

THE IRRADIATION EFFECTS ON THE SOLVENT EXTRACTION SYSTEM OF
TRIBUTYL PHOSPHATE-DODECANE

by

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NOMENCLATURE

Mwd	mega-watt days
tonne	metric ton, 1000 kilograms
MeV	10^6 electron volts
keV	10^3 electron volts
eV	electron volt
G(X)	the yield of "X" component in molecules per 100 eV
ϵ	electron fraction
mm	millimeter
torr	millimeters of mercury (pressure)
$^{\circ}\text{K}$	degrees Kelvin
N	gram equivalent weight of solute per liter of solution
M	molecular formula weight per liter of solution
rad	100 ergs absorbed energy per gram of material
min	minute
$^{\circ}\text{C}$	degrees Centigrade
ml	milliliter, 10^{-3} liter
g	gram
mp	millipoise, 10^{-3} poise
pK	negative log of the ionization constant
v/o	volume per cent

1.0 INTRODUCTION

Nuclear fuel is reprocessed to recover fissile and fertile species such as uranium, plutonium, and thorium from the accumulated fission products. Solvent extraction, ion exchange, and pyrometallurgical processes are three reprocessing techniques. Of the three, solvent extraction is the best developed and most common.

A solvent extraction is the separation of various chemical species dissolved in a solvent, by the transfer of certain chemical species to another solvent, immiscible with the first solvent. The transfer takes place due to the greater solubility of the transferred constituents in the second solvent than in the first solvent.

Dilute nitric acid is typically used to dissolve the spent fuel and tributyl phosphate (TBP) is used as the second immiscible solvent in the Purex system [5]. TBP is used as the solvent due to its ability to form a complex with uranium, plutonium, and thorium in the forms, $\text{UO}_2(\text{NO}_3)_2 \cdot 2\text{TBP}$, $\text{Pu}(\text{NO}_3)_4 \cdot 2\text{TBP}$, and $\text{Th}(\text{NO}_3)_4 \cdot 2\text{TBP}$. These complexes are more soluble in the organic phase, TBP, than in the inorganic phase.

TBP is an organic ester of phosphoric acid and the structure is shown in Fig. 1. TBP has tetrahedral symmetry with one double oxygen-phosphorous bond and three single butoxy-phosphorous bonds. Physical properties of TBP are listed in Appendix A. TBP is stable toward oxidation and nitric acid, the usual aqueous phase, and undergoes alkaline hydrolysis [1,2]. The hydrolysis produces dibutyl phosphate (DBP), and monobutyl phosphate (MBP), and phosphoric acid. Since this hydrolysis reaction is slow, it is a minor problem. Pure TBP is viscous and the density is within 3% of water, therefore it must be diluted for solvent extraction use.

The most commonly used diluent is dodecane, generally industrial grade of about 90% purity. Dodecane is used because it is economical, has a density of 0.75 g/cm^3 , and its radiolytic degradation products don't significantly complex with materials such as zirconium [3]. The physical properties of dodecane are also listed in Appendix A.

The Purex solvent extraction process, using a mixture of dodecane and TBP, dates back to the early post World War II development work to find a replacement for the Hanford bismuth precipitation process [4,5]. Due to its use in the production of plutonium for the weapons program, a great deal of experience is available. However, this experience is with low burnup fuel, typically less than 1000 Mwd/tonne. Very little experience is available with high burnup fuel, 30,000 Mwd/tonne or greater, that is produced by current generation light water reactors (LWR). Even less experience is available concerning light water thermal breeder reactors or liquid metal fast breeder reactor fuels, where burnups of greater than 100,000 Mwd/tonne are foreseen.

High fuel burnups not only increase the specific activity of the spent fuel, but also cause an increase in the fraction of energy released in the form of alpha particles. Alpha particles have a higher Linear Energy Transfer (L.E.T.) than beta or gamma radiation, where L.E.T. is defined as the energy deposited per unit length of track [6]. Since high L.E.T. irradiation causes the damage to a medium to be more localized and concentrated, there is the possibility for significantly different chemical reactions to dominate, than dominate for low L.E.T. irradiations.

For low L.E.T. experimental results to be meaningful to the reprocessing problems for the near future, the emphasis should be placed on basic understanding of the chemical mechanism and the reaction rates. This kind of

basic knowledge is necessary to allow reasonable extrapolation to the conditions to be encountered, as opposed to the typical reprocessing literature which is a tabulation of radiolysis products with little information about reaction mechanisms.

In this work irradiations of dodecane, TBP, and mixtures of the two were made and analyzed. Gaseous products were measured by a combination of gas chromatography and pressure-volume-temperature analysis. Acid products were determined by potentiometric titration. This work was performed as a first step in the understanding of the physical and chemical reactions involved in the degradation of reprocessing solvents.

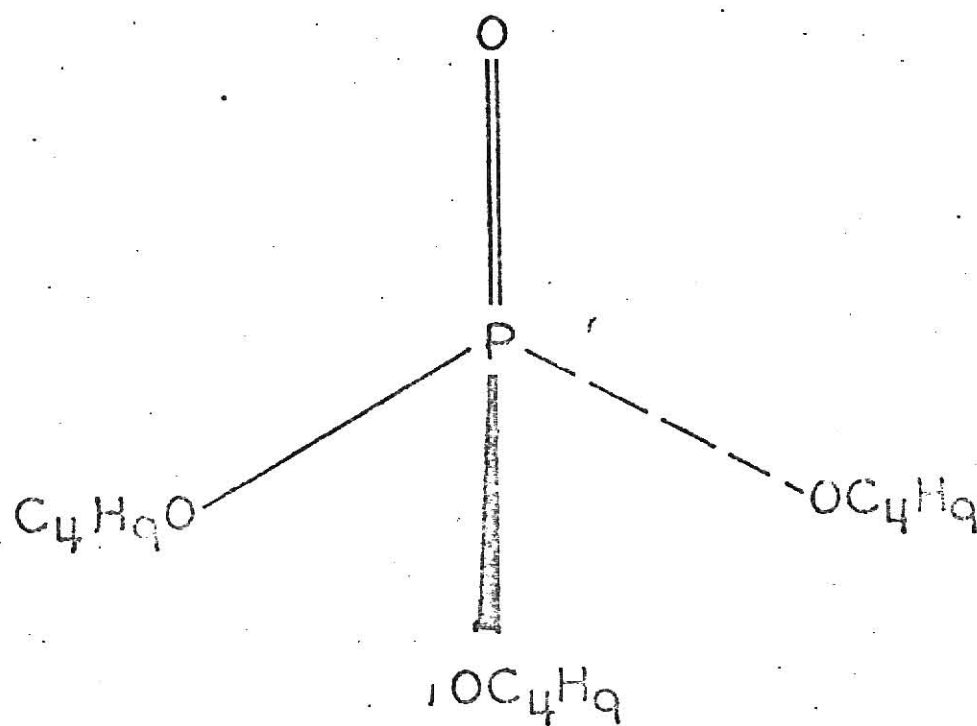


Fig. 1: Molecular Structure of TBP.

2.0 THEORY

2.1 Types of Radiation

Radiation encountered in the reprocessing of spent nuclear fuel will be gamma, beta, and alpha. In the following, the types and significance of primary interactions of these radiations will be discussed.

2.1.1 Beta Particles

Beta particles are energetic electrons produced by the decay of an unstable nucleus. The betas encountered in nuclear fuel reprocessing have an average energy of about 0.5 MeV and are the result of fission product decay. Due to the low penetrating capability of beta particles, essentially all the energy of the beta particles is deposited in the processing stream.

The three major types of interaction of an electron with matter are bremsstrahlung, elastic scattering, and inelastic scattering. The atomic mass of dodecane or tributyl phosphate (TBP) is too low for significant bremsstrahlung and too high for significant elastic scattering. Inelastic scattering will dominate.

Inelastic scattering occurs when an electron undergoes a coulombic interaction with the electrons in the irradiated material, producing ionized and excited molecular states. The electron ejected from the molecules by the beta particle will have sufficient energy to cause further ionization and excitation. Cascades of second, third, and higher order electrons can be produced [7]. The excited states produced by the beta particles and subsequent electrons are similar to those produced by optical transitions, however more triplet states are observed due to a relaxation of the optical selection rules [8].

2.1.2 Gamma Rays

Since the source of gamma radiation in nuclear fuel reprocessing is fission product decay, almost all gamma rays produced will have an energy of less than 10 MeV [9]. Below 10 MeV the two major photon interaction mechanisms are Compton scattering and the photoelectric effect.

In Compton scattering, the incident photon interacts with an electron of the medium, causing the electron to be accelerated and the incident photon to be scattered at a lower energy. The energy of the scattered photon and electron can be calculated from the energy of the incident photon (see almost any introductory physics text such as "Nuclear Physics," by I. Kaplan). For example, a 0.661 MeV incident gamma ray will produce a scattered electron of approximately 0.5 MeV maximum energy. This scattered electron undergoes inelastic scattering with the electrons of the medium.

Photoelectric effect is the process of absorption of an incident photon by an atom which then ejects an inner shell electron with an energy equal to the incident photon energy minus the electron binding energy. This leaves the atom ionized and in an excited state. However this mechanism is of secondary importance to Compton scattering except for photon energies of less than 100 keV [7,10].

The result of photon absorption is the same for both Compton scattering and photoelectric effect, energetic electrons, ionization, and excited states. Beta particles produce these same species, therefore beta radiation can be simulated by gamma radiation. However, gamma and beta radiation can not be treated identically due to the much greater penetrating power of gamma radiation, which allows significant energy to escape the reprocessing stream.

2.1.3 Alpha Particles

Alpha particles are heavy, energetic, charged particles (He^{2+}), produced from the decay of an unstable nucleus. They interact with matter much the same way as beta particles, bremsstrahlung, elastic scattering, and inelastic scattering. However, since the alpha particle is more massive than a beta particle, the alpha particles move more slowly and inelastic scattering is the only significant interaction.

The alpha particle has a much higher Linear Energy Transfer (L.E.T.) resulting in a shorter interaction track and a higher density of interactions than a beta or gamma. These interactions have the same intermediate products as beta or gamma particles, but the much higher density can lead to a different distribution of final reaction products.

2.1.4 Relative Importance

Traditionally, the only significant radiation encountered in nuclear fuel reprocessing was beta and gamma rays. High burnup fuel however, has significant transuranic isotopes present that are very active alpha emitters, so the effect of high L.E.T. radiation should be considered.

2.2 Radiolysis Products

Alkane radiolysis results in hydrogen production, olefin generation, dimerization, and generation of fragments C_1 to C_n which can then combine in pairs. The major species evolved are hydrogen, dimer, and olefin.

Hexane, C_6H_{14} , has been studied in considerable detail and the following, Table 1, is a tabulation of radiolysis products [11]. The G value is defined as the number of that species produced per 100 eV of absorbed energy. These types of results are typical for alkane radiolysis. However, alkane structure

Table 1. Radiolysis Products from Liquid Hexane (C_6H_{14})

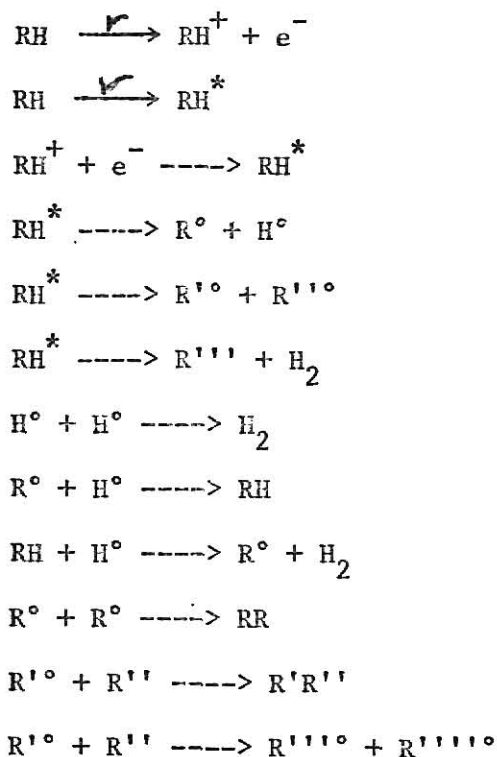
Product	G Value
H_2	5.0
CH_4	0.12
C_2H_2	0.3
C_2H_4	0.3
C_3H_6	0.13
C_3H_8	0.42
C_4H_8	0.03
C_4H_{10}	0.50
i- C_4H_{10}	0
C_5H_{12}	0.30
trans- C_6H_{12}	0.2
i- C_6H_{14}	0
C_7	0.15
C_8	0.53
C_9	0.45
C_{10}	0.43
C_{11}	0.02
C_{12}	2.0

does effect the final product yield. Branched alkanes with more tertiary carbon hydrogen bonds generally have greater methane yields and long chain alkanes have a definite tendency to cleave carbon-carbon bonds toward the middle of the chain. This leads to a final radiolysis product yield shifted toward species with a greater number of carbon atoms than the parent.

Numerous mechanisms have been proposed to account for these observed results, the one currently favored is a sequence of radical reactions. Excited states, produced directly by ionizing radiation or as a result of geminate ion recombination, are the source of radical species. Ion

mechanisms in a nonpolar medium, such as an alkane, are thought to be quite limited due to the rapid geminate recombination rate. Conductivity measurements on aliphatic hydrocarbons indicate that less than 3% of the electrons produced as a result of ionization, diffuse into the bulk medium before recombination with the parent ion [12].

The radical reaction mechanism can be represented by the following reaction sequence:



Where R, R', R'', R''', and R'''' are alkyl groups of differing length.

Product yields (G_m) from the irradiation of a homogeneous mixture of two materials would be expected to be equal to the yield from a single component (G_a) times its electron fraction (E_a) plus the yield from the second component (G_b) times its electron fraction (E_b), this is called the mixture law.* However, significant deviation from this situation has been observed in binary solutions such as cyclohexane/benzene.

*Mathematically, $G_m = G_A E_A + G_B E_B$.

The cyclohexane/benzene system is one of the best studied mixture law deviations and there is still considerable disagreement concerning the mechanism. The deviation mechanism has been suggested to be excitation transfer, chemical scavenging, or charge transfer.

The presence of benzene suppresses the yield of hydrogen, cyclohexyl radical, and the dicyclohexyl radical [13]. This result can be interpreted as excitation transfer from excited cyclohexane to benzene, which would undergo a radiative or a nonradiative transition to the ground state. This excitation transfer would have prevented the formation of radical species from the excited state of cyclohexane [13,14,15]. Since benzene can undergo electronic transition without breaking chemical bonds, the total number of radical species has decreased. Another interpretation is that of Dyne [13], who suggested that the benzene quenched $C_6H_{12}^+$ and e^- prior to geminate recombination of the ions which would lead to excited state cyclohexane. A case can be made for chemical quenching by radical addition to benzene, since phenylcyclohexane and dicyclohexadienyl have been observed [13] in cyclohexane/benzene mixtures and not in irradiations of the pure components.

The mixture of TBP and dodecane has considerable potential for deviation from mixture law. The combination of nonpolar dodecane and polar TBP opens the possibility of significant ion reactions since the increased polarity of the solute could reduce the geminate recombination rate of the parent alkane ions.

The literature concerning TBP/dodecane mixture law deviations is limited and conflicting. Barelko [16] and Williams [17] observed mixture law deviations. However Burger [18] did not observe deviation from the mixture law with a mixture of TBP and iso-octane. Differences in experimental techniques of dosimetry, sample purification, and duration and rate of radiation exposure makes evaluation of the literature difficult.

Another difficulty in looking at mixture law deviations of the TBP/dodecane system is that the literature of the single components is poor. For example, the reported radiolysis yields of gaseous products from TBP range from a G value (number of molecules/100 eV) of less than 0.24 [19] to 3.1 [20]. The dodecane literature is almost nonexistent; Barelko [16] reports a gas yield, volatile at an unknown temperature, of $G = 3.8$ and Dewhurst [21] reports a $G(\text{H}_2) = 4.9$ and $G(\text{CH}_4) = 0.05$. TBP radiolysis yields and products are tabulated in Table 2.

Table 2. Radiolysis Yields of Pure TEP: Low L.E.T. Irradiation

$G(H_2)$	$G(DBP)$	$G(MBP)$	$G(Acid)$	$G(CH_4)$	$G(BuOH)$	$G(Total\ Hydrolysis)$	Reference
2.02			3.71	0.032			[this work]
1.72	2.44	0.14	2.58	0.072			[20]
2.5	1.9	0.3	2.2		0.65		[27]
1.11	1.52	0.12	1.64	0.05	0.0		[17]
1.5	1.86					0.24	[19]
(total gas)					0.78		[16]
1.59	2.25	0.39	2.64	0.07	0.13		[28]

3.0 EXPERIMENTAL PROCEDURE

The preparation and irradiation of samples and the determination of product yields are discussed in this section.

3.1 Sample Preparation

The samples consisted of dodecane, tributyl phosphate (TBP), and mixtures of these two components. Preparation consisted of sample purification and degassing. These steps are outlined below.

3.1.1 Sample Purification

The tributyl phosphate (TBP) (Matheson, Coleman & Bell Inc., white label grade) was found, from potentiometric titrations, to have less than 0.1% acid impurities and was used as received. The dodecane (Matheson, Coleman and Bell, Inc., practical grade) was treated with fuming sulfuric acid (30% SO_3) to remove unsaturated impurities and then the remaining unreacted acid and impurities were removed. Residual acid was neutralized by the addition of a 2% sodium carbonate solution. Next the dodecane was passed through approximately 24 inches of 28-200 mesh silica gel at about one drop per second. This step was performed twice. The resulting dodecane was analyzed on a Cary 14 UV spectrophotometer and found to be near manufacturers specifications for spectral grade dodecane. UV spectrometry results are shown in Appendix B.

