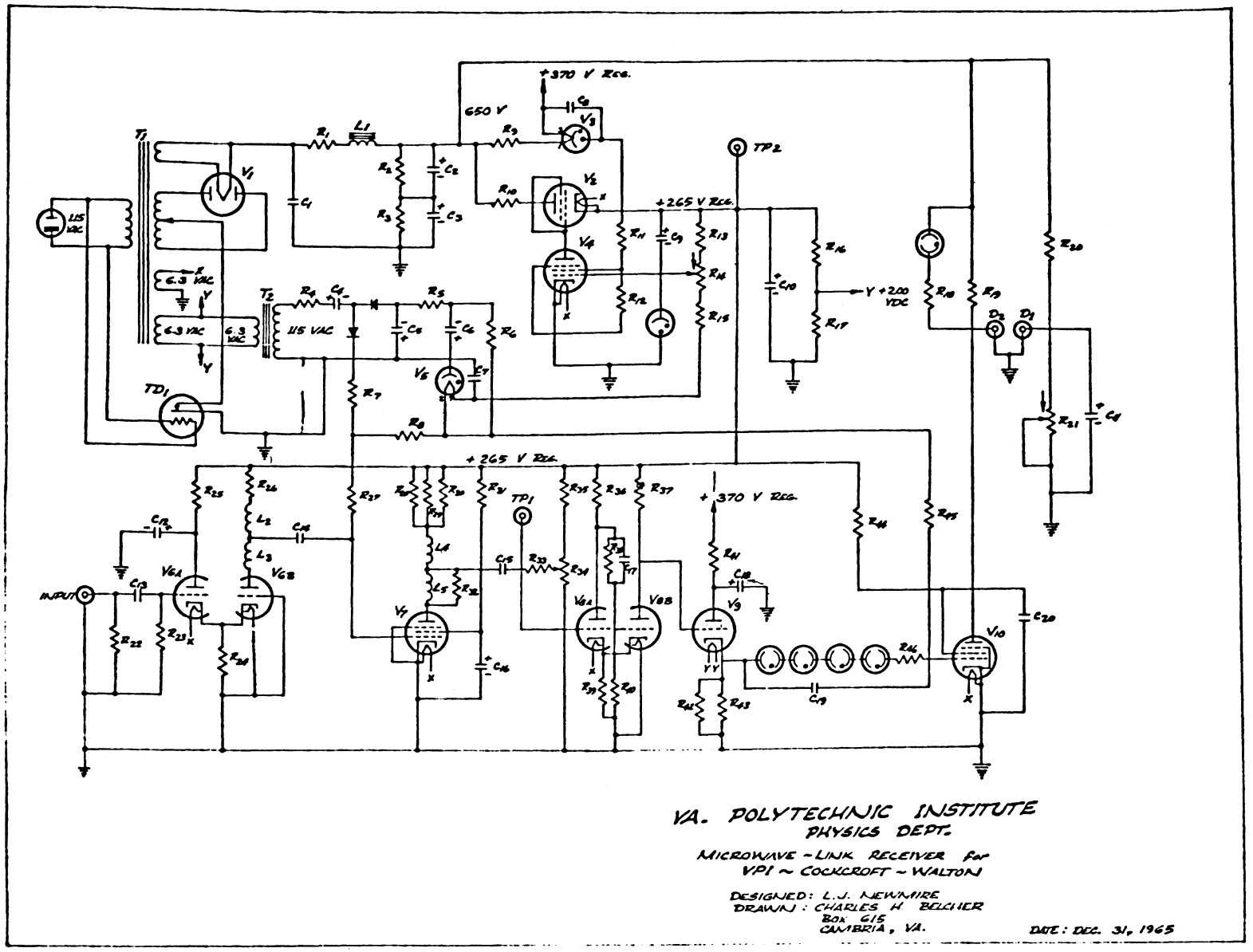

Figure 8. Microwave receiver.

