

POSITRON LONG LIFETIME MEASUREMENTS IN
SOME ORGANIC LIQUIDS

By

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PREFACE

In spite of the large amount of theoretical and experimental work that has been done on the phenomenon of positron annihilation in different materials, there are still many suppositions and conjectures that suggest even more work on such a matter, and it was the main purpose of this paper to find values of positron lifetimes before annihilation in unreported organic compounds, hoping that they add a small contribution to the present knowledge on this subject.

The equipment used was another good reason to have chosen this field of work, since its cost is relatively low and also part of it is already available at the University of San Luis Potosi, Mexico, where work on this promising field of physics is intended to commence.

I want to express my sincere gratitude to the very valuable guidance and assistance of Dr. Betty D. Pollak, the financial support of INIC, the frequent help and advices of Miss Concepción San José, and of course the good will and patience of my wife María Isabel G. de Ledegma who typed the thesis, all of which specially contributed towards the completion of this paper.

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CHAPTER I.

INTRODUCTION

Dirac's Theory

The existence of positrons, the anti-particles of electrons, was predicted in 1928 by the relativistic quantum theory of P. A. M. Dirac (1) which was published in 1930 (2). Dirac assumed that the equations describing the electron were invariant under Lorentz transformations, and required a symmetry property between electronic states of positive and negative total energy. Dirac thus inferred that electrons could occupy a continuum of negative as well as a continuum of positive energy states. The positive energy states correspond to what we commonly know as the electron, and the negative energy states to what we identify with a positron.

Although this result was first taken as evidence for the failure of Dirac's electron theory, since the positron had not so far been found experimentally, four years later, C. D. Anderson (3), working in the California Institute of Technology detected the positron in a cloud chamber experiment.

Dirac assumed in his theory negative energy states all of which were filled up with electrons which did not produce an external electric field, and did not contribute to the total charge, momentum and energy of the system, such that when one of those negative levels

was empty, this would represent a moving positron, as opposed to the moving electron corresponding to the positive level.

Positron - Electron Annihilation.

The process of annihilation of an electron-positron pair is achieved when an electron falls into the empty state or hole representing the positron. Charge is conserved in this process but not the number of particles. The mass - energy conversion is always $\geq 2m_0c^2$ where m_0 is the electron mass.

Annihilation processes may be expressed symbolically as



where n is an integer that can have values of 1, or 2, or 3 depending upon the environment and/or the state of the electron-positron pair at the instant of annihilation, and theories are developed by choosing the best parameters that can explain adequately the annihilation process. Such a choice of parameters depends of course on the several factors involving annihilation, i. o. a) annihilation of a positron and electron when either one is a free particle, where following Dirac (1) the mean life is inversely proportional to the density of the medium for free annihilation and is expected to vary from element to element; b) annihilation of the positron and electron from the bound, unstable state called positronium which serves to explain the long lifetime and which theory will be developed in more detail in Chapter II; c) annihilation of positrons before they come to rest and where the collision cross section is the important parameter; d) annihilation of positrons when they are slowed down to thermal velocities, in

which the electron density and molecular structure play important parts.

A good approach to the study of positron annihilation in matter is that of lifetimes, which are relatively easy to measure experimentally. Such measurements give information about the interior of the sample where the positron annihilates, since the annihilation rate of the gamma rays produced by the process depends on the product of the wave functions of the annihilating pair. The gamma rays produced in the annihilation can pass through the sample without being appreciably attenuated or scattered thus reducing complications and errors in the experimental results.

Indubitably, the presence of a positron in the sample alters its electronic configuration. This is shown quantum mechanically by a distortion of the electron wave function and must be taken into account in order to interpret the annihilation data. However, when the positron captures an electron to form a bound system, the electron wave function is not altered as much, and more information can be obtained since now the positron lifetime depends not only on its environment but on the internal properties of the bound system as well.

In general, positrons entering a condensed medium have shown a complex lifetime spectrum (4). When the positron enters the sample under study, it will lose energy mainly by inelastic collision and then it will find itself in a region where both direct

annihilation and positronium formation cross sections are important and become competing factors. At still lower energies direct annihilation may occur (5). Some of the positrons are found to decay with an intensity I_1 and a short lifetime of about 10^{-10} seconds called the γ_1 component, while the rest decay with an intensity I_2 and a meanlife of about 10^{-9} seconds, which is called the anomalous long lifetime, or γ_2 component.