3.1.2 Degassing

The samples were degassed in two steps. The first was to pass dry clean helium through the sample for a period of four to eight hours. This reduced the concentration of atmospheric gases, which shortened the time and improved the results of the cell degassing.

The irradiation cell (Fig. 2) was attached to the high vacuum manifold by means of a "butt" joint using 10 mm, thick wall glass tubing. The cell was evacuated and held at 10^{-6} torr for at least 24 hours to outgas the cell walls. The cell was then repressurized to slightly greater than one atmosphere with clean dry helium. The side arm of the cell was then flame cracked open, allowed to cool, and a five ml sample injected using a syringe with a four inch needle. Care was exercised to minimize the amount of sample material deposited on wall of side arm to reduce the chance of pyrolysis. Carbon monoxide and other unidentified substances found in some of the early irradiations are attributed to pyrolysis. The sample was frozen to 195 °K with a dry ice/ethanol bath, side arm resealed, and the cell evacuated. The dry ice/ethanol bath was used instead of liquid nitrogen (77 °K) to remove possible gaseous pyrolysis products. Once the cell was evacuated to under 10^{-5} torr, the cell was isolated from the vacuum system and thawed to room temperature. The freeze-pump-thaw cycle was repeated until the system pressure increased to less than 10^{-5} torr upon opening the cell to the vacuum system.

At this time the 10 mm thick wall glass tubing connecting the cell to the vacuum system manifold was gently flamed to drive off residual sample material. The tube wall was then heated to softening temperature of glass, allowing the tube walls to collapse inward, making a vacuum seal and was then removed.

3.2 Irradiation and Dosimetry

The irradiations were performed in a Gammacell (AECL 220) with repeatable positioning provided by an aluminum sample holder shown in Fig. 3. Dosimetry was done with the Fricke Dosimeter (Fe^{+2} in 0.8N H_2SO_4) [22]. Due to strong axial and radial variations in dose rates, care was

exercised to have the dosimeter solution and the sample occupy the same position in the sample holder. This was done by using the same diameter glass tubing for the dosimeter cell as was used in sample cell and filling both cells to the same height.

Eighteen irradiations of dosimeters were done in each of the twelve positions of the sample holder, over an eight month period. The dose rates were normalized to the date of the first irradiation and the mean and standard deviation were determined. This dose rate, approximately 1100 rad/m, with a decay correction was used to calculate the dose absorbed by the dosimeter solution. A computer program was written to handle these calculations and a sample output and the program listing are shown in Appendix C. Dose correction factors were applied to correct for the difference in absorbed dose in dosimeter and organic sample solutions. These correction factors were calculated on an electron density basis and are found in Appendix A, Table A-3.

3.3 Gas Analysis

Analysis of the gaseous radiolytic products was done in two steps; determination of the total amount of gas and then the species of gases present. The total number of moles of gas volatile at 161 °K was determined by a pressure-volume-temperature (PVT) analysis on a Saunders-Taylor apparatus [23], shown in Fig. 4. Saunders-Taylor calibration data is given in Appendix D. A sample of the gas extracted was collected in a gas sampler, shown in Fig. 5. This gas sample was analyzed at 40 °C on a Varian 90P3 gas chromatograph, using a six foot, 60-80 mesh, molecular sieve 5A column, and a thermal conductivity detector. The gases were identified by the elution time and the relative amounts were determined by comparison

to calibration data for known amounts of nitrogen, oxygen, hydrogen, methane and carbon monoxide. Varian 90P3 calibration data are found in Appendix E. Detection of less than 0.1 micromoles was achieved.

3.4 Acid Determination: Potentiometric Titration

The yield of dibutyl phosphate (DBP) and monobutyl phosphate (MBP) was determined by titrating the irradiated solution with 0.012 M KOH. The equivalence point was determined with a Fisher model 38 pH meter equipped with a combination electrode. Two or four ml of the irradiated sample was pipetted into 10 ml of distilled water. The sample was mixed until broken down to an emulsion, at which time the water soluble DBP and MBP were in contact with the distilled water and were dissolved. The sample was then titrated with constant mixing. Titrations were made on unirradiated TBP, dodecane, distilled water, DBP, MBP, and a mixture of DBP and MBP. The purified MBP and DBP were produced by the procedure in Reference [30]. Correction factors for the residual acid concentration in TBP and water were applied. The dodecane was found to have insignificant acid impurities. A 90% DBP - 10% MBP mixture was prepared and titrated. The second proton from the diprotic MBP was detected and the MBP fraction was found to agree with the known value. However, the break in the titration curve was such that any lower concentration of MBP would not be resolved. Therefore, a yield of MBP, 10% of the DBP yield, seems to be the limit of resolution with potentiometric analysis. Sample plots of the titration curves are shown in Fig. 6 thru Fig. 11.

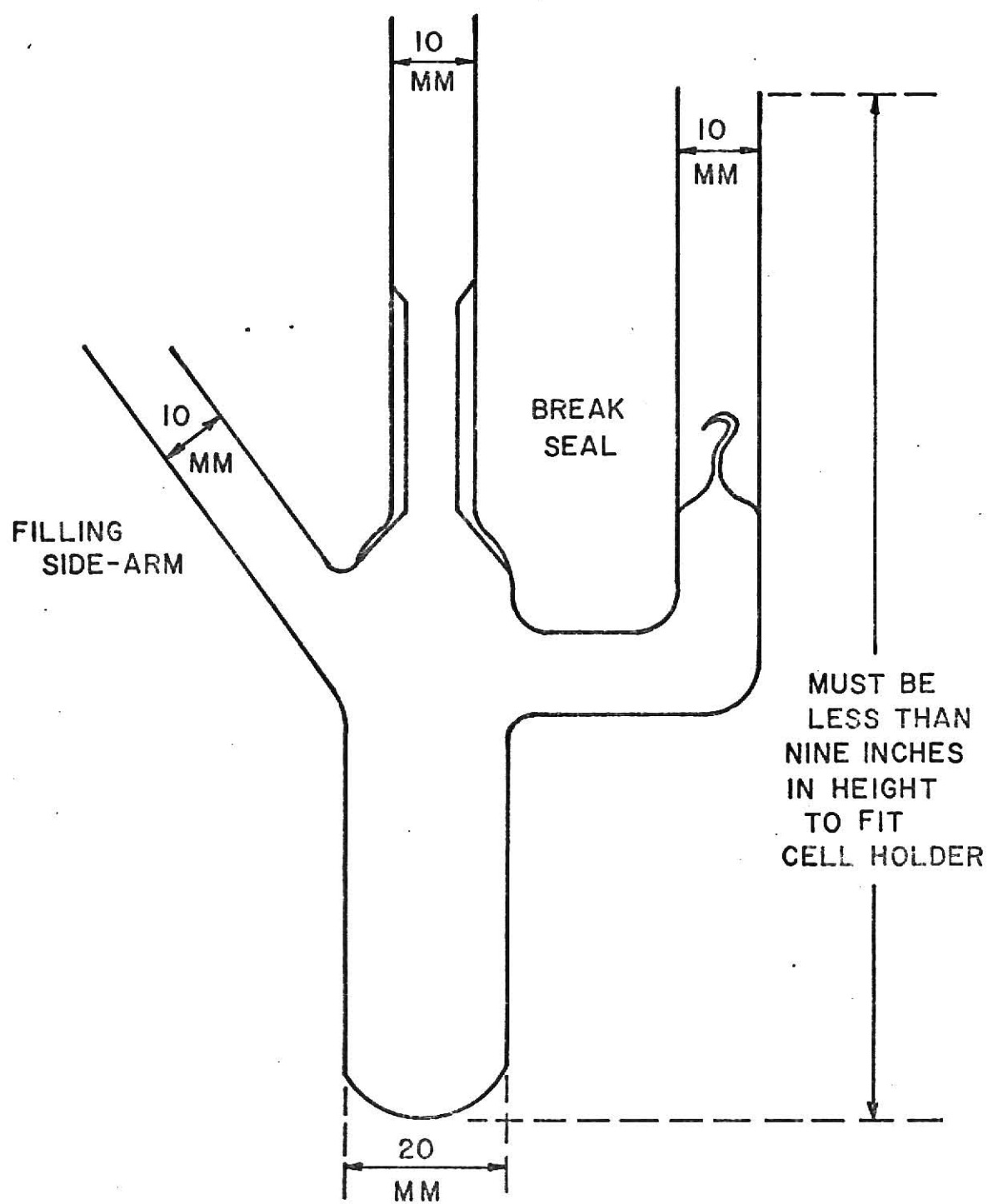


Fig. 2: Sketch of the irradiation cell[29]

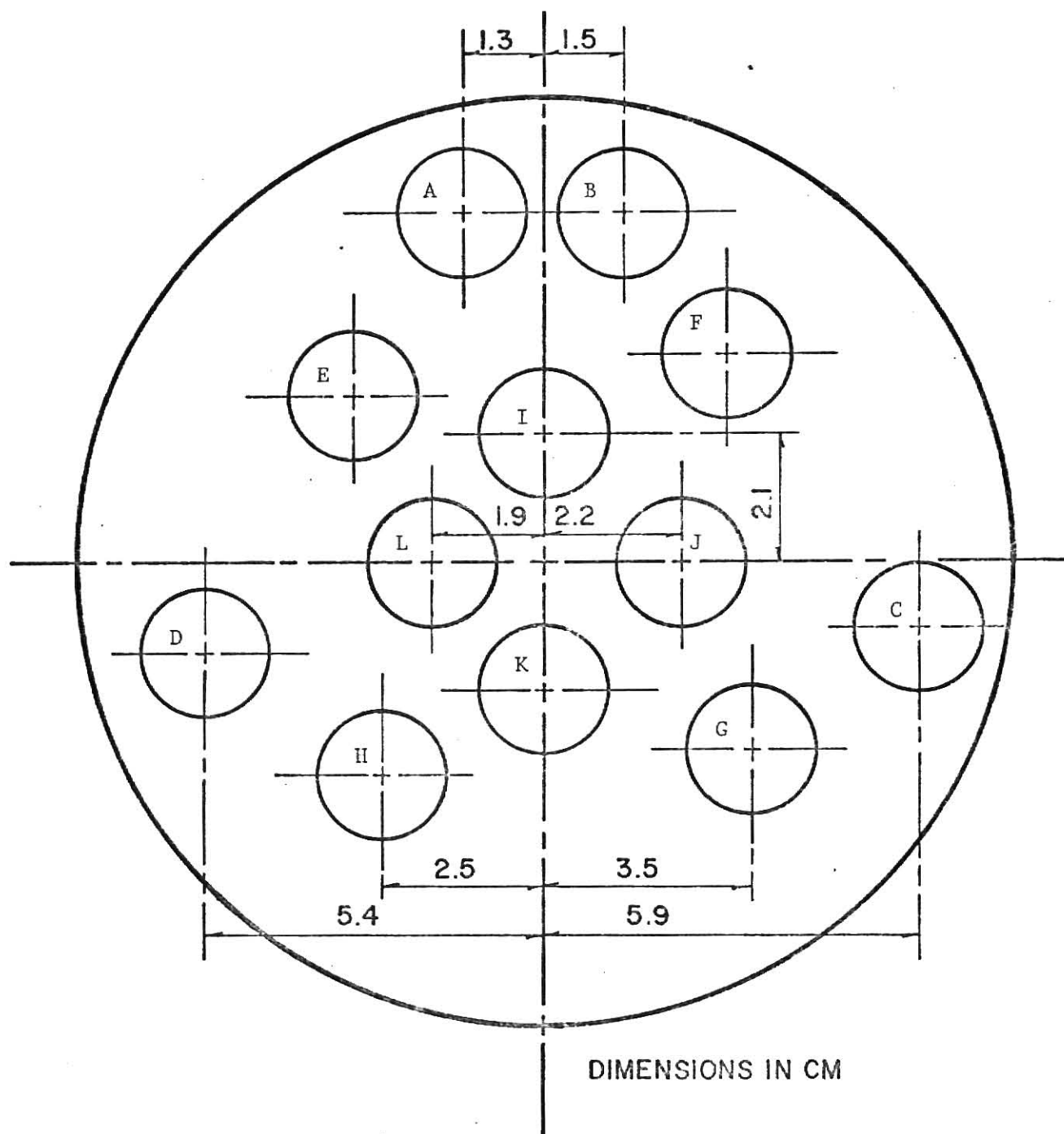


Fig. 3: Schematic of sample cell holder.