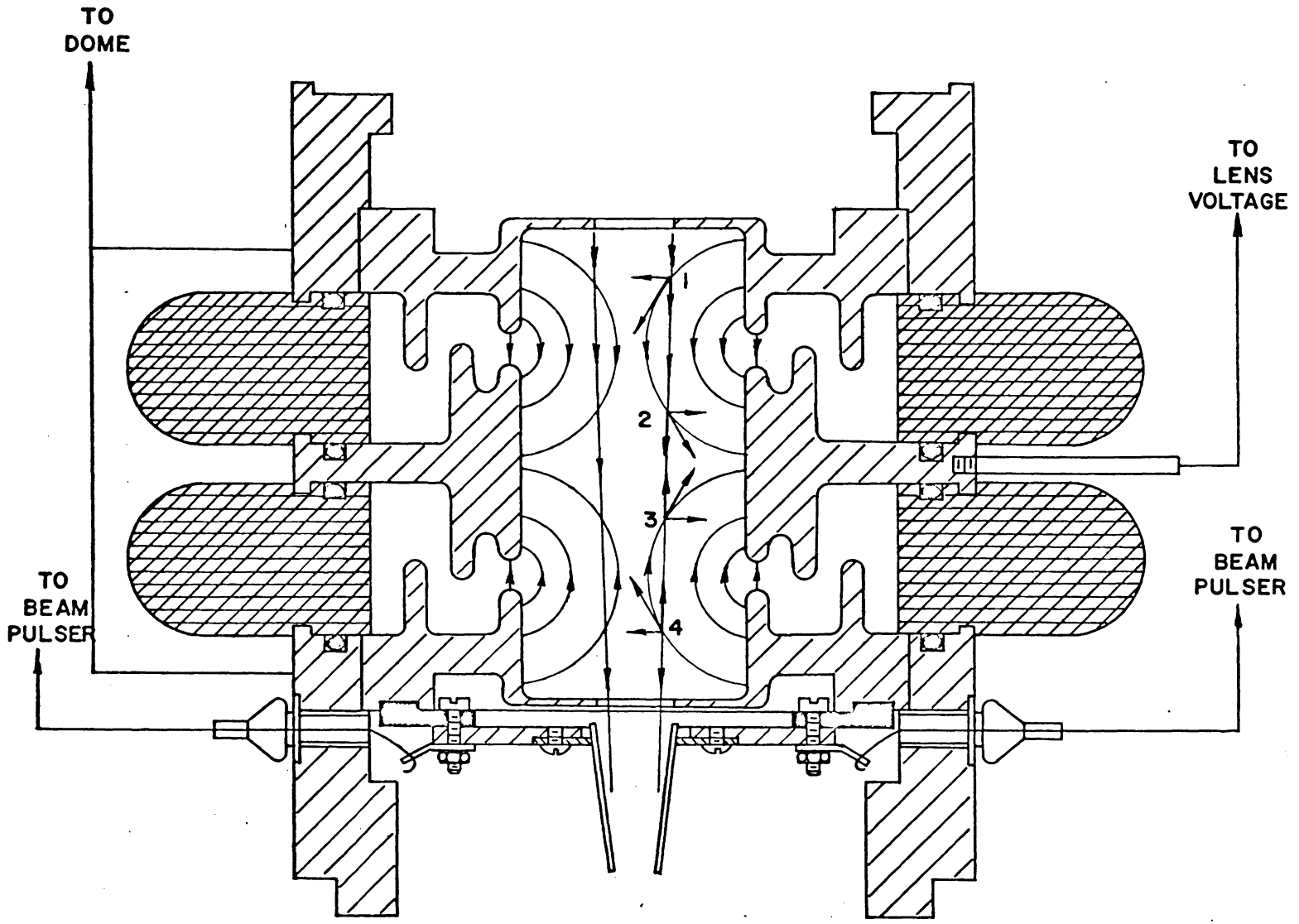

Figure 9. Electrostatic focusing lens.

common point. In passing through regions 3 and 4, the particles receive another kick toward the axis. The force is outward at region 3 and inward at region 4, but the protons stay longer in the latter region. Thus, there is a net impulse toward the axis. Electrostatic lens of this type have been used to focus both positive and negative particles.

When using a terminal pulsing system, a large number of ions are "dumped" into the terminal end of the accelerating tube. Therefore, relatively fast pumping speeds are necessary for the accelerator system to remain in equilibrium with the ion source. To increase the pumping speed of the system, the three and one-fourth inches diameter tube was shortened from 33 accelerator electrodes spaced one inch apart to 17 electrodes of the same spacing. The first two electrodes are used for focusing. The tube opens into a manifold nine inches in diameter and 18 inches in length, to which an NRC six-inch diffusion pump with multi-coolant baffle and drift tube are attached. The forepump is a Model 1397 Duo-Seal manufactured by the Welch Scientific Company. This pumping system is used to maintain a system pressure of 10^{-6} torr with a beam current of 800 microamperes.