The intensity I_2 has been found to vary from 2% to 53% and according to some reports is mainly due to the decay of triplet positronium by "pickoff" annihilation with bound lattice electrons (6). It is recognized that certain correlations between I_2 and various properties of the molecules can be used as information about molecular structures.

This paper aims toward checking some reported values on long lifetimes in condensed (liquified) materials, and making new measurements that may contribute to reinforce the present theories on positron annihilation.

CHAPTER II.

THEORY.

Positronium Formation and Annihilation.

It is natural to assume that positrons and electrons attract each other because of their opposite electrical charges, so that before annihilation takes place they would momentarily behave as a quasi-stable system. Mohorovicic (7) as early as 1934 proposed such a bound state, Ruark (8) suggested the name positronium for the system, and it was Dutsch (9), (10) who verified experimentally its existence in 1951.

This positronium "atom" can be treated as an isolated two body system similar to the hydrogen atom, having potential energy of the form $-e^2/r$ where e is the electronic charge and r is the distance between the charges, kinetic energy equal to one half that of the hydrogen atom, with an energy in the n th quantum state of

$$W_n = - \frac{2\pi^2 \cdot m e^4}{n^2 h^2} \left[\frac{1}{2} \left(1 + \frac{m}{M_H} \right) \right]$$

where m is the electron mass and M_H the hydrogen atom mass, and a wave function in the ground state expressed as

$$\Psi_0 = \pi^{-1/2} (1/2a_0)^{3/2} \exp. (-r/2a_0)$$

where $a_0 = \hbar^2 / m e^2$

represents the Bohr radius for the ground level of the hydrogen atom.

Later work by Ball and Graham (4) reaffirmed the theory of positronium, which is said to be in the singlet state (1S) when the positron and electron are bound with their spins antiparallel, or in the triplet state (3S) when their spins are parallel. The former is called the parapositronium and the latter the ortho-positronium.

Energetics of positronium formation are well described by the "Ore gap" mechanism, as explained by De Zafra and Joyner (11) in their experiment on the temperature effect on positron annihilation. Figure 1 shows how the Ore gap is directly determined by the ionization potential E_i and the lowest excitation potential E_o of the surrounding atoms as well as the positronium binding energy E_p .

Positronium formation by positrons above the E_o level is unlikely, due to the preference for inelastic collision, and impossible below the bottom of the Ore gap due to lack of sufficient energy for electron capture.

If it is assumed that positrons will be more or less evenly distributed energy-wise between zero and E_o after undergoing their last possible excitation (inelastic) collision during the slowing down process, then the ratio of the width of the Ore gap to the width from zero to E_o indicates that fraction of entering positrons which form positronium. In the case of metals, an Ore gap does not exist because of the large number of conduction electrons which lower the binding energy below 6.8 ev.

Once positronium is formed, information about the positrons lifetime is carried out to the observer by gamma rays which transport the