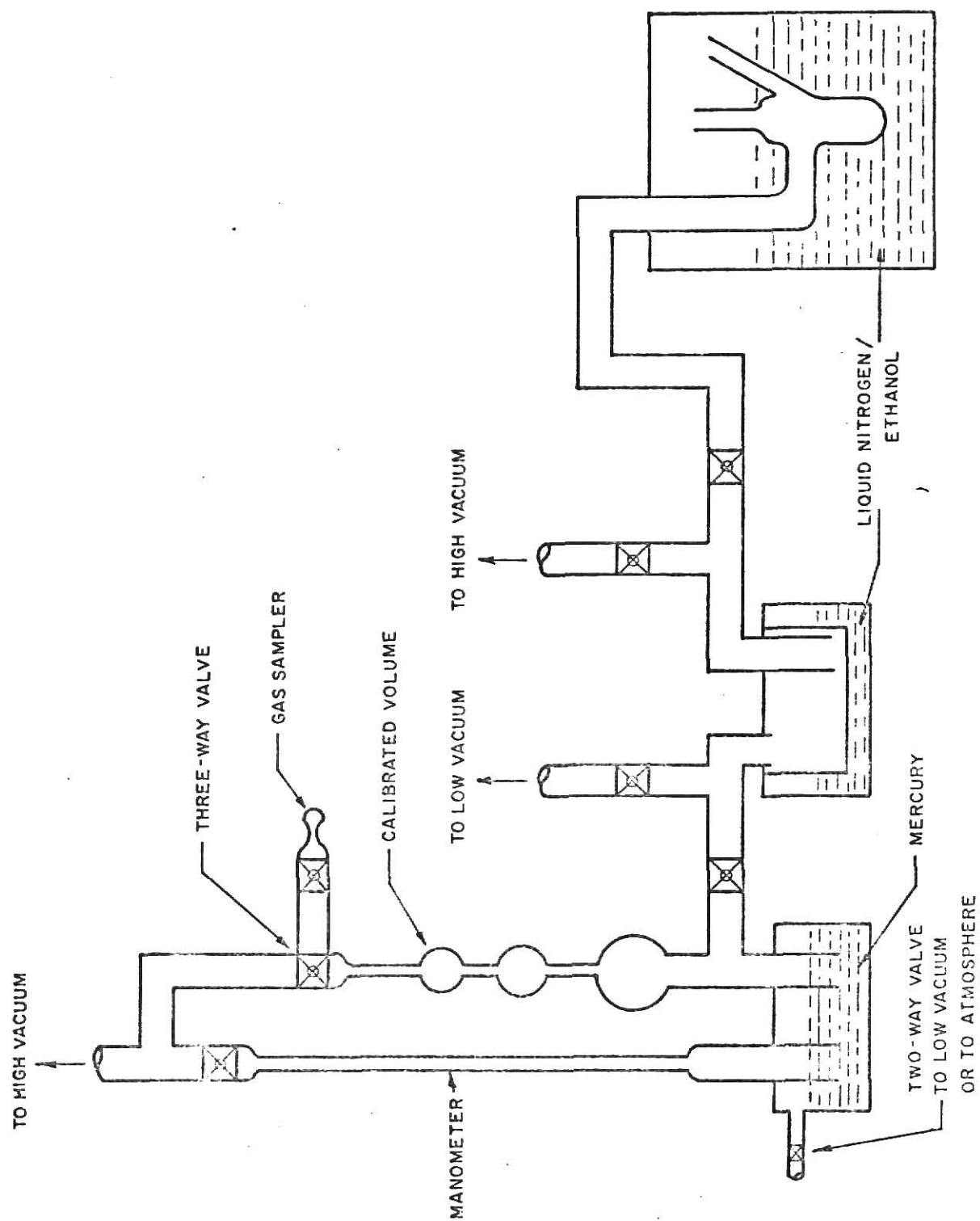


Fig. 4: Schematic view of Saunders-Taylor apparatus

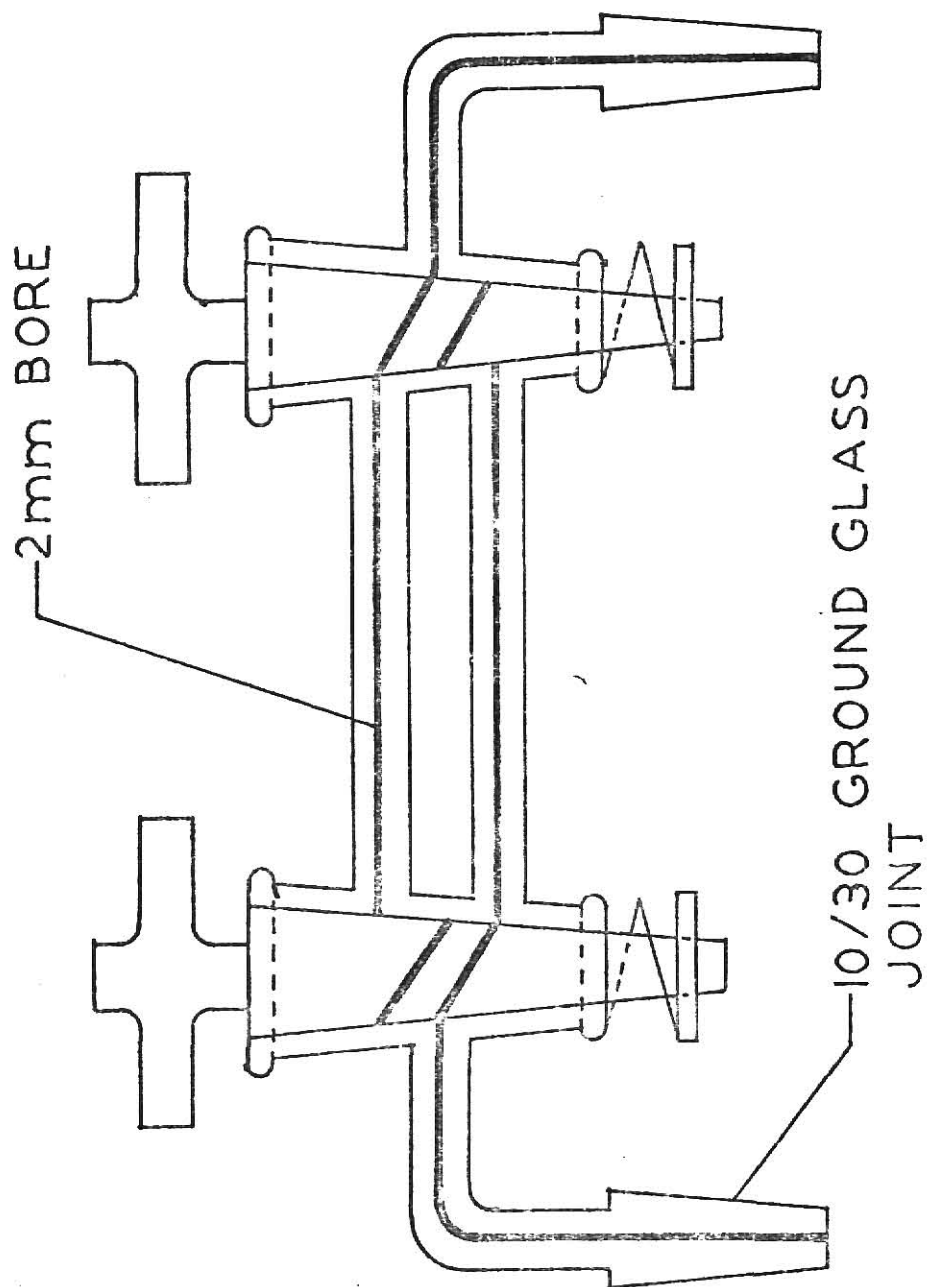


Fig. 5: Schematic view of gas sampler

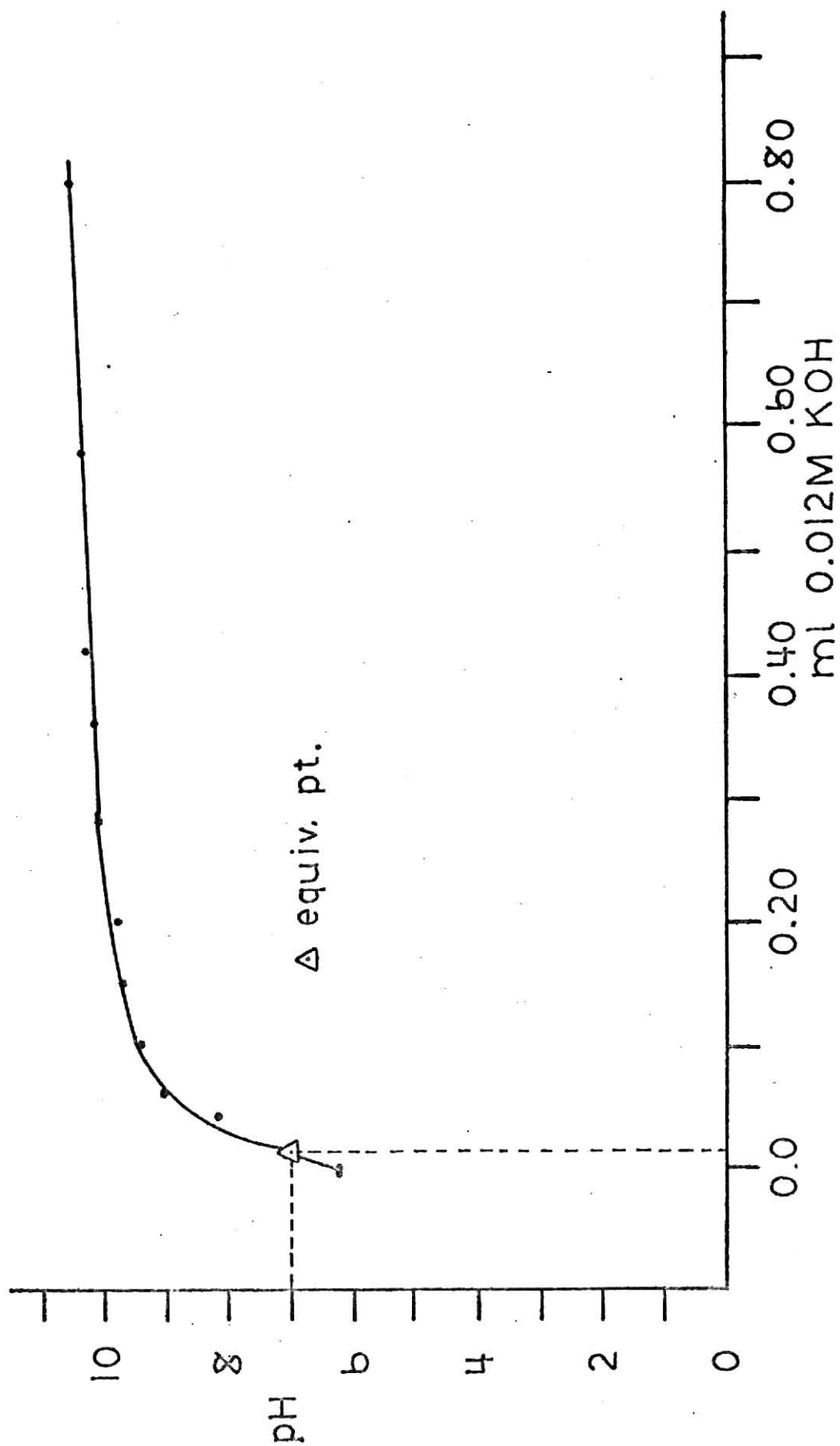


Fig. 6: Titration curve for a 10 ml sample of H_2O_2 .

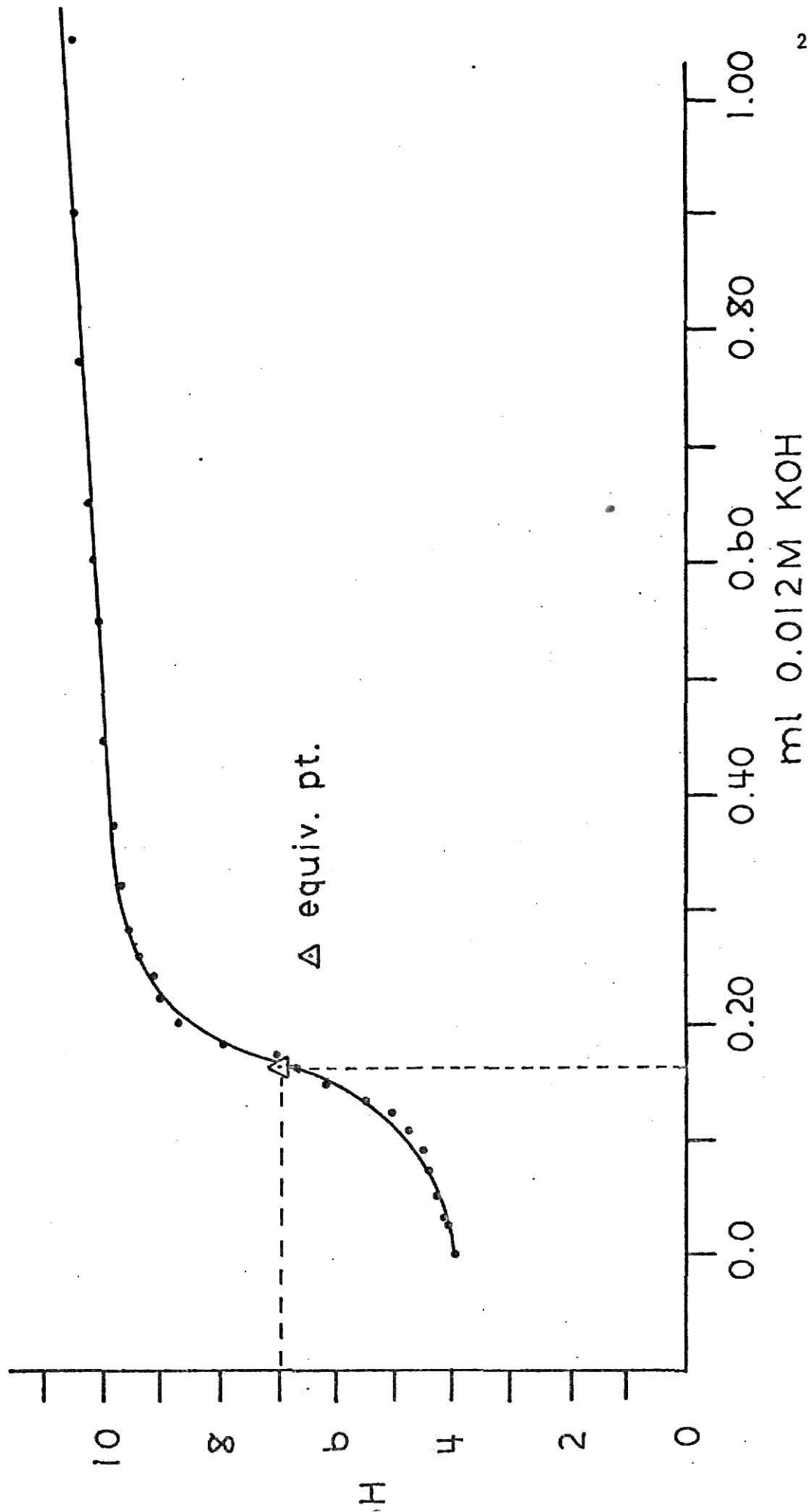


Fig. 7: Titration curve for 2 ml of TBP in 10 ml of H_2O .

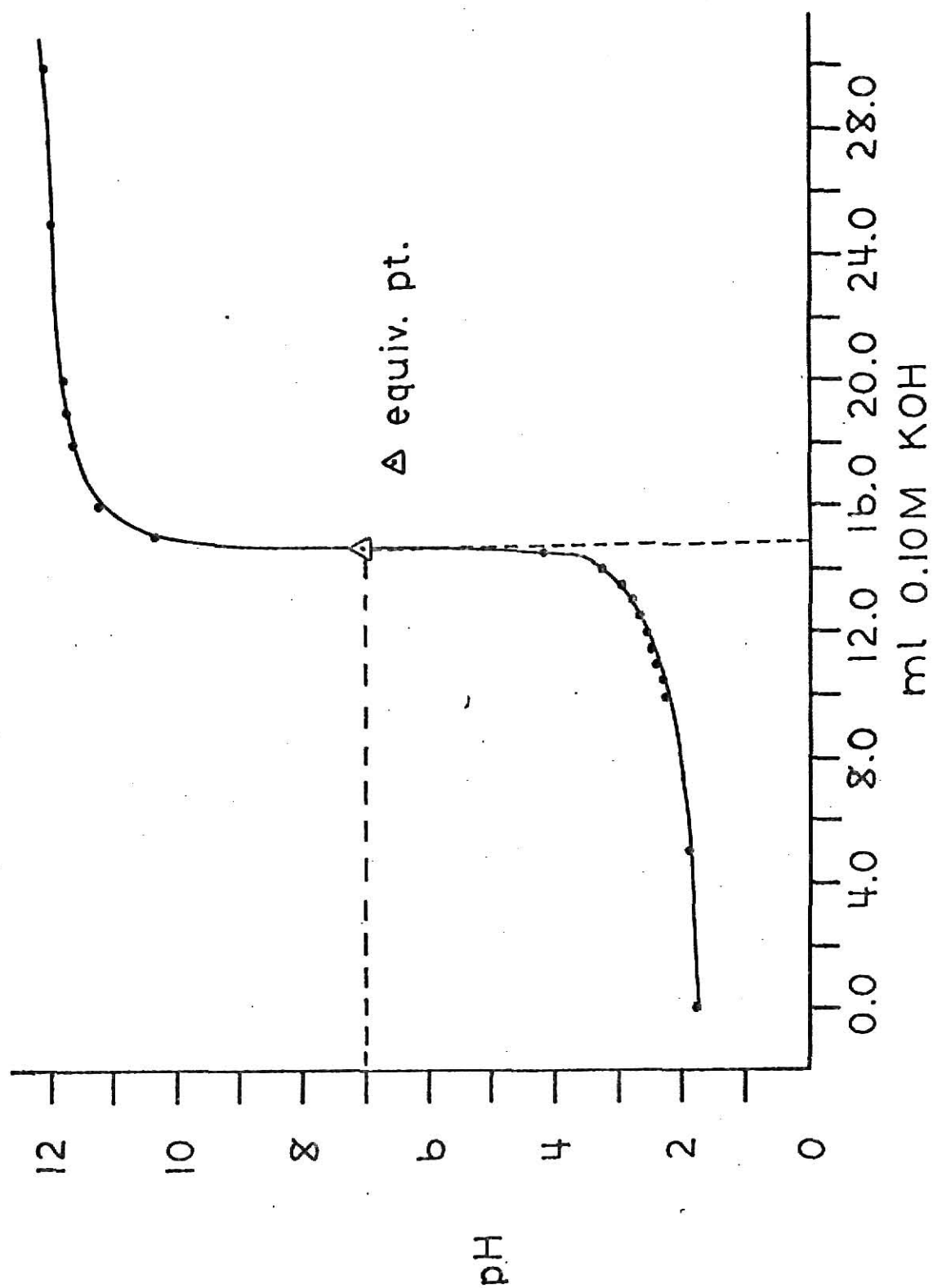


Fig. 8: Titration curve for 15 ml of 0.1N DBP.

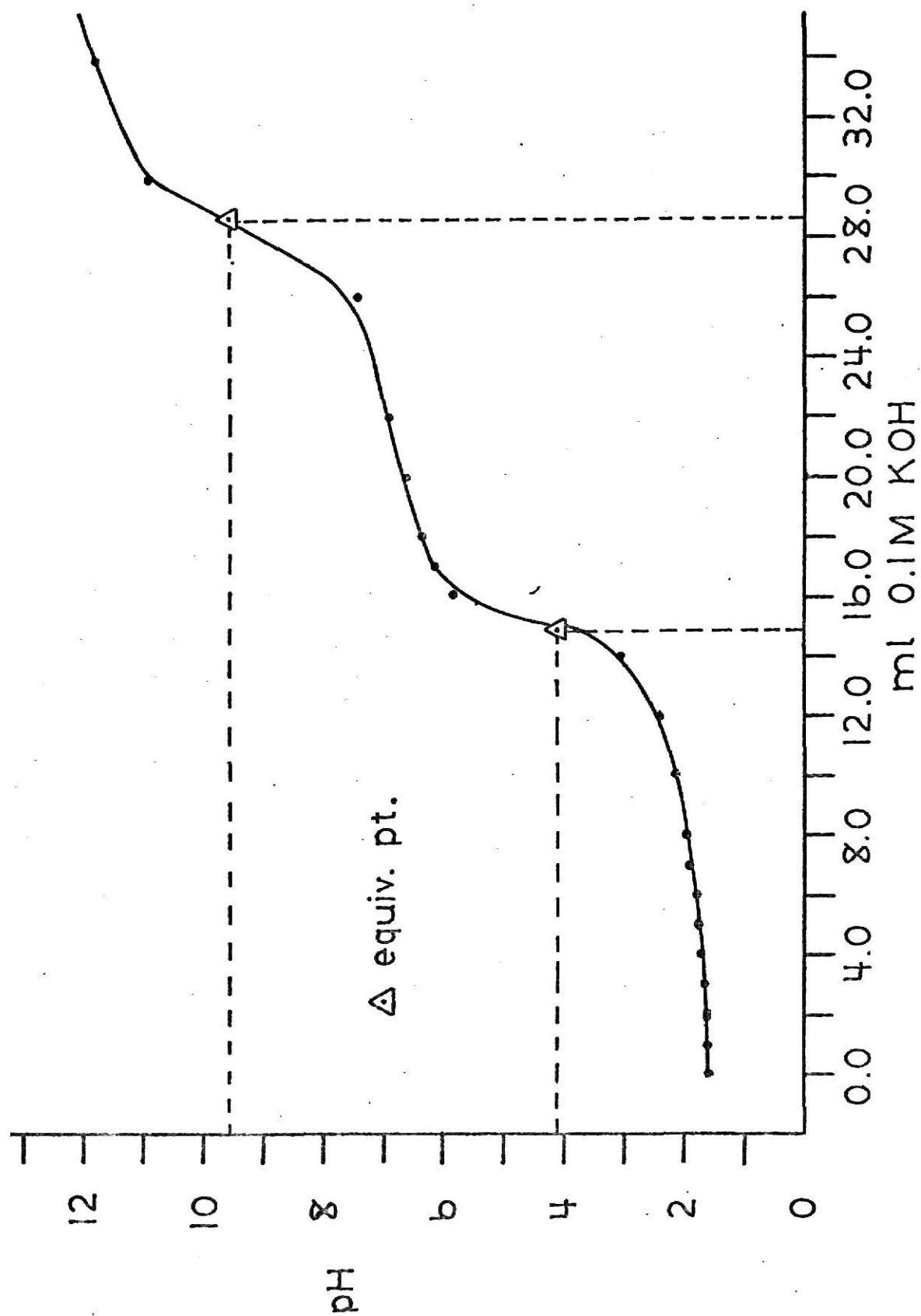


Fig. 9: Titration curve for 15 ml of 0.1N MBP.