The heavy ice target used on the previous accelerator assembly was replaced with a zirconium "drive-in" target. This target is water-cooled when operating with a constant beam current greater than 100 microamperes and air-cooled for a pulsed beam.

The high voltage power supply is a Model 4250-2, 0-250,000 volts DC, reversible polarity, supply manufactured by the Beta Electric Corporation using the voltage doubler principle.

APPENDIX II

Computer Programs

```
$JOB            517150001LAMBDA VS. B SQUARED
$IBJOB           DECK,GO
$IBFTC HGJ1
      DIMENSION R(50), H(50), Y(50), X(50)
      V=220000.
      READ(5,101)NN
      NNN=1
    1 J=0
      READ(5,102)K,N,ALAN
      DO2 I=1,N,1
    2 READ(5,103)R(I),H(I),Y(I)
    3 EPI=0.71*ALAN
      DO 4 I=1,N,1
      B= 2.405/(R(I)+EPI)
      A=3.1415/(H(I)+2.0*EPI)
    4 X(I)=B*B+A*A
      A=0.0
      B=0.0
      C=0.0
      D = 0.0
      E=0.0
      F=0.0
      G=0.0
      DO 5 I=1,N,1
      A=A+X(I)
      B=B+X(I)*X(I)
      C=C+X(I)*X(I)*X(I)
      D=D+X(I)*X(I)*X(I)*X(I)
      E=E+Y(I)
      F=F+Y(I)*X(I)
    5 G=G+Y(I)*X(I)*X(I)
      Z=N
      DET=Z*(B*D-C*C)-A*(A*D-C*B)+B*(A*C-B*B)
      DET1=E*(B*D-C*C)-F*(A*D-C*B)+G*(A*C-B*B)
      DET2=Z*(F*D-G*C)-A*(E*D-G*B)+B*(E*C-F*B)
      DET3=Z*(B*G-C*F)-A*(A*G-C*E)+B*(A*F-B*E)
      A1=DET1/DET
      A2=DET2/DET
      A3=DET3/DET
      J=J+1
      ALAO=ALAN
      ALAN=3.0*A2/V
      IF(ABS((ALAO-ALAN)/ALAO)-.001)6,6,3
    6 D1=0.0
      DO 7 I=1,N,1
      ZZ=Y(I)-A1-A2*X(I)-A3*X(I)*X(I)
```

```
      7 D1=D1+ZZ*ZZ
        ALPH0=D1/((Z-3.)*DET)
        ALPH1=SQRT(ALPH0*(B*D-C*C))
        ALPH2=SQRT(ALPH0*(Z*D-B*B))
        ALPH3=SQRT(ALPH0*(Z*B-A*A))
        WRITE (7,104)K,N
        WRITE (7,105)J
        WRITE (7,106)ALAN
        WRITE (7,107)
        DO 8 I=1,N,1
      8 WRITE (7,108)R(I),H(I),X(I)
        WRITE (7,109)A1,ALPH1
        WRITE (7,110)A2,ALPH2
        WRITE (7,111)A3,ALPH3
        IF(NN-NNN)10,10,9
      9 NNN=NNN+1
        GO TO 1
     10 STOP
    101 FORMAT(I3)
    102 FORMAT(I3,I3,E14.7)
    103 FORMAT(E10.3,E11.4,E12.5)
    104 FORMAT(24HRESULTS FOR DATA SET NO.,I3,10HCONTAINING,I3,6HPOINTS)
    105 FORMAT(I5,32HITERATIONS HAVING BEEN PERFORMED)
    106 FORMAT(19HLAMBDA TRANSPORT IS,E14.7)
    107 FORMAT(4X,6HRADIUS,7X,6HHEIGHT,6X,10H B SQUARED)
    108 FORMAT(E14.7,3X,E14.7,3X,E14.7)
    109 FORMAT(3HA1=,E14.7,9H ERROR IS, E14.7)
    110 FORMAT(3HA2=,E14.7,9H ERROR IS ,E14.7)
    111 FORMAT(3HA3=,E14.7,9H ERROR IS,E14.7)
        END
$ENTRY          HGJ1
$IBSYS
```

```
$JOB            517150001LMDA VS. BSQ FIX A1
$IBJOB           DECK,GO
$IBFTC HGJ2
      DIMENSION R(50), H(50), Y(50), X(50)
      V=220000.
      READ(5,101)NN
      NNN=1