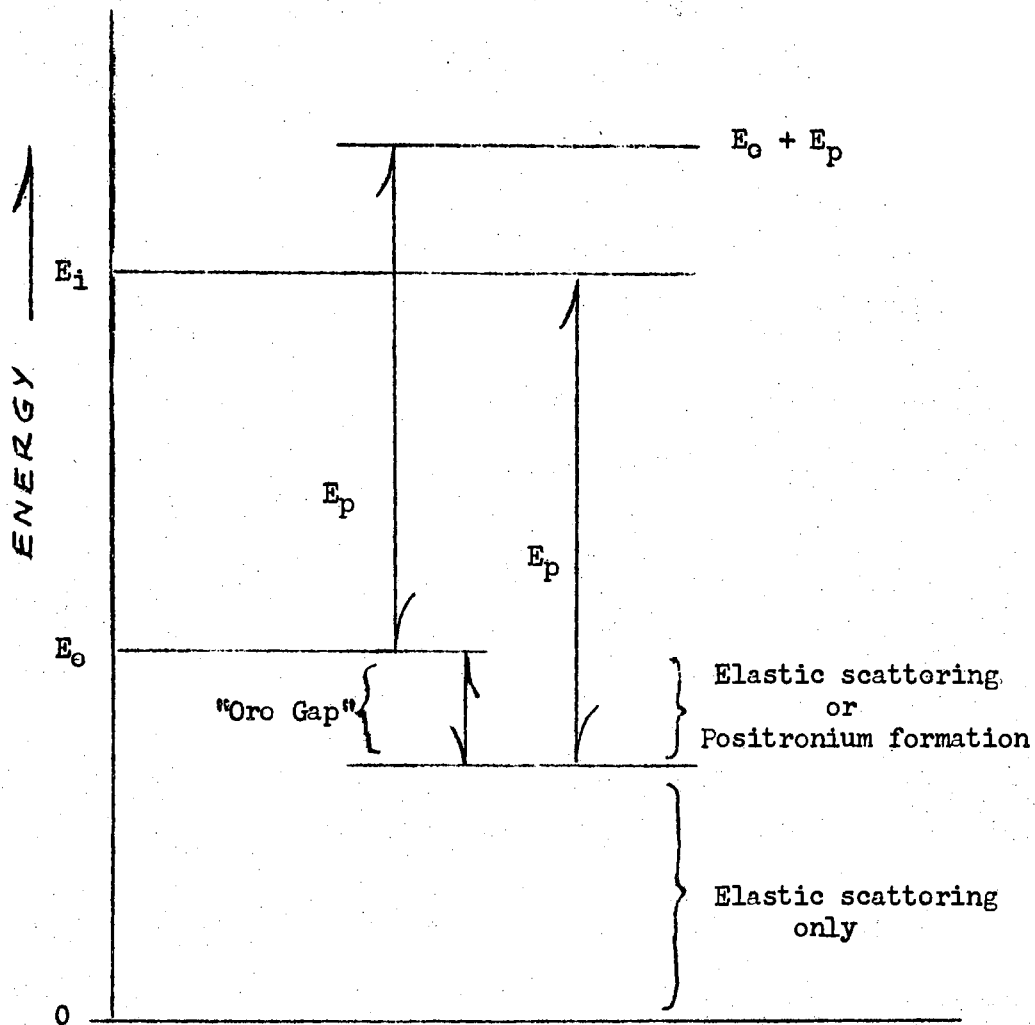


Figure 1. Energetics of Positronium Formation, (After De Zafra and Joyner (11)).

E_i = Ionization Potential of Surrounding Atoms

E_0 = Excitation Potential of Surrounding Atoms

E_p = Positronium Binding Energy

energy released in the positron-electron pair annihilation.

As predicted by Ore and Powell (12), the singlet state, having a total angular momentum of zero, decays with emission of two quanta and the triplet state with a total angular momentum of one, decays with the emission of three gamma rays. Discussions on this matter are, in general, restricted to low energies between the annihilating particles, since from annihilation-in-flight studies by Heitler (1), the probability that a positron annihilates while in flight is about 2% for 500 Kev positrons, while other experimenters (13) have shown that the momentum of the annihilating pair is about $mc/137$ corresponding to an energy of just a few electron volts.

The Long Lifetime Component.

Upon studying the time distribution of the annihilating radiation emerging from samples of condensed material following the absorption of positrons by the samples, Bell and Graham (4) reported that many of the studied materials showed a complex time decay curve with two distinct components of comparable intensity and different lifetimes, one of the order of 10^{-10} sec. and another of the order of 10^{-9} sec. These lifetime components are called the τ_1 and τ_2 components respectively. The τ_2 component is also referred to as the anomalous component because of its unexpected appearance in the results.

Bell and Graham (4) explained the τ_2 component found in experiments with a mechanism of ortho-parapositronium conversion (triplet state to singlet state) which depopulates the triplet state with a meanlife of 10^{-9} sec. to the rapid decaying singlet state, mainly

due to collisions with atoms of the sample material. Ferrell (5) theoretically worked on this explanation and complemented it with a process called "pickoff" annihilation, in which the decay of triplet positronium is due to the annihilation of its positron with a lattice electron in the neighboring atom or molecule.

The mechanism of "pickoff" annihilation was further observed by Green and Bell (6) while working on the variation in the amounts of positronium formed in liquids and amorphous solids.

In certain materials, a collision induces an actual exchange between the electron in the positronium atom and an electron of opposite spin from a neighboring atom that has two closely spaced energy levels of opposite spin, one of which is empty, and where the positronium atom must furnish an amount of energy equal to the difference in levels.

Lifetimes are also altered by materials whose energy level spacing is of the order of thermal energies, and which are known as "quenching" agents.

Another effect that hastens the destruction of positronium is the mixing of the 1S and 3S states by means of strong magnetic fields (14).

Bell and Graham (4) also found out that the long lifetime component decreased, in general, as the sample is cooled, although later work has shown that the temperature effect is in reality a density effect (11).

Annihilation Lifetime in Solids.

From measurements of Bell and Graham (4) it was settled that in metals, positrons have a single lifetime of the order of 10^{-10} seconds, but in amorphous solids such as plastics, fused quartz, etc., two lifetimes are observed: one of the order of 10^{-10} seconds, and the "anomalous" lifetime of the order of 10^{-9} seconds.