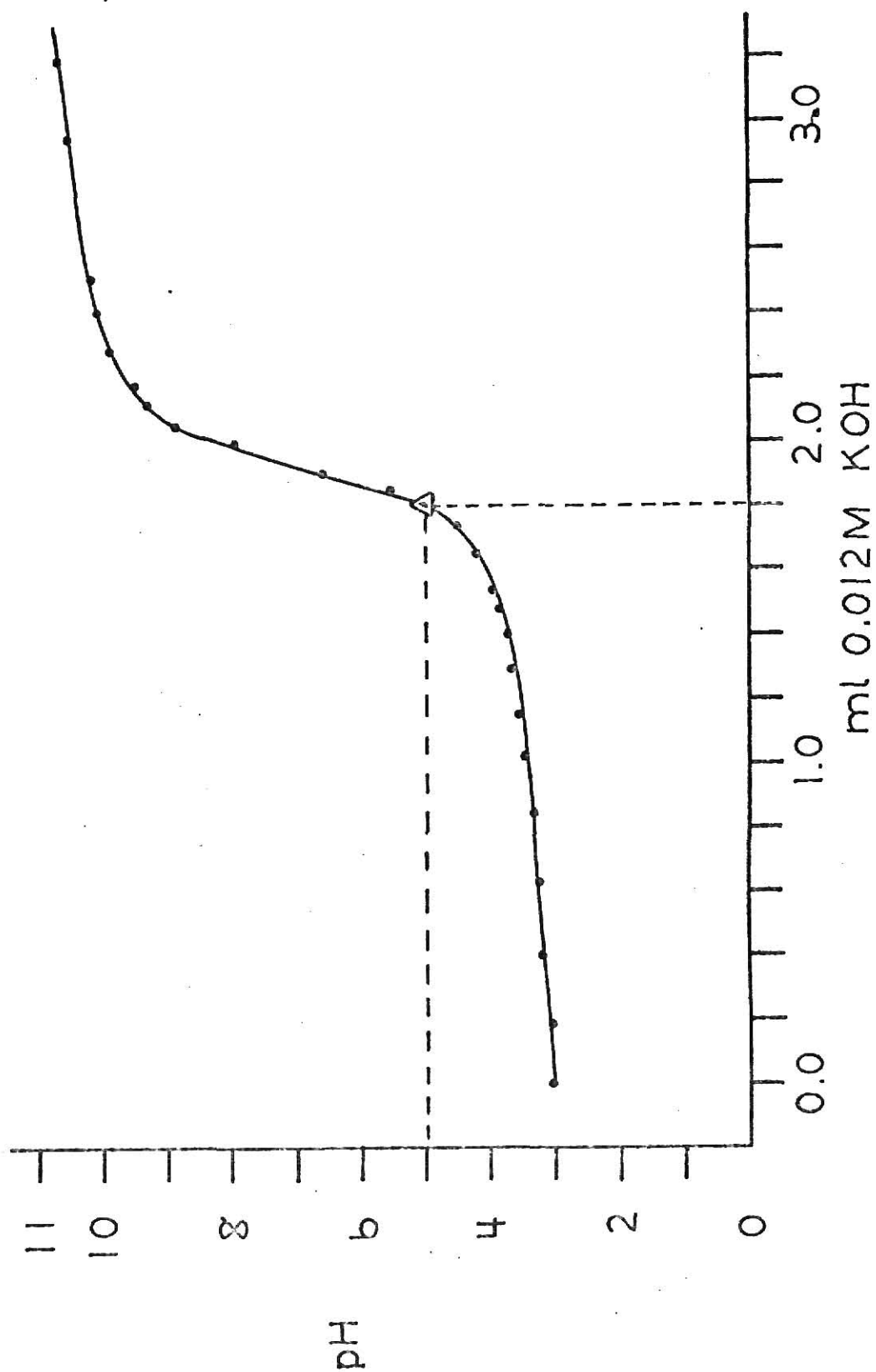


Fig 10: Titration curve for 2 ml of irradiated TBP in 10 ml of H_2O .
(TBP sample T-182)

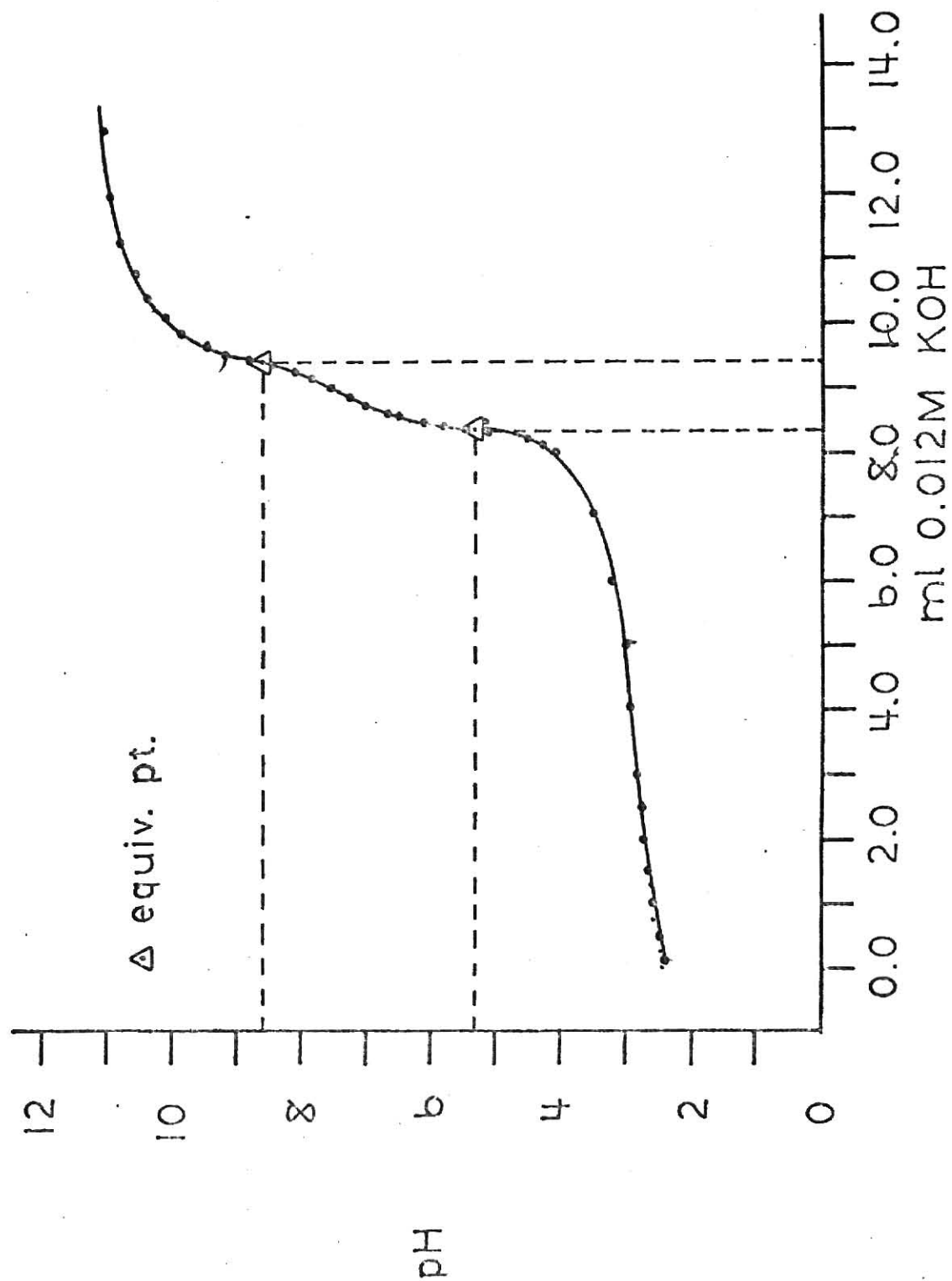


Fig. 11: Titration curve of 10 ml of 0.01N DBP and 0.002N MBP.

4.0 RESULTS AND CONCLUSIONS

This section deals with the results of the irradiation of dodecane, tributyl phosphate (TBP), and mixtures of the two and an analysis of the results of the irradiations. A reaction mechanism inferred by these results is proposed.

4.1 Dodecane

There is little reported data on the yields of hydrogen and methane from dodecane. Uncertainties concerning the effects of different experimental techniques also confuse the evaluation of the literature. For these reasons, the yield of hydrogen and methane from dodecane were determined. Degassed 5 ml samples were irradiated at absorbed dose rates varying from 4.1×10^{16} eV/g-min to 4.9×10^{16} eV/g-min and to absorbed doses varying from 6.1×10^{18} eV/g to 1.75×10^{20} eV/g.

A plot of G (hydrogen) as a function of dose is shown in Fig. 12. The solid lines shown on Fig. 12 are results of a least squares fit and the dashed lines are the standard deviation. No statistically significant dose dependence was observed for the hydrogen yield. Therefore, the yield of hydrogen was averaged over all doses. The average yield of hydrogen was $G(\text{H}_2) = 6.71$ with a standard deviation of 0.23. The methane yield was detectable, but no conclusion concerning dose dependence could be made. The average methane yield was, $G(\text{CH}_4) = 0.051$ and a standard deviation of 0.017.

4.2 TBP

Degassed 5 ml samples were irradiated at absorbed dose rates varying from 5.2×10^{16} eV/g-min to 6.0×10^{16} eV/g-min and to absorbed doses varying from 2.2×10^{19} eV/g to 1.75×10^{20} eV/g. A plot of $G(\text{H}_2)$ as a function of dose is shown in Fig. 12. No statistically significant dose dependence was observed for the hydrogen yield. Therefore, averaging over

doses, $G(H_2) = 2.02$ with a standard deviation of 0.13. Methane was also observed, but the yield was less than from dodecane. The average methane yield, $G(CH_4)$, was 0.032 with a standard deviation of 0.008.

A titration was performed on each irradiated sample of TBP to determine the yield of dibutyl phosphate (DBP) and monobutyl phosphate (MBP). At no absorbed dose was the MBP fraction of the total acid yield greater than 10%, which was determined to be the limit of MBP resolution for the titrimetric analysis (see Section 3.4). Since the DBP and MBP could not be resolved, the results of the titrations are reported as the yield of total acid. A plot of $G(\text{acid})$ as a function of dose is shown in Fig. 12.

A slight dose dependence was observed for the yield of acid but was statistically insignificant. Therefore, averaging over all doses, acid yield, $G(\text{acid})$, was 3.71 with a standard deviation of 0.21.

4.3 Mixtures

Mixtures of TBP and dodecane, with electron fractions of dodecane of 40%, 70%, 80%, 90%, and 95% were prepared. Degassed 5 ml samples were irradiated at absorbed dose rates varying from 4.1×10^{16} eV/g-min to 6.0×10^{16} eV/g-min and to absorbed doses varying from 1.0×10^{19} eV/g to 5.0×10^{19} eV/g. These samples were analyzed for gases volatile at 161 °K and amount of acid. Hydrogen and methane were the observed gaseous products and MBP was not resolvable from DBP.

Plots of $G(H_2)$ and $G(\text{acid})$ as a function of electron fraction of dodecane of TBP are shown in Fig. 13 and Fig. 14 respectively. These plots are nonlinear, indicating a significant deviation from the mixture law. The yield of hydrogen is less than would be predicted by the mixture law and the yield of acid is greater. The depressed hydrogen yield and increased

acid yield indicates that the two components are interacting and that TBP is being decomposed as a result.

4.4 Discussion

The depressed hydrogen yields and increased acid yields in the dodecane/TBP mixtures indicates that sensitized decomposition of TBP is taking place and that there is the possibility of a dodecane protection reaction occurring.

The dodecane protection reaction is inferred by the decrease in hydrogen yield in the presence of TBP. Confirmation of protection of dodecane by measurement of dodecene degradation products such as dodecene, dodecanol, and dodecane dimer was not done. This was due to the lack of a gas chromatograph capable of separating high molecular weight alkanes, olefins, and alcohols. However, all samples were saved for future analysis.

The yield of dodecane degradation products and how they are affected by sensitized decomposition or protection reactions is quite important to the Purex process since they radically effect process efficiency and are quite difficult to remove. Dodecene could be a particular problem with high burn up fuels where a significant amount of radioiodine, a prevalent fission product, would be present. Iodine readily adds across the double bounds of an olefin like dodecene and the halogenated dodecene's effect on the process is not known. It might interfere with decontamination of the fissile species or the presence of the halogen group might be exploitable as a means to scrub the solvent stream of dodecene prior to solvent recycle.

The effect of sensitized TBP decomposition on a Purex process could also be important, due to increased DBP and MBP yield.

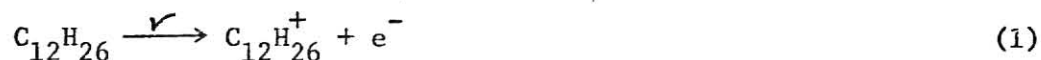
DBP, a strong monoprotic acid, causes a reduction in the attainable decontamination factor due to the zirconium and niobium complexes which are not scrubbed out by nitric acid washes [24].

At the dose rates encountered in this work, less than 10% of the total acid yield was MBP. However, there is a possible dose rate dependence which could increase the MBP yield significantly at very high fuel activities. This could be quite important since MBP forms a complex with plutonium that is insoluble in both the organic and aqueous phases. Large, high burn up Purex plants could then have considerable problems with accidental nuclear criticality due to deposition of this MBP/Pu complex in the process piping.

4.5 Mechanism

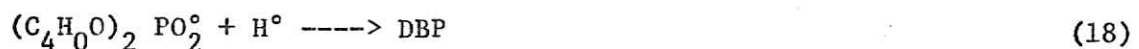
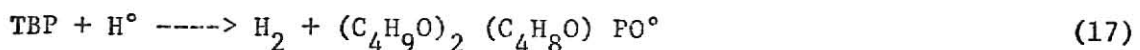
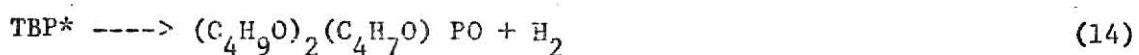
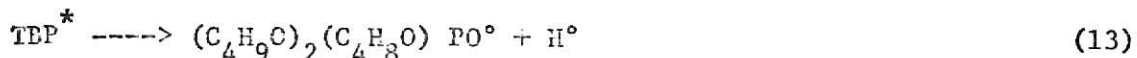
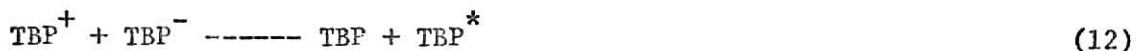
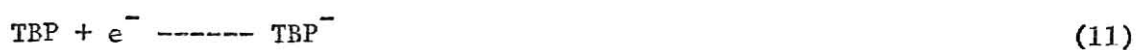
Making the assumption that the energy absorbed by a component is directly proportional to the electron fraction, a reaction scheme is proposed to account for the observed results. The proposed mechanism is in three parts, the pure dodecane reactions, TBP reactions and the reactions coupling TBP and dodecane.

The dodecane reactions are:



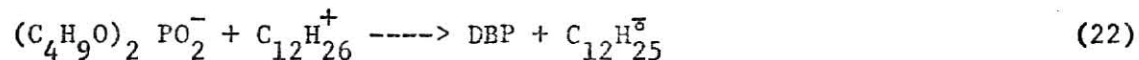
Reactions (1) and (2) produce the primary intermediate species. Reactions (4), (5), and (7) are the hydrogen producing reactions. Reaction (6) is the only significant hydrogen consumption reaction in pure dodecane. Disproportionation and dimerization of $C_{12}H_{25}^\circ$ are not dealt with since the yield of olefins and dimer were not measured in this work. Also, there was insufficient data to include methane production in this reaction mechanism.

The TBP reactions are:



Reactions (8) thru (12) are the producers of the primary intermediates. Reactions (13), (14), and (17) are hydrogen producing. Reactions (16), (18), and (19) are hydrogen consuming. Methane, TBP dimer, and some TBP radicals are not considered due to lack of data.

Reactions between TBP and dodecane are shown below:



If reaction (11) takes place and it utilizes electrons produced from dodecane it can produce an ionized form and depress the amount of excited state dodecane. Also, the resulting TBP^- can lead to the production of a DBP precursor and a butyl radical that are hydrogen consuming, this is shown in reaction (21). In reaction (22) hydrogen is abstracted from the dodecane, forming a dodecane radical which will consume hydrogen. MBP production was neglected due to the lack of experimental evidence of a diprotic acid in any of the titrations.

4.6 Summary

The average yield of hydrogen from dodecane was found to be, $G(\text{H}_2) = 6.71$ with a standard deviation of 0.23, as compared to the $G(\text{H}_2) = 4.9$ in Ref. [11]. The yield of hydrogen for TBP was, $G(\text{H}_2) = 2.02$ with a standard deviation of 0.13, this falls within the range of values reported in literature. The acid yield was, $G(\text{Acid}) = 3.71$ with standard deviation of 0.21, this was well above literature values. The yield of hydrogen in the mixtures was less than expected by mixture law and the acid yields are greater than expected by mixture law, therefore sensitized decomposition of TBP is taking place.

4.7 Further Work

The identity and the yields of dodecane degradation products need to be investigated. This information would help refine the proposed reaction

mechanisms and confirm or deny dodecane protection. A more sophisticated gas chromatograph than was available for this work may be able to perform this analysis, but it will be complicated by the presence of TBP in mixtures.

Gas chromatograph analysis of the gaseous radiolysis products uncovered small air leaks in a few of the samples; however, the measured oxygen fraction was far smaller than the fraction encountered in normal air. Apparently, oxygen consumption took place during irradiation. Since full scale Purex operations take place in an air saturated environment, this effect should be explored.