    1 J=0
      READ(5,102)K,N,ALAN
      READ(5,112)A1
      DO2 I=1,N,1
    2 READ(5,103)R(I),H(I),Y(I)
    3 EPI=0.71*ALAN
      DO 4 I=1,N,1
      B= 2.405/(R(I)+EPI)
      A=3.1415/(H(I)+2.0*EPI)
    4 X(I)=B*B+A*A
      A=0.0
      B=0.0
      C=0.0
      D = 0.0
      F=0.0
      G=0.0
      DO 5 I=1,N,1
      A=A+X(I)
      B=B+X(I)*X(I)
      C=C+X(I)*X(I)*X(I)
      D=D+X(I)*X(I)*X(I)*X(I)
      F=F+Y(I)*X(I)
    5 G=G+Y(I)*X(I)*X(I)
      Z=N
      DET=B*D-C*C
      Q=F-A1*A
      T=G-A1*B
      A2=(Q*D-T*C)/DET
      A3=(T*B-Q*C)/DET
      J=J+1
      ALAO=ALAN
      ALAN=3.0*A2/V
      IF(ABS((ALAO-ALAN)/ALAO)-.001)6,6,3
    6 D1=0.0
      DO 7 I=1,N,1
      ZZ=Y(I)-A1-A2*X(I)-A3*X(I)*X(I)
    7 D1=D1+ZZ*ZZ
      ALPH0=D1/((Z-2.)*DET)
      ALPH1=0.0
      ALPH2=SQRT(ALPH0*D)
      ALPH3=SQRT(ALPH0*B)
      WRITE (7,104)K,N
      WRITE (7,105)J
```

```
      WRITE (7,106)ALAN
      WRITE (7,107)
      DO 8 I=1,N,1
      S=Y(I)-A1-A2*X(I)-A3*X(I)*X(I)
    8 WRITE (7,108)R(I),H(I),X(I),S
      WRITE (7,109)A1,ALPH1
      WRITE (7,110)A2,ALPH2
      WRITE (7,111)A3,ALPH3
      IF(NN-NNN)10,10,9
    9 NNN=NNN+1
      GO TO 1
   10 STOP
  101 FORMAT(I3)
  102 FORMAT(I3,I3,E14.7)
  103 FORMAT(E10.3,E11.4,E12.5)
  104 FORMAT(24HRESULTS FOR DATA SET NO.,I3,10HCONTAINING,I3,6HPOINTS)
  105 FORMAT(I5,32HITERATIONS HAVING BEEN PERFORMED)
  106 FORMAT(19HLAMBDA TRANSPORT IS,E14.7)
  107 FORMAT(4X,6HRADIUS,11X,6HHEIGHT,9X,10H B SQUARED,7X,6H YM-YC)
  108 FORMAT(E14.7,3X,E14.7,3X,E14.7,3X,E14.7)
  109 FORMAT(3HA1=,E14.7,9H ERROR IS, E14.7)
  110 FORMAT(3HA2=,E14.7,9H ERROR IS ,E14.7)
  111 FORMAT(3HA3=,E14.7,9H ERROR IS,E14.7//)
  112 FORMAT(E11.4)
      END
$ENTRY          HGJ2
$IBSYS
```

ABSTRACT

Neutron diffusion parameter measurements have been made at 21°C in 20, 50, and 80 percent concentrations of heavy and light water, including pure H_2O and D_2O as moderators. Pulsed neutron techniques were used with a 250-kv Cockcroft-Walton accelerator as the pulsed source. The range of bucklings used was from 0.05 cm^{-2} to 0.70 cm^{-2}.

The values for the diffusion coefficient, D_o; the diffusion cooling coefficient, C; and the transport mean free path, λ_{tr}, were determined from the expansion of the decay constant to second order using an iteration procedure. The values of D_o, C, and λ_{tr} are tabulated below for the various concentrations, including pure H_2O and D_2O.

Percent D_2O	$D_o \times 10^{-4}(cm^2 sec^{-1})$	$C \times 10^{-5}(cm^4 sec^{-1})$	$\lambda_{tr}(cm)$
0	3.634 ± 0.048	0.161 ± 0.008	0.443
20	3.779 ± 0.017	0.170 ± 0.032	0.515
50	6.964 ± 0.044	0.74 ± 0.12	0.915
80	11.75 ± 0.68	2.19 ± 0.42	1.602
100	19.66 ± 1.37	3.56 ± 1.08	2.670