Energy losses for the positron entering the solid are largely due to inelastic collisions with the solid thermal lattice vibrations. As a result, the positrons attain thermal equilibrium with the lattice in a time short compared with its mean lifetime as stated by De Benedetti et al (15), and once thermalized, the positrons diffuse randomly through the solid without any further energy gain or loss on the average, and then are eventually annihilated. De Benedetti et al calculated a thermalization time of 3×10^{-10} sec. for gold, and concluded that positrons entering gold were thermalized before annihilation.

For metals as was mentioned at the beginning of this section, Bell and Graham (4) showed in 1953 that positron lifetimes are of the order of 10^{-10} seconds, which puts in evidence the calculations made by De Benedetti et al in 1950. However, five years later, Lee-Whiting (16) calculated a thermalization time of 3×10^{-12} seconds for metals, which is considerably shorter than the lifetime of positrons in the medium. Lee-Whiting took into consideration other factors that make the positron lose energy upon entering the solid, such as ordinary Coulomb scattering and free electron screening effects.

Positrons after being thermalized, form triplet positronium in some fraction of the cases. The triplet positronium is then converted to the singlet state, resulting in a lifetime shorter than that of the triplet state.

In the ionic crystals, which may in general be considered as elastic spheres, when positrons are thermalized they become practically "swarmed" by free electrons before positronium can be formed, as pointed out by Ferroll (5) who showed that the nonformation of positronium is essentially due to the fact that there is no room in the crystal for a positronium atom. The observed lifetimes are of the order of 10^{-10} seconds although their meanlife varies over a wider range from sample to sample, and when they belong to the Halido family, they show a linear dependence on the halogen ion radius as observed by B. D. Pollak (17), rather than the cubic dependence reported by Ferroll (5).

Annihilation Lifetimes in Liquids.

Since the experimental studies of positron interaction in solids and liquids by Borke and Horeford (18), it is known that the annihilation of positrons in liquids follows the same general pattern of the amorphous solids and plastics, with respect to the complex decay mode and the resulting intensities.

In liquids, there are at least two independent annihilation states with lifetimes of the order 10^{-10} seconds and 10^{-9} seconds. Reports of experiments on magnetic "quenching" (18), free radical interaction (19), and larger 3λ ratios (20) indicate the formation of positronium in these materials, which in turn explains the observation

of the anomalous lifetime component. This τ_2 component also varies strongly from sample to sample.

Berko and Zucholli (19) found that the long lifetime component could be altered by introducing a proper "quenching" agent in the liquid without altering the intensity, while Green and Bell (6) also found it possible to "quench" the intensity without appreciably changing the long lifetime observed in water.

Annihilation Lifetime in Gases.

Experimental verification of the existence of positronium was first reported by Deutsch (10), whose work (9) showed that annihilation of positrons in gases has also a complex spectrum, similar to that observed in amorphous solids and liquids, with the difference that in gases the density which affects the free electron cross section is greatly reduced, resulting in a longer lifetime for annihilation by direct collision.

Actually, three separate lifetime components are observed: a short lived component of the order of 10^{-10} seconds due to annihilation of parapositronium, a second lifetime of the order of magnitude predicted by the Dirac cross section and inversely dependent on gas pressure, and a third lifetime of the order of 10^{-7} seconds, independent of pressure and due to the annihilation of ortho-positronium.

CHAPTER III.

EXPERIMENTAL METHODS

The Electronic Circuit

The basic electronic circuitry used in most of the works referred to is essentially the same, (compare figures 2 and 3). It is designed to detect and measure a delayed coincidence between a 1.28 Mev nuclear gamma ray, also called the "prompt" γ -ray, and the 0.51 Mev annihilation radiation of the positron which was emitted almost simultaneously with the prompt γ -ray; these two events, the emission of the nuclear gamma radiation and the positron emission occur very conveniently in the isotope Na^{22} , which is thus the most suitable among the positron sources used in positron lifetime studies. This isotope is a long-lived positron emitter. It has been thoroughly studied (22), (23), and has a half-life of 2.6 years. See figure 4.

Most investigators have used a fast-slow coincidence circuit to record a "delayed triple coincidence". This delayed coincidence between the 1.28 Mev nuclear gamma ray and the 0.51 Mev annihilation quanta is set as a function of an artificial delay in the time channel.

According to Newton (24), who was among the first in developing the mathematical form of delayed coincidence resolution curves