Another area needing investigation is that of high Linear Energy Transfer irradiation of dodecane and TBP. A proposed technique is neutron bombardment of boron doped samples of dodecane or TBP to produce in situ alpha irradiation. High Linear Energy Transfer irradiation might have significantly different radiolysis yields due to much higher local dose rates encountered inside the radiation track. MBP yields could be significantly affected.

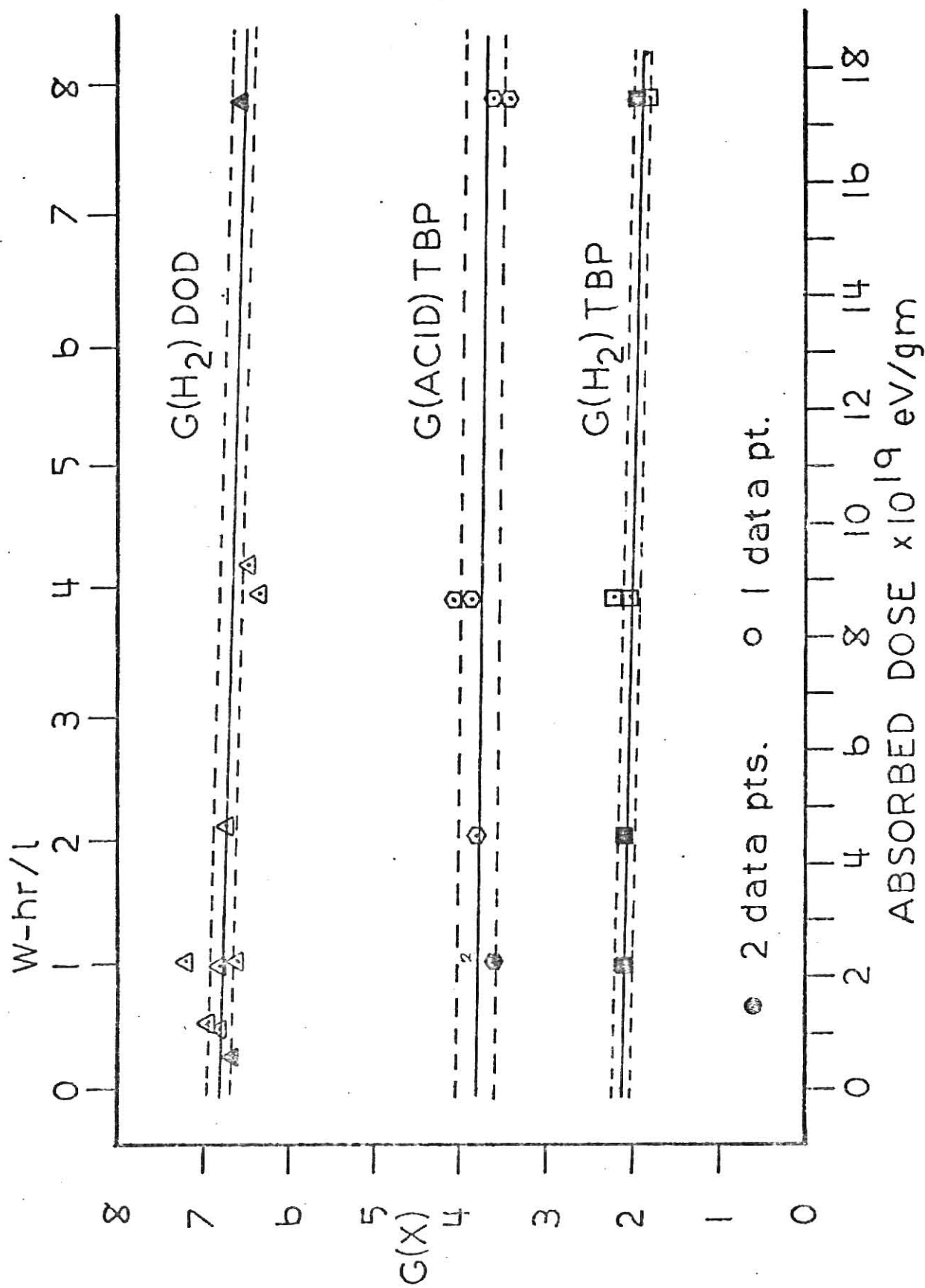


Fig. 12: Plot of product yields as a function of dose for TBP and dodecane.

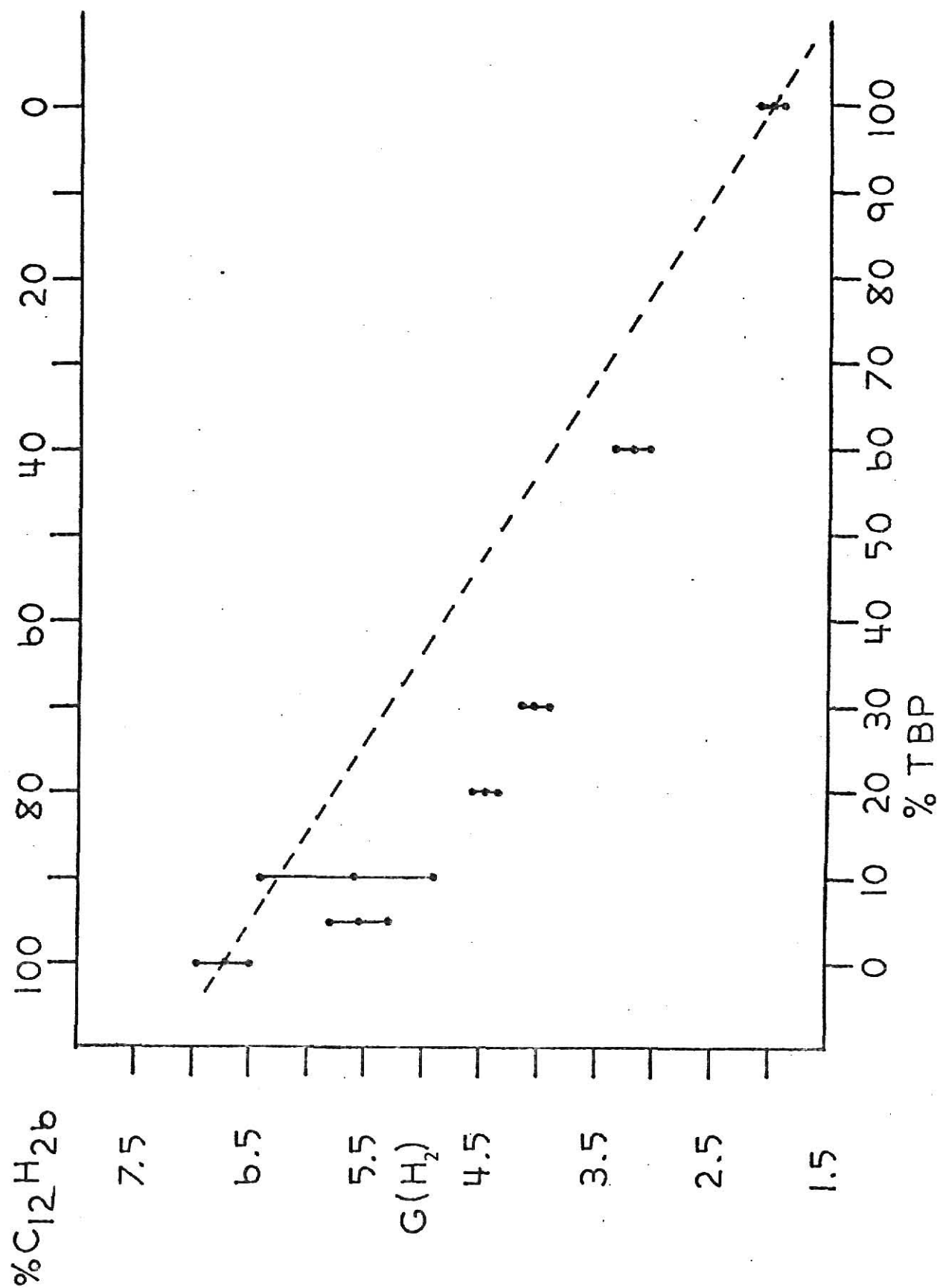


Fig. 13: Plot of $G(H_2)$ as a function of electron fraction TBP in Dodecane, absorbed dose, 2 watt-hr/liter.

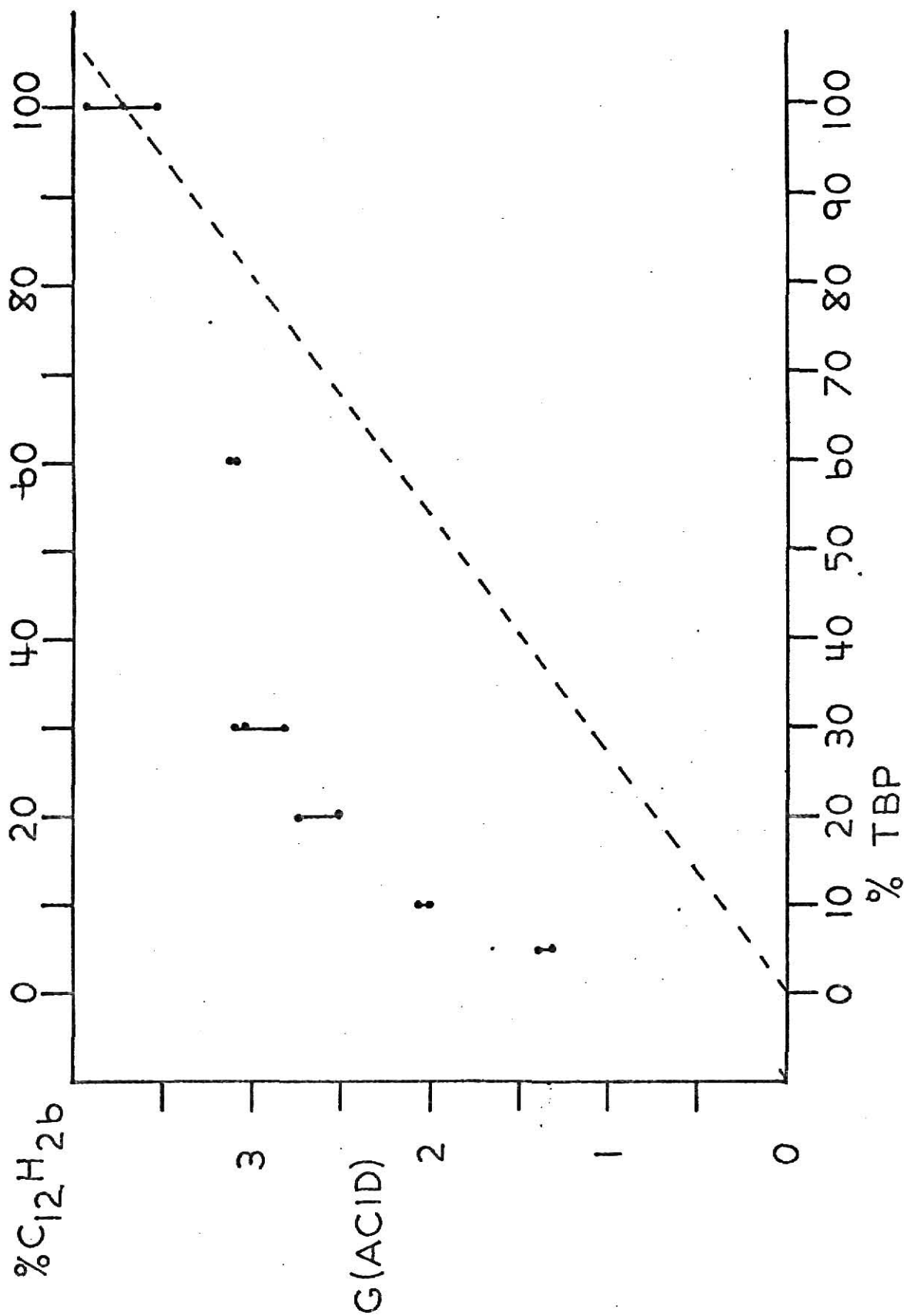


Fig. 14: Plot of G(Acid) as a function of electron fraction TBP in Dodecane, absorbed dose, 2 watt-hr/liter.

5. BIBLIOGRAPHY

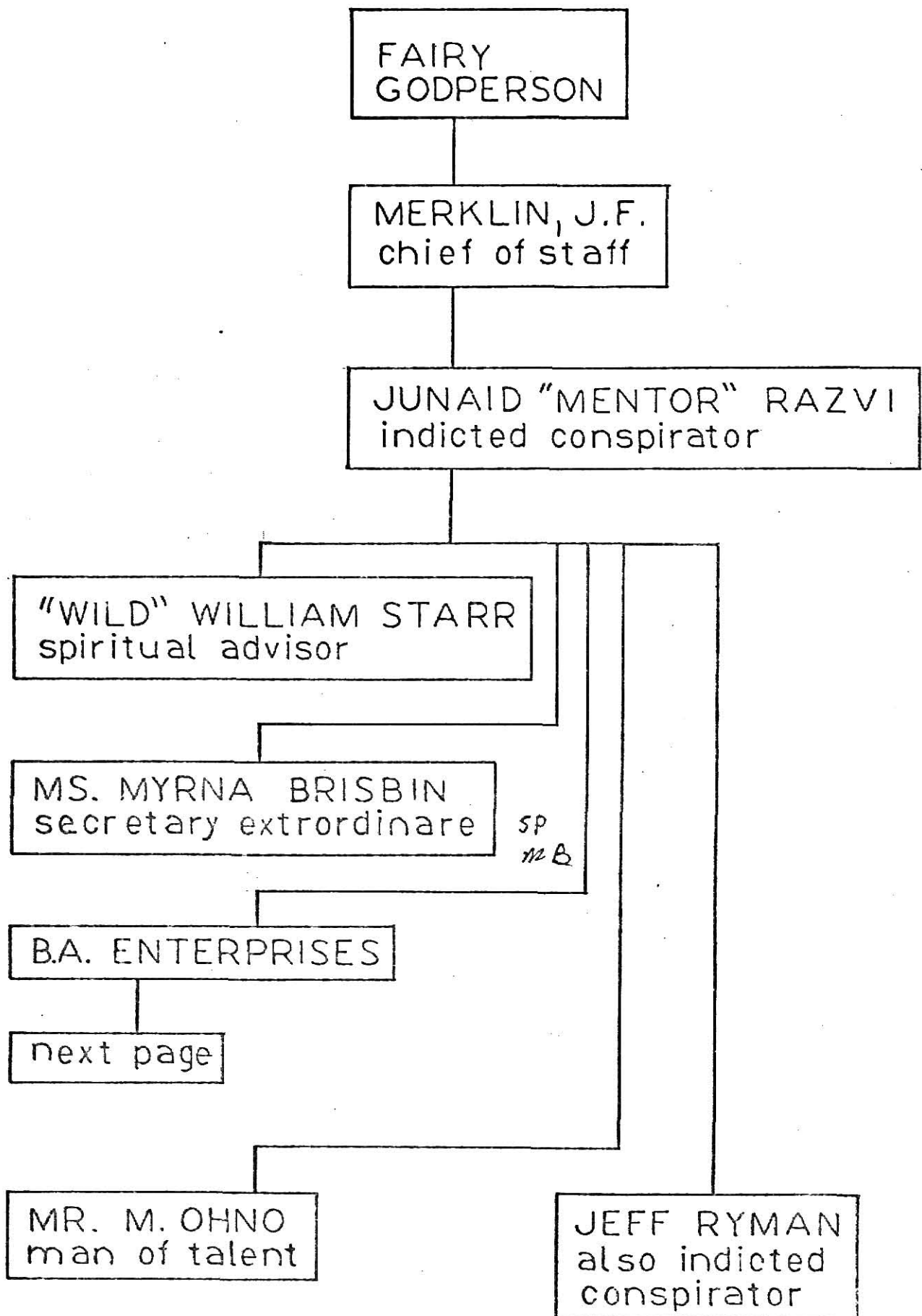
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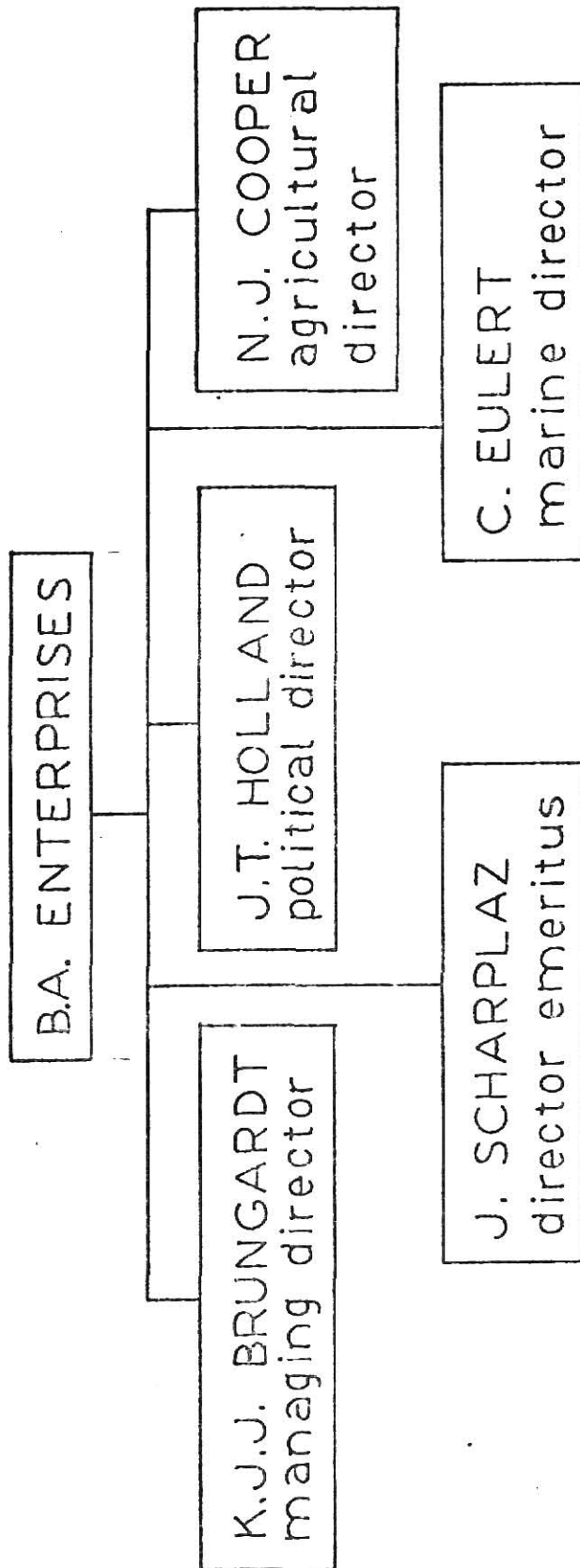
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I would like to thank my Fairy Godperson and staff for getting me thru my masters. Table of organization on following two pages.

I would like to also thank the Nuclear Engineering Department faculty for causing me to do this thesis, it was one of the luckiest things to happen to me.





APPENDIX A

Some Physical Properties of the TBP-Dodecane System.

Table A-1 Physical properties of butyl phosphates.

Property	TBP	DBP	MBP
*molecular weight	263.3	210.2	154.1
*density (gm/cm ³)	0.97	1.065	1.22
*viscosity (mp, 25 °C)	33	520	4300
**boiling point	289 °C		
***pK		1.72	1.89 & 6.84
**solubility in water (gm/liter)	0.2		

*[2]

**[25]

***[18]

Table A-2 Physical properties of dodecane*

Property	Dodecane
molecular weight	170.34
melting point	-9.6 °C
boiling point	216.3 °C
density	0.7487 (gm/cm ³)

*[26]

Table A-3 TBP-Dodecane mixture data.

TBP Electron Fraction	Dodecane Electron Fraction	TBP v/o	Dodecane v/o	Density (gm/cm ³)	Dose Correction Factor*
100	0	100	0.0	0.9727	0.9407
60	40	54.8	45.2	0.871	0.8589
30	70	25.7	74.3	0.8063	0.8063
20	80	16.8	83.2	0.7863	0.7902
10	90	8.2	91.8	0.7671	0.7747
5	95	4.0	95.9	0.7569	0.7672
0	100	0.0	100.0	0.7487	0.7598

*Dose Correction Factor corrects for the difference in electron density of these mixtures and the Fricke dosimeter solution.

APPENDIX B: UV spectrophotometer Results

Figure B-1 shows the results of UV spectrophotometric analysis of practical grade dodecane and dodecane after purification. Included on Figure B-1 is a plot of the maximum permissible absorbance for spectrophotometric grade dodecane as given in the Aldrich Catalog-Handbook of Organic and Biochemicals, 1977-1978, Aldrich Chemical Company, Inc. This plot indicates that the purified dodecane used in this work is near spectrophotometric grade in purity.

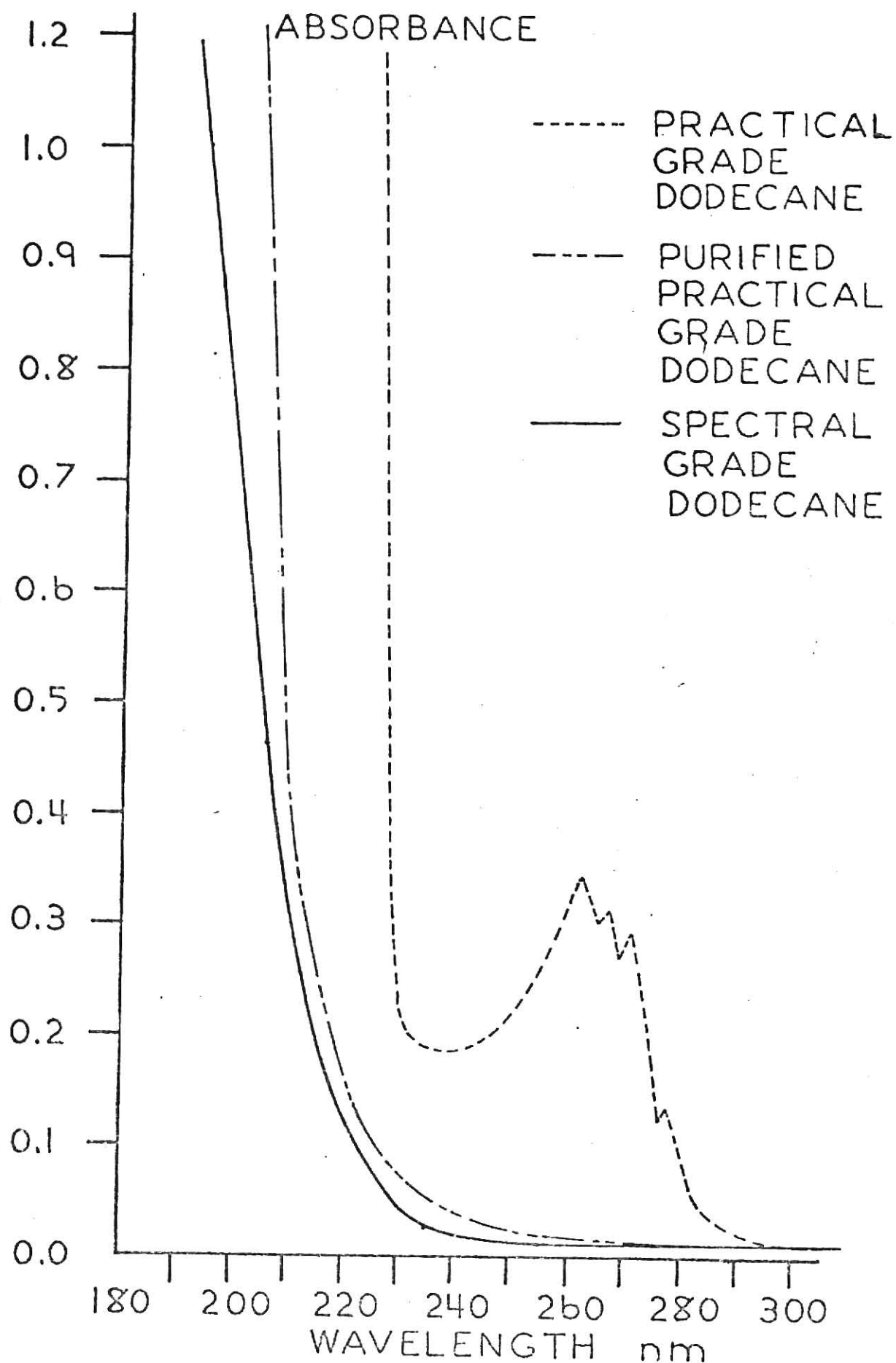


Fig. B-1: UV spectrophotometric results for spectral grade, purified practical grade, and unpurified practical grade dodecane.

APPENDIX C: Dose Rate Program

A sample output from the program used to calculate the dose rate is shown on the following pages. The program first calculates the dose rate for the twelve positions of sample holder on January 30, 1976, this is done by using the results of eighteen Fricke dosimeter runs that were preformed over a period of six months. Each Fricke measurement is corrected to the dose rate of 1/30/76 by a decay correction factor.

The next step of the program is the calculation of dose rate for the irradiated sample. This is done by correcting the January 30, 1976 dose rate at each position to the date of the sample irradiation by means of a decay correction factor. The resulting dose rates are those that would be absorbed by a Fricke dosimeter solution on that specific date, so a correction factor for the difference in electron density of Fricke solution and irradiated sample must be applied. This correction factor is listed in Appendix A, Table A-3.


```

1  SJOB
2  JR
3  IMPLICIT REAL*(A-L,D-Z)
4  DIMENSION X(40),TYPE(40),CY(40),SIGCY(40),LAMDAT(40),XLAM(40),TITL
5  IF(40),TYM(40),SAMPLE(40)
6  WRITE(6,14)
7  18  FORMAT(1F1)
8  EC=2121.5500
9  SIGEL=8.7210
10  GFE3=15.600
11  SIGFE=C.CDO
12  RPO=1024.500
13  SIGRFC=1.500
14  TIME=20.CDC
15  TMO=2.CDC
16  YEAR=365.000
17  LAM=5.26300
18  LAMDA = DLOG(TMO)/(LAM * YEAR)
19  999 READ(5,1,END=99)NPIS,TITL
20  NPIS = # OF FRICKE RUNS MADE AT THIS POSITION
21  TITL = POSITION IN THE GAMMA CELL
22  C
23  C
24  C
25  WRITE(6,17)
26  17  FORMAT(1HC)
27  READ(5,2)(X(M),M=1,NPIS)
28  READ(5,2)(TYME(M),M=1,NPIS)
29  M=C
30  NP=C
31  NP=N+1
32  NP=NF+1
33  READ(5,48)SAMPLE(M),TYM(M),NPI
34  IF(NPI.NE.0.C) GO TO 339
35  48  FORMAT(A8,F10.5,I2)
36  C
37  C
38  C
39  X(M) = MEASURED ABSORBANCE OF FRICKE SOLUTION
40  TYME(M) = NUMBER OF DAYS AFTER 1/30/76 THAT THE GIVEN FRICKE RUN WAS MADE
41  TYPE(M) IS THE NUMBER OF DAYS AFTER 1/30/76 THAT A DOSE RATE IS WANTED
42  SAMPLE(M) IS THE I.D. NUMBER OF SAMPLE(ALPHANUMERIC)
43  C
44  C
45  C
46  29  FORMAT(12,A4)
47  2  FORMAT(5F10.5)
48  SUM=C.CDO
49  SUMX=C.CDO
50  SIGMA=C.CDO
51  SIGMA=C.CDO
52  DO 32 M=1,NPIS
53  22  WEIGHT=1.CDO
54  31  SUM=SUM+WEIGHT
55  31  LAMDAT(M)=LAMDA * TYME(M)
56  31  XLAM(M)=X(M) * CEXP(LAMDAT(M))
57  31  SUMX=SUMX+WEIGHT*XLAM(M)
58  32  CONTINUE
59  C
60  C
61  C
62  EVALUATE DOSE RATES FOR EXPERIMENTALLY DETERMINED ABSORBANCES
63  41  XMEAN=SUMX/SUM
64  51  DO 52 M=1,NPIS
65  52  SIGMA=SIGMA+(XLAM(M)-XMEAN)**2
66  52  CONTINUE

```

```

46 FREE=NPTS-1
47 54 SIGMA= DSQRT(SIGMA/FREE)
48 64 SIGMA=SIGMA/DSQRT(SUM)
49 70 CONTINUE

C      EVALUATE CCSE RATES
C
50 DR=(XMEAN/ED)*(6.C23E23/RHC)*(100.0/TIME)*(1.0/GFE3)
51 SIGDR=(SIGMAM/XMEAN)**2+(SIGED/ED)**2+(SIGFE3/GFE3)**2+(SIGRHO/RHO)
52 11**2
53 SIGDR=DR*DSQRT(SIGDR)
54 DRAD=DR*1.602D-14
55 SIGRL=SIGDR*1.602D-14
56 CCNV=C.027341D-16
57 DRWL=DR*CCNV
58 SIGDWL=(SIGDR/DR)**2+2.1437D-6
59 SIGDWL=DRWL*DSQRT(SIGDWL)
60 WRITE(6,10)TTL
10 FCFORMAT(1X,POSITION,A4,2X,DATE,2X,2X,ABSCREANCE,4X,DOSE RATE
2W,1)
11 FCFORMAT(1X,POSITION,A4,2X,DATE,2X,2X,ABSCREANCE,4X,DOSE RATE
2W,1)
12 FCFORMAT(12X,F7.1,5X,F8.3,16X,D10.4,2X,C10.4,1X,D10.4,3X,D1
10.4,2X,C10.4)
63 DRY=DR
64 SIGDR)=SIGDR
65 DO 777 M=1,NP
66 LAMT=TYM(M)*LAMDA
67 DR=DR/DEXP(LAMT)
68 DR=DR/DEXP(LAMT)
69 SIGDR=SIGDR/DEXP(LAMT)
70 DRAD=DR*1.602D-14
71 SIGDR=DR*1.602D-14
72 DRWL=DR*CCNV
73 SIGDWL=(SIGDR/DR)**2+2.1437D-6
74 SIGDWL=DRWL*DSQRT(SIGDWL)
75 WRITE(6,11)TTL
76 FCFORMAT(1X,POSITION,A4,2X,DATE,2X,2X,ABSCREANCE,4X,DOSE RATE
2W,1)
77 FCFORMAT(12X,F7.1,5X,F8.3,16X,D10.4,2X,C10.4,1X,D10.4,3X,D10.
2W,1)
78 177
79 177
80 777 CONTINUE
81 GC TC 999
82 99 STOP
83 ENB

$ENTRY

```

19805

POSITION A	DATE	ABSORBANCE	DOSE RATE	EV/GM-MIN	SIGDR-EV	RAD/MIN	SIGDR-RAD	WAIT/L	SIGDR-W
POSITION A	DATE	0.787	DOSE RATE	0.6990	0.14520	0.11200	0.23260	0.19110	0.37900
POSITION A	277.0	SAMPLE #	DOSE RATE	EV/GM-MIN	SIGDR-EV	RAD/MIN	SIGDR-RAD	WAIT/L	SIGDR-W
POSITION A	DATE	C-141	DOSE RATE	0.63250	0.13140	0.10130	0.21050	0.17290	0.36010
POSITION A	295.0	SAMPLE #	DOSE RATE	EV/GM-MIN	SIGDR-EV	RAD/MIN	SIGDR-RAD	WAIT/L	SIGDR-W
POSITION A	DATE	REF	DOSE RATE	0.62840	0.13050	0.10070	0.20910	0.17180	0.35770

POSITION B	DATE	ABSORBANCE	DOSE RATE	EV/GM-MIN	SIGDR-EV	RAD/MIN	SIGDR-RAD	WAIT/L	SIGDR-W
POSITION B	DATE	0.791	DOSE RATE	0.70230	0.14720	0.11250	0.23580	0.19200	0.40330
POSITION B	DATE	C-121	DOSE RATE	EV/GM-MIN	SIGDR-EV	RAD/MIN	SIGDR-RAD	WAIT/L	SIGDR-W
POSITION B	DATE	SAMPLE #	DOSE RATE	0.63780	0.13360	0.10220	0.21410	0.17440	0.36630
POSITION B	318.0	SAMPLE #	DOSE RATE	EV/GM-MIN	SIGDR-EV	RAD/MIN	SIGDR-RAD	WAIT/L	SIGDR-W
POSITION B	DATE	D-95.51	DOSE RATE	0.62610	0.13120	0.10030	0.21020	0.17120	0.35960

POSITION C	DATE	ABSORBANCE	DOSE RATE	EV/GM-MIN	SIGDR-EV	RAD/MIN	SIGDR-RAD	WAIT/L	SIGDR-W
POSITION C	DATE	0.790	DOSE RATE	0.70170	0.14490	0.11240	0.23210	0.19190	0.37100
POSITION C	DATE	SAMPLE #	DOSE RATE	EV/GM-MIN	SIGDR-EV	RAD/MIN	SIGDR-RAD	WAIT/L	SIGDR-W
POSITION C	DATE	C-122	DOSE RATE	0.63720	0.13160	0.10210	0.21080	0.17420	0.36060
POSITION C	DATE	SAMPLE #	DOSE RATE	EV/GM-MIN	SIGDR-EV	RAD/MIN	SIGDR-RAD	WAIT/L	SIGDR-W
POSITION C	DATE	D-1.22	DOSE RATE	0.63500	0.13110	0.10170	0.21000	0.17360	0.35930
POSITION C	DATE	SAMPLE #	DOSE RATE	EV/GM-MIN	SIGDR-EV	RAD/MIN	SIGDR-RAD	WAIT/L	SIGDR-W
POSITION C	DATE	T-111	DOSE RATE	0.63410	0.13090	0.10160	0.20970	0.17340	0.35880
POSITION C	DATE	SAMPLE #	DOSE RATE	EV/GM-MIN	SIGDR-EV	RAD/MIN	SIGDR-RAD	WAIT/L	SIGDR-W
POSITION C	DATE	D-113	DOSE RATE	0.63380	0.13090	0.10150	0.20960	0.17330	0.35870
POSITION C	DATE	SAMPLE #	DOSE RATE	EV/GM-MIN	SIGDR-EV	RAD/MIN	SIGDR-RAD	WAIT/L	SIGDR-W
POSITION C	DATE	T-122	DOSE RATE	0.63160	0.13040	0.10120	0.20890	0.17270	0.35740
POSITION C	DATE	SAMPLE #	DOSE RATE	EV/GM-MIN	SIGDR-EV	RAD/MIN	SIGDR-RAD	WAIT/L	SIGDR-W
POSITION C	DATE	T-183	DOSE RATE	0.63130	0.13030	0.10110	0.20880	0.17260	0.35730
POSITION C	DATE	SAMPLE #	DOSE RATE	EV/GM-MIN	SIGDR-EV	RAD/MIN	SIGDR-RAD	WAIT/L	SIGDR-W
POSITION C	DATE	D-421	DOSE RATE	0.63060	0.13020	0.10100	0.20860	0.17240	0.35690
POSITION C	DATE	SAMPLE #	DOSE RATE	EV/GM-MIN	SIGDR-EV	RAD/MIN	SIGDR-RAD	WAIT/L	SIGDR-W
POSITION C	DATE	C-7.51	DOSE RATE	0.62840	0.12970	0.10070	0.20780	0.17180	0.35560
POSITION C	DATE	SAMPLE #	DOSE RATE	EV/GM-MIN	SIGDR-EV	RAD/MIN	SIGDR-RAD	WAIT/L	SIGDR-W
POSITION C	DATE	C-9.51	DOSE RATE	0.62680	0.12940	0.10040	0.20730	0.17140	0.35470
POSITION C	DATE	SAMPLE #	DOSE RATE	EV/GM-MIN	SIGDR-EV	RAD/MIN	SIGDR-RAD	WAIT/L	SIGDR-W
POSITION C	DATE	D-9.53	DOSE RATE	0.62570	0.12920	0.10020	0.20690	0.17110	0.35410

POSITION D	DATE	ABSORBANCE	DOSE RATE	EV/GM-MIN	SIGDR-EV	RAD/MIN	SIGDR-RAD	WAIT/L	SIGDR-W
POSITION D	DATE	0.794	DOSE RATE	0.70530	0.14620	0.11300	0.23410	0.19280	0.40800
POSITION D	DATE	SAMPLE #	DOSE RATE	EV/GM-MIN	SIGDR-EV	RAD/MIN	SIGDR-RAD	WAIT/L	SIGDR-W
POSITION D	DATE	C-112	DOSE RATE	0.63580	0.13270	0.10250	0.21250	0.17490	0.36360
POSITION D	DATE	SAMPLE #	DOSE RATE	EV/GM-MIN	SIGDR-EV	RAD/MIN	SIGDR-RAD	WAIT/L	SIGDR-W
POSITION D	DATE	C-1.21	DOSE RATE	0.63820	0.13230	0.10220	0.21200	0.17450	0.36270
POSITION D	DATE	SAMPLE #	DOSE RATE	EV/GM-MIN	SIGDR-EV	RAD/MIN	SIGDR-RAD	WAIT/L	SIGDR-W
POSITION D	DATE	T-112	DOSE RATE	0.63720	0.13210	0.10210	0.21170	0.17420	0.36220
POSITION D	DATE	SAMPLE #	DOSE RATE	EV/GM-MIN	SIGDR-EV	RAD/MIN	SIGDR-RAD	WAIT/L	SIGDR-W
POSITION D	DATE	T-143	DOSE RATE	0.63500	0.13170	0.10170	0.21090	0.17360	0.36090
POSITION D	DATE	SAMPLE #	DOSE RATE	EV/GM-MIN	SIGDR-EV	RAD/MIN	SIGDR-RAD	WAIT/L	SIGDR-W
POSITION D	DATE	C-422	DOSE RATE	0.63380	0.13140	0.10150	0.21050	0.17330	0.36020
POSITION D	DATE	SAMPLE #	DOSE RATE	EV/GM-MIN	SIGDR-EV	RAD/MIN	SIGDR-RAD	WAIT/L	SIGDR-W
POSITION D	DATE	C-7.52	DOSE RATE	0.63160	0.13090	0.10120	0.20980	0.17270	0.35890
POSITION D	DATE	SAMPLE #	DOSE RATE	EV/GM-MIN	SIGDR-EV	RAD/MIN	SIGDR-RAD	WAIT/L	SIGDR-W
POSITION D	DATE	C-9.52	DOSE RATE	0.63000	0.13060	0.10090	0.20920	0.17220	0.35800
POSITION D	DATE	SAMPLE #	DOSE RATE	EV/GM-MIN	SIGDR-EV	RAD/MIN	SIGDR-RAD	WAIT/L	SIGDR-W

POSITION	D	318.0 DATE	322.0 DATE	D-95.52 SAMPLE #	D-8.51 SAMPLE #	DOSE RATE:	0.6288D 17 EV/GM-MIN	0.6279D 17	0.1304D 16 SIGDR-EV	0.1302D 16	0.1007D 04 RAD/MIN	0.1006D 04	0.2089D 02 SIGDR-RAD	0.2086D 02	0.1719D 00 WAT/L	0.1717D 00	0.3574D-02 SIGDR-W	0.3568D-02
POSITION E		DATE	DATE	ABSORBANCE		DOSE RATE:	EV/GM-MIN		SIGDR-EV		RAD/MIN		SIGDR-RAD		WAT/L		SIGDR-W	
POSITION E		DATE	DATE	0.733 SAMPLE #		DOSE RATE:	0.6508D 17		0.1328D 16		0.1043D 04		0.2128D 02		0.1770D 00		0.3640D-02	
POSITION E		DATE	DATE	0.733 SAMPLE #		DOSE RATE:	EV/GM-MIN		SIGDR-EV		RAD/MIN		SIGDR-RAD		WAT/L		SIGDR-W	
POSITION E		DATE	DATE	0.733 SAMPLE #		DOSE RATE:	0.5910D 17		0.1206D 16		0.09468D 03		0.1932D 02		0.1616D 00		0.3106D-02	
POSITION E		DATE	DATE	0.733 SAMPLE #		DOSE RATE:	EV/GM-MIN		SIGDR-EV		RAD/MIN		SIGDR-RAD		WAT/L		SIGDR-W	
POSITION E		DATE	DATE	0.733 SAMPLE #		DOSE RATE:	0.5876D 17		0.1199D 16		0.09413D 03		0.1921D 02		0.1607D 00		0.3287D-02	
POSITION E		DATE	DATE	0.733 SAMPLE #		DOSE RATE:	EV/GM-MIN		SIGDR-EV		RAD/MIN		SIGDR-RAD		WAT/L		SIGDR-W	
POSITION E		DATE	DATE	0.733 SAMPLE #		DOSE RATE:	0.5855D 17		0.1175D 16		0.09379D 03		0.1914D 02		0.1601D 00		0.3275D-02	
POSITION E		DATE	DATE	0.733 SAMPLE #		DOSE RATE:	EV/GM-MIN		SIGDR-EV		RAD/MIN		SIGDR-RAD		WAT/L		SIGDR-W	
POSITION E		DATE	DATE	0.733 SAMPLE #		DOSE RATE:	0.5825D 17		0.1189D 16		0.09332D 03		0.1904D 02		0.1593D 00		0.3259D-02	
POSITION E		DATE	DATE	0.733 SAMPLE #		DOSE RATE:	EV/GM-MIN		SIGDR-EV		RAD/MIN		SIGDR-RAD		WAT/L		SIGDR-W	
POSITION E		DATE	DATE	0.733 SAMPLE #		DOSE RATE:	0.5813D 17		0.1186D 16		0.09312D 03		0.1900D 02		0.1589D 00		0.3252D-02	
POSITION E		DATE	DATE	0.733 SAMPLE #		DOSE RATE:	EV/GM-MIN		SIGDR-EV		RAD/MIN		SIGDR-RAD		WAT/L		SIGDR-W	
POSITION E		DATE	DATE	0.733 SAMPLE #		DOSE RATE:	0.5810D 17		0.1186D 16		0.09308D 03		0.1900D 02		0.1589D 00		0.3250D-02	
POSITION E		DATE	DATE	0.733 SAMPLE #		DOSE RATE:	EV/GM-MIN		SIGDR-EV		RAD/MIN		SIGDR-RAD		WAT/L		SIGDR-W	
POSITION E		DATE	DATE	0.733 SAMPLE #		DOSE RATE:	0.5796D 17		0.1183D 16		0.09285D 03		0.1895D 02		0.1585D 00		0.3242D-02	
POSITION E		DATE	DATE	0.733 SAMPLE #		DOSE RATE:	EV/GM-MIN		SIGDR-EV		RAD/MIN		SIGDR-RAD		WAT/L		SIGDR-W	
POSITION E		DATE	DATE	0.733 SAMPLE #		DOSE RATE:	0.5774D 17		0.1182D 16		0.09282D 03		0.1894D 02		0.1584D 00		0.3241D-02	
POSITION E		DATE	DATE	0.733 SAMPLE #		DOSE RATE:	EV/GM-MIN		SIGDR-EV		RAD/MIN		SIGDR-RAD		WAT/L		SIGDR-W	
POSITION E		DATE	DATE	0.733 SAMPLE #		DOSE RATE:	0.5785D 17		0.1181D 16		0.09268D 03		0.1891D 02		0.1582D 00		0.3236D-02	
POSITION E		DATE	DATE	0.733 SAMPLE #		DOSE RATE:	EV/GM-MIN		SIGDR-EV		RAD/MIN		SIGDR-RAD		WAT/L		SIGDR-W	
POSITION E		DATE	DATE	0.733 SAMPLE #		DOSE RATE:	0.5769D 17		0.1177D 16		0.09241D 03		0.1886D 02		0.1577D 00		0.3227D-02	

POSITION F		DATE	DATE	ABSORBANCE		DOSE RATE:	EV/GM-MIN		SIGDR-EV		RAD/MIN		SIGDR-RAD		WAT/L		SIGDR-W	
POSITION F		DATE	DATE	0.734 SAMPLE #		DOSE RATE:	0.6517D 17		0.1345D 16		0.1044D 04		0.2155D 02		0.1782D 00		0.3683D-02	
POSITION F		DATE	DATE	0.734 SAMPLE #		DOSE RATE:	EV/GM-MIN		SIGDR-EV		RAD/MIN		SIGDR-RAD		WAT/L		SIGDR-W	
POSITION F		DATE	DATE	0.734 SAMPLE #		DOSE RATE:	0.5918D 17		0.1222D 16		0.09401D 03		0.1957D 02		0.1618D 00		0.3349D-02	
POSITION F		DATE	DATE	0.734 SAMPLE #		DOSE RATE:	EV/GM-MIN		SIGDR-EV		RAD/MIN		SIGDR-RAD		WAT/L		SIGDR-W	
POSITION F		DATE	DATE	0.734 SAMPLE #		DOSE RATE:	0.5884D 17		0.1215D 16		0.09427D 03		0.1946D 02		0.1609D 00		0.3330D-02	
POSITION F		DATE	DATE	0.734 SAMPLE #		DOSE RATE:	EV/GM-MIN		SIGDR-EV		RAD/MIN		SIGDR-RAD		WAT/L		SIGDR-W	
POSITION F		DATE	DATE	0.734 SAMPLE #		DOSE RATE:	0.5834D 17		0.1204D 16		0.09346D 03		0.1929D 02		0.1595D 00		0.3301D-02	
POSITION F		DATE	DATE	0.734 SAMPLE #		DOSE RATE:	EV/GM-MIN		SIGDR-EV		RAD/MIN		SIGDR-RAD		WAT/L		SIGDR-W	
POSITION F		DATE	DATE	0.734 SAMPLE #		DOSE RATE:	0.5821D 17		0.1202D 16		0.09325D 03		0.1925D 02		0.1592D 00		0.3294D-02	
POSITION F		DATE	DATE	0.734 SAMPLE #		DOSE RATE:	EV/GM-MIN		SIGDR-EV		RAD/MIN		SIGDR-RAD		WAT/L		SIGDR-W	
POSITION F		DATE	DATE	0.734 SAMPLE #		DOSE RATE:	0.5804D 17		0.1198D 16		0.09298D 03		0.1920D 02		0.1587D 00		0.3284D-02	
POSITION F		DATE	DATE	0.734 SAMPLE #		DOSE RATE:	EV/GM-MIN		SIGDR-EV		RAD/MIN		SIGDR-RAD		WAT/L		SIGDR-W	
POSITION F		DATE	DATE	0.734 SAMPLE #		DOSE RATE:	0.5802D 17		0.1198D 16		0.09295D 03		0.1919D 02		0.1586D 00		0.3283D-02	
POSITION F		DATE	DATE	0.734 SAMPLE #		DOSE RATE:	EV/GM-MIN		SIGDR-EV		RAD/MIN		SIGDR-RAD		WAT/L		SIGDR-W	
POSITION F		DATE	DATE	0.734 SAMPLE #		DOSE RATE:	0.5794D 17		0.1196D 16		0.09282D 03		0.1916D 02		0.1584D 00		0.3278D-02	
POSITION F		DATE	DATE	0.734 SAMPLE #		DOSE RATE:	EV/GM-MIN		SIGDR-EV		RAD/MIN		SIGDR-RAD		WAT/L		SIGDR-W	
POSITION F		DATE	DATE	0.734 SAMPLE #		DOSE RATE:	0.5777D 17		0.1193D 16		0.09255D 03		0.1911D 02		0.1580D 00		0.3269D-02	

POSITION G		DATE	DATE	ABSORBANCE		DOSE RATE:	EV/GM-MIN		SIGDR-EV		RAD/MIN		SIGDR-RAD		WAT/L		SIGDR-W	
POSITION G		DATE	DATE	0.739 SAMPLE #		DOSE RATE:	0.6560D 17		0.1342D 16		0.1051D 04		0.2150D 02		0.1794D 00		0.3679D-02	
POSITION G		DATE	DATE	0.739 SAMPLE #		DOSE RATE:	EV/GM-MIN		SIGDR-EV		RAD/MIN		SIGDR-RAD		WAT/L		SIGDR-W	
POSITION G		DATE	DATE	0.739 SAMPLE #		DOSE RATE:	0.5928D 17		0.1213D 16		0.09496D 03		0.1943D 02		0.1621D 00		0.3324D-02	
POSITION G		DATE	DATE	0.739 SAMPLE #		DOSE RATE:	EV/GM-MIN		SIGDR-EV		RAD/MIN		SIGDR-RAD		WAT/L		SIGDR-W	
POSITION G		DATE	DATE	0.739 SAMPLE #		DOSE RATE:	0.5902D 17		0.1208D 16		0.09455D 03		0.1935D 02		0.1614D 00		0.3310D-02	
POSITION G		DATE	DATE	0.739 SAMPLE #		DOSE RATE:	EV/GM-MIN		SIGDR-EV		RAD/MIN		SIGDR-RAD		WAT/L		SIGDR-W	
POSITION G		DATE	DATE	0.739 SAMPLE #		DOSE RATE:	0.5856D 17		0.1206D 16		0.09445D 03		0.1932D 02		0.1612D 00		0.3307D-02	
POSITION G		DATE	DATE	0.739 SAMPLE #		DOSE RATE:	EV/GM-MIN		SIGDR-EV		RAD/MIN		SIGDR-RAD		WAT/L		SIGDR-W	
POSITION G		DATE	DATE	0.739 SAMPLE #		DOSE RATE:	0.5874D 17		0.1202D 16		0.09411D 03		0.1925D 02		0.1606D 00		0.3295D-02	
POSITION G		DATE	DATE	0.739 SAMPLE #		DOSE RATE:	EV/GM-MIN		SIGDR-EV		RAD/MIN		SIGDR-RAD		WAT/L		SIGDR-W	

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POSITION	G	DATE	307.C	DCSE RATE:	0.5872D 17	0.1201D 16	0.94C7D 03	0.1925D 02	0.1606D 00	0.3293C-02
POSITION G	314.0	DATE	0.0	DCSE RATE: <td>0.5872D 17</td> <td>0.1198D 16</td> <td>0.9384C 03</td> <td>0.1920D 02</td> <td>0.1601D 00</td> <td>0.3285C-02</td>	0.5872D 17	0.1198D 16	0.9384C 03	0.1920D 02	0.1601D 00	0.3285C-02
POSITION G	318.C	DATE	0.0	DCSE RATE: <td>0.5849D 17</td> <td>0.1197D 16</td> <td>0.9370D 03</td> <td>0.1917D 02</td> <td>0.1592D 00</td> <td>0.3280D-02</td>	0.5849D 17	0.1197D 16	0.9370D 03	0.1917D 02	0.1592D 00	0.3280D-02
POSITION G	321.0	DATE	0.0	DCSE RATE: <td>0.5843D 17</td> <td>0.1195D 16</td> <td>0.9360C 03</td> <td>0.1915D 02</td> <td>0.1597D 00</td> <td>0.3277D-02</td>	0.5843D 17	0.1195D 16	0.9360C 03	0.1915D 02	0.1597D 00	0.3277D-02
POSITION G	326.0	DATE	0.0	DCSE RATE: <td>0.5832D 17</td> <td>0.1193D 16</td> <td>0.9343D 03</td> <td>0.1912D 02</td> <td>0.1595D 00</td> <td>0.3271D-02</td>	0.5832D 17	0.1193D 16	0.9343D 03	0.1912D 02	0.1595D 00	0.3271D-02
POSITION G	334.C	DATE	0.0	DCSE RATE: <td>0.5815D 17</td> <td>0.1190D 16</td> <td>0.9316D 03</td> <td>0.1906D 02</td> <td>0.1590D 00</td> <td>0.3261D-02</td>	0.5815D 17	0.1190D 16	0.9316D 03	0.1906D 02	0.1590D 00	0.3261D-02

POSITION	H	DATE	318.C	DCSE RATE:	0.5848D 17 <th>0.1186D 16 <th>0.9368D 03 <th>0.1899D 02 <th>0.1599D 00 <th>0.3250D-02</th> </th></th></th></th>	0.1186D 16 <th>0.9368D 03 <th>0.1899D 02 <th>0.1599D 00 <th>0.3250D-02</th> </th></th></th>	0.9368D 03 <th>0.1899D 02 <th>0.1599D 00 <th>0.3250D-02</th> </th></th>	0.1899D 02 <th>0.1599D 00 <th>0.3250D-02</th> </th>	0.1599D 00 <th>0.3250D-02</th>	0.3250D-02
POSITION H	267.0	DATE	0.0	DCSE RATE: <td>0.5835D 17</td> <td>0.1185D 16</td> <td>0.9359D 03</td> <td>0.1898D 02</td> <td>0.1598D 00</td> <td>0.3249D-02</td>	0.5835D 17	0.1185D 16	0.9359D 03	0.1898D 02	0.1598D 00	0.3249D-02
POSITION H	277.C	DATE	0.0	DCSE RATE: <td>0.5820D 17</td> <td>0.1184D 16</td> <td>0.9349D 03</td> <td>0.1897D 02</td> <td>0.1597D 00</td> <td>0.3248D-02</td>	0.5820D 17	0.1184D 16	0.9349D 03	0.1897D 02	0.1597D 00	0.3248D-02
POSITION H	281.0	DATE	0.0	DCSE RATE: <td>0.5805D 17</td> <td>0.1183D 16</td> <td>0.9339D 03</td> <td>0.1896D 02</td> <td>0.1596D 00</td> <td>0.3247D-02</td>	0.5805D 17	0.1183D 16	0.9339D 03	0.1896D 02	0.1596D 00	0.3247D-02
POSITION H	296.C	DATE	0.0	DCSE RATE: <td>0.5790D 17</td> <td>0.1182D 16</td> <td>0.9329D 03</td> <td>0.1895D 02</td> <td>0.1595D 00</td> <td>0.3246D-02</td>	0.5790D 17	0.1182D 16	0.9329D 03	0.1895D 02	0.1595D 00	0.3246D-02
POSITION H	306.0	DATE	0.0	DCSE RATE: <td>0.5775D 17</td> <td>0.1181D 16</td> <td>0.9319D 03</td> <td>0.1894D 02</td> <td>0.1594D 00</td> <td>0.3245D-02</td>	0.5775D 17	0.1181D 16	0.9319D 03	0.1894D 02	0.1594D 00	0.3245D-02
POSITION H	312.0	DATE	0.0	DCSE RATE: <td>0.5760D 17</td> <td>0.1180D 16</td> <td>0.9309D 03</td> <td>0.1893D 02</td> <td>0.1593D 00</td> <td>0.3244D-02</td>	0.5760D 17	0.1180D 16	0.9309D 03	0.1893D 02	0.1593D 00	0.3244D-02
POSITION H	318.C	DATE	0.0	DCSE RATE: <td>0.5745D 17</td> <td>0.1179D 16</td> <td>0.9299D 03</td> <td>0.1892D 02</td> <td>0.1592D 00</td> <td>0.3243D-02</td>	0.5745D 17	0.1179D 16	0.9299D 03	0.1892D 02	0.1592D 00	0.3243D-02

POSITION	I	DATE	295.C	DCSE RATE:	0.6098D 17 <th>0.1237D 16 <th>0.9765D 03 <th>0.1982D 02 <th>0.1667D 00 <th>0.3391C-02</th> </th></th></th></th>	0.1237D 16 <th>0.9765D 03 <th>0.1982D 02 <th>0.1667D 00 <th>0.3391C-02</th> </th></th></th>	0.9765D 03 <th>0.1982D 02 <th>0.1667D 00 <th>0.3391C-02</th> </th></th>	0.1982D 02 <th>0.1667D 00 <th>0.3391C-02</th> </th>	0.1667D 00 <th>0.3391C-02</th>	0.3391C-02
POSITION I	295.C	DATE	0.0	DCSE RATE: <td>0.6083D 17</td> <td>0.1236D 16</td> <td>0.9755D 03</td> <td>0.1981D 02</td> <td>0.1666D 00</td> <td>0.3390C-02</td>	0.6083D 17	0.1236D 16	0.9755D 03	0.1981D 02	0.1666D 00	0.3390C-02
POSITION I	295.C	DATE	0.0	DCSE RATE: <td>0.6068D 17</td> <td>0.1235D 16</td> <td>0.9745D 03</td> <td>0.1980D 02</td> <td>0.1665D 00</td> <td>0.3389C-02</td>	0.6068D 17	0.1235D 16	0.9745D 03	0.1980D 02	0.1665D 00	0.3389C-02
POSITION I	295.C	DATE	0.0	DCSE RATE: <td>0.6053D 17</td> <td>0.1234D 16</td> <td>0.9735D 03</td> <td>0.1979D 02</td> <td>0.1664D 00</td> <td>0.3388C-02</td>	0.6053D 17	0.1234D 16	0.9735D 03	0.1979D 02	0.1664D 00	0.3388C-02
POSITION I	295.C	DATE	0.0	DCSE RATE: <td>0.6038D 17</td> <td>0.1233D 16</td> <td>0.9725D 03</td> <td>0.1978D 02</td> <td>0.1663D 00</td> <td>0.3387C-02</td>	0.6038D 17	0.1233D 16	0.9725D 03	0.1978D 02	0.1663D 00	0.3387C-02
POSITION I	295.C	DATE	0.0	DCSE RATE: <td>0.6023D 17</td> <td>0.1232D 16</td> <td>0.9715D 03</td> <td>0.1977D 02</td> <td>0.1662D 00</td> <td>0.3386C-02</td>	0.6023D 17	0.1232D 16	0.9715D 03	0.1977D 02	0.1662D 00	0.3386C-02
POSITION I	295.C	DATE	0.0	DCSE RATE: <td>0.6008D 17</td> <td>0.1231D 16</td> <td>0.9705D 03</td> <td>0.1976D 02</td> <td>0.1661D 00</td> <td>0.3385C-02</td>	0.6008D 17	0.1231D 16	0.9705D 03	0.1976D 02	0.1661D 00	0.3385C-02
POSITION I	295.C	DATE	0.0	DCSE RATE: <td>0.5993D 17</td> <td>0.1230D 16</td> <td>0.9695D 03</td> <td>0.1975D 02</td> <td>0.1660D 00</td> <td>0.3384C-02</td>	0.5993D 17	0.1230D 16	0.9695D 03	0.1975D 02	0.1660D 00	0.3384C-02

POSITION	J	DATE	295.C	DCSE RATE:	0.5513D 17 <th>0.1122D 16 <th>0.8832D 03 <th>0.1778D 02 <th>0.1507D 00 <th>0.3077D-02</th> </th></th></th></th>	0.1122D 16 <th>0.8832D 03 <th>0.1778D 02 <th>0.1507D 00 <th>0.3077D-02</th> </th></th></th>	0.8832D 03 <th>0.1778D 02 <th>0.1507D 00 <th>0.3077D-02</th> </th></th>	0.1778D 02 <th>0.1507D 00 <th>0.3077D-02</th> </th>	0.1507D 00 <th>0.3077D-02</th>	0.3077D-02
POSITION J	279.C	DATE	0.0	DCSE RATE: <td>0.5503D 17</td> <td>0.1121D 16</td> <td>0.8822D 03</td> <td>0.1777D 02</td> <td>0.1506D 00</td> <td>0.3076D-02</td>	0.5503D 17	0.1121D 16	0.8822D 03	0.1777D 02	0.1506D 00	0.3076D-02
POSITION J	281.0	DATE	0.0	DCSE RATE: <td>0.5488D 17</td> <td>0.1120D 16</td> <td>0.8812D 03</td> <td>0.1776D 02</td> <td>0.1505D 00</td> <td>0.3075D-02</td>	0.5488D 17	0.1120D 16	0.8812D 03	0.1776D 02	0.1505D 00	0.3075D-02
POSITION J	295.C	DATE	0.0	DCSE RATE: <td>0.5473D 17</td> <td>0.1119D 16</td> <td>0.8802D 03</td> <td>0.1775D 02</td> <td>0.1504D 00</td> <td>0.3074D-02</td>	0.5473D 17	0.1119D 16	0.8802D 03	0.1775D 02	0.1504D 00	0.3074D-02
POSITION J	295.C	DATE	0.0	DCSE RATE: <td>0.5458D 17</td> <td>0.1118D 16</td> <td>0.8792D 03</td> <td>0.1774D 02</td> <td>0.1503D 00</td> <td>0.3073D-02</td>	0.5458D 17	0.1118D 16	0.8792D 03	0.1774D 02	0.1503D 00	0.3073D-02
POSITION J	295.C	DATE	0.0	DCSE RATE: <td>0.5443D 17</td> <td>0.1117D 16</td> <td>0.8782D 03</td> <td>0.1773D 02</td> <td>0.1502D 00</td> <td>0.3072D-02</td>	0.5443D 17	0.1117D 16	0.8782D 03	0.1773D 02	0.1502D 00	0.3072D-02
POSITION J	295.C	DATE	0.0	DCSE RATE: <td>0.5428D 17</td> <td>0.1116D 16</td> <td>0.8772D 03</td> <td>0.1772D 02</td> <td>0.1501D 00</td> <td>0.3071D-02</td>	0.5428D 17	0.1116D 16	0.8772D 03	0.1772D 02	0.1501D 00	0.3071D-02
POSITION J	295.C	DATE	0.0	DCSE RATE: <td>0.5413D 17</td> <td>0.1115D 16</td> <td>0.8762D 03</td> <td>0.1771D 02</td> <td>0.1500D 00</td> <td>0.3070D-02</td>	0.5413D 17	0.1115D 16	0.8762D 03	0.1771D 02	0.1500D 00	0.3070D-02

POSITION	K	DATE	295.C	DCSE RATE:	0.6038D 17 <th>0.1224D 16 <th>0.9673D 03 <th>0.1960D 02 <th>0.1651D 00 <th>0.3354C-02</th> </th></th></th></th>	0.1224D 16 <th>0.9673D 03 <th>0.1960D 02 <th>0.1651D 00 <th>0.3354C-02</th> </th></th></th>	0.9673D 03 <th>0.1960D 02 <th>0.1651D 00 <th>0.3354C-02</th> </th></th>	0.1960D 02 <th>0.1651D 00 <th>0.3354C-02</th> </th>	0.1651D 00 <th>0.3354C-02</th>	0.3354C-02
POSITION K	267.0	DATE	0.0	DCSE RATE: <td>0.6023D 17</td> <td>0.1223D 16</td> <td>0.9663D 03</td> <td>0.1959D 02</td> <td>0.1650D 00</td> <td>0.3353C-02</td>	0.6023D 17	0.1223D 16	0.9663D 03	0.1959D 02	0.1650D 00	0.3353C-02
POSITION K	267.0	DATE	0.0	DCSE RATE: <td>0.6008D 17</td> <td>0.1222D 16</td> <td>0.9653D 03</td> <td>0.1958D 02</td> <td>0.1649D 00</td> <td>0.3352C-02</td>	0.6008D 17	0.1222D 16	0.9653D 03	0.1958D 02	0.1649D 00	0.3352C-02
POSITION K	295.C	DATE	0.0	DCSE RATE: <td>0.5993D 17</td> <td>0.1221D 16</td> <td>0.9643D 03</td> <td>0.1957D 02</td> <td>0.1648D 00</td> <td>0.3351C-02</td>	0.5993D 17	0.1221D 16	0.9643D 03	0.1957D 02	0.1648D 00	0.3351C-02
POSITION K	295.C	DATE	0.0	DCSE RATE: <td>0.5978D 17</td> <td>0.1220D 16</td> <td>0.9633D 03</td> <td>0.1956D 02</td> <td>0.1647D 00</td> <td>0.3350C-02</td>	0.5978D 17	0.1220D 16	0.9633D 03	0.1956D 02	0.1647D 00	0.3350C-02
POSITION K	295.C	DATE	0.0	DCSE RATE: <td>0.5963D 17</td> <td>0.1219D 16</td> <td>0.9623D 03</td> <td>0.1955D 02</td> <td>0.1646D 00</td> <td>0.3349C-02</td>	0.5963D 17	0.1219D 16	0.9623D 03	0.1955D 02	0.1646D 00	0.3349C-02
POSITION K	295.C	DATE	0.0	DCSE RATE: <td>0.5948D 17</td> <td>0.1218D 16</td> <td>0.9613D 03</td> <td>0.1954D 02</td> <td>0.1645D 00</td> <td>0.3348C-02</td>	0.5948D 17	0.1218D 16	0.9613D 03	0.1954D 02	0.1645D 00	0.3348C-02
POSITION K	295.C	DATE	0.0	DCSE RATE: <td>0.5933D 17</td> <td>0.1217D 16</td> <td>0.9603D 03</td> <td>0.1953D 02</td> <td>0.1644D 00</td> <td>0.3347C-02</td>	0.5933D 17	0.1217D 16	0.9603D 03	0.1953D 02	0.1644D 00	0.3347C-02

POSITION	L	DATE	295.C	DCSE RATE:	0.6064D 17 <th>0.1221D 16 <th>0.9714D 03 <th>0.1957D 02 <th>0.1658D 00 <th>0.3348D-02</th> </th></th></th></th>	0.1221D 16 <th>0.9714D 03 <th>0.1957D 02 <th>0.1658D 00 <th>0.3348D-02</th> </th></th></th>	0.9714D 03 <th>0.1957D 02 <th>0.1658D 00 <th>0.3348D-02</th> </th></th>	0.1957D 02 <th>0.1658D 00 <th>0.3348D-02</th> </th>	0.1658D 00 <th>0.3348D-02</th>	0.3348D-02
POSITION L	295.C	DATE	0.0	DCSE RATE: <td>0.6049D 17</td> <td>0.1220D 16</td> <td>0.9704D 03</td> <td>0.1956D 02</td> <td>0.1657D 00</td> <td>0.3347D-02</td>	0.6049D 17	0.1220D 16	0.9704D 03	0.1956D 02	0.1657D 00	0.3347D-02
POSITION L	295.C	DATE	0.0	DCSE RATE: <td>0.6034D 17</td> <td>0.1219D 16</td> <td>0.9694D 03</td> <td>0.1955D 02</td> <td>0.1656D 00</td> <td>0.3346D-02</td>	0.6034D 17	0.1219D 16	0.9694D 03	0.1955D 02	0.1656D 00	0.3346D-02
POSITION L	295.C	DATE	0.0	DCSE RATE: <td>0.6019D 17</td> <td>0.1218D 16</td> <td>0.9684D 03</td> <td>0.1954D 02</td> <td>0.1655D 00</td> <td>0.3345D-02</td>	0.6019D 17	0.1218D 16	0.9684D 03	0.1954D 02	0.1655D 00	0.3345D-02
POSITION L	295.C	DATE	0.0	DCSE RATE: <td>0.6004D 17</td> <td>0.1217D 16</td> <td>0.9674D 03</td> <td>0.1953D 02</td> <td>0.1654D 00</td> <td>0.3344D-02</td>	0.6004D 17	0.1217D 16	0.9674D 03	0.1953D 02	0.1654D 00	0.3344D-02
POSITION L	295.C	DATE	0.0	DCSE RATE: <td>0.5989D 17</td> <td>0.1216D 16</td> <td>0.9664D 03</td> <td>0.1952D 02</td> <td>0.1653D 00</td> <td>0.3343D-02</td>	0.5989D 17	0.1216D 16	0.9664D 03	0.1952D 02	0.1653D 00	0.3343D-02
POSITION L	295.C	DATE	0.0	DCSE RATE: <td>0.5974D 17</td> <td>0.1215D 16</td> <td>0.9654D 03</td> <td>0.1951D 02</td> <td>0.1652D 00</td> <td>0.3342D-02</td>	0.5974D 17	0.1215D 16	0.9654D 03	0.1951D 02	0.1652D 00	0.3342D-02
POSITION L	295.C	DATE	0.0	DCSE RATE: <td>0.5959D 17</td> <td>0.1214D 16</td> <td>0.9644D 03</td> <td>0.1950D 02</td> <td>0.1651D 00</td> <td>0.3341D-02</td>	0.5959D 17	0.1214D 16	0.9644D 03	0.1950D 02	0.1651D 00	0.3341D-02

APPENDIX D

Saunders-Taylor Calibration Data (Done by O. Block, 5/21/71)

Table D-1

<u>Volume Number</u>	<u>Free Volume in ml at 25°*</u>
V ₁	76.5759 ± 0.0929
V ₂	8.5446 ± 0.0706
V ₃	0.8360 ± 0.0027
V ₄	0.2149 ± 0.0071
V ₅	0.1396 ± 0.0079
V ₆	0.0825 ± 0.00736

*Density of Hg was taken to be 13.5340 gm/cm³ at 25 °C.

APPENDIX E

Varian 90P3 Gas Chromatograph Calibration Data

The Varian 90P3 was calibrated by injecting known amounts of hydrogen, methane, oxygen, nitrogen, and carbon monoxide. The area under the resulting peaks was determined by cutting out the peak and weighing. The peak weight in grams as a function of micromoles of gas at a particular attenuation, was the data for a least squares fit program, fitting to the formula $y = ax$, y being peak weight, x being moles of gas, and a being slope. Below is a tabulation of a for hydrogen, methane, oxygen, nitrogen, and carbon monoxide.

Table E-1

<u>Gas</u>	<u>Attenuation</u>	<u>Slope, a (micromoles/gm)</u>
H ₂	x1	0.009684 ± 0.004174
CH ₄	x16	0.0578 ± 0.0003382
O ₂	x16	0.02915 ± 0.000446
N ₂	x32	0.01472 ± 0.001832
CO	x16	0.002779 ± 0.0004515

THE IRRADIATION EFFECTS ON A MODEL SOLVENT EXTRACTION
SYSTEM OF TRIBUTYL PHOSPHATE-DODECANE

by

JOE PASCHAL HOLLAND

B.S., Kansas State University, 1974

AN ABSTRACT OF A MASTER'S THESIS

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the requirements for the degree

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ABSTRACT

The radiation effects on the system of tributyl phosphate (TBP) and dodecane, which is the solvent extraction system used for the Purex process, was studied.

TBP, dodecane, and mixtures of TBP and dodecane were irradiated at an absorbed dose rate of about 5.0×10^{16} eV/g-min with absorbed doses of 6.1×10^{18} eV/g to 1.75×10^{20} eV/g.

The irradiated samples were analyzed for gaseous products noncondensable at 161 °K and acid products from TBP. The gaseous products observed were hydrogen and methane and the acid products were dibutyl phosphate (DBP) and monobutyl phosphate (MBP). The average yield of hydrogen from dodecane was, $G(H_2) = 6.71$, $G(H_2)$ from TBP was 2.02, and $G(\text{Acid})$ from TBP was 3.71.

No significant dose dependence was observed from TBP or dodecane. Deviation from mixture law was observed for the yields of hydrogen and acid products for the mixtures of TBP and dodecane. Sensitized decomposition of TBP seems indicated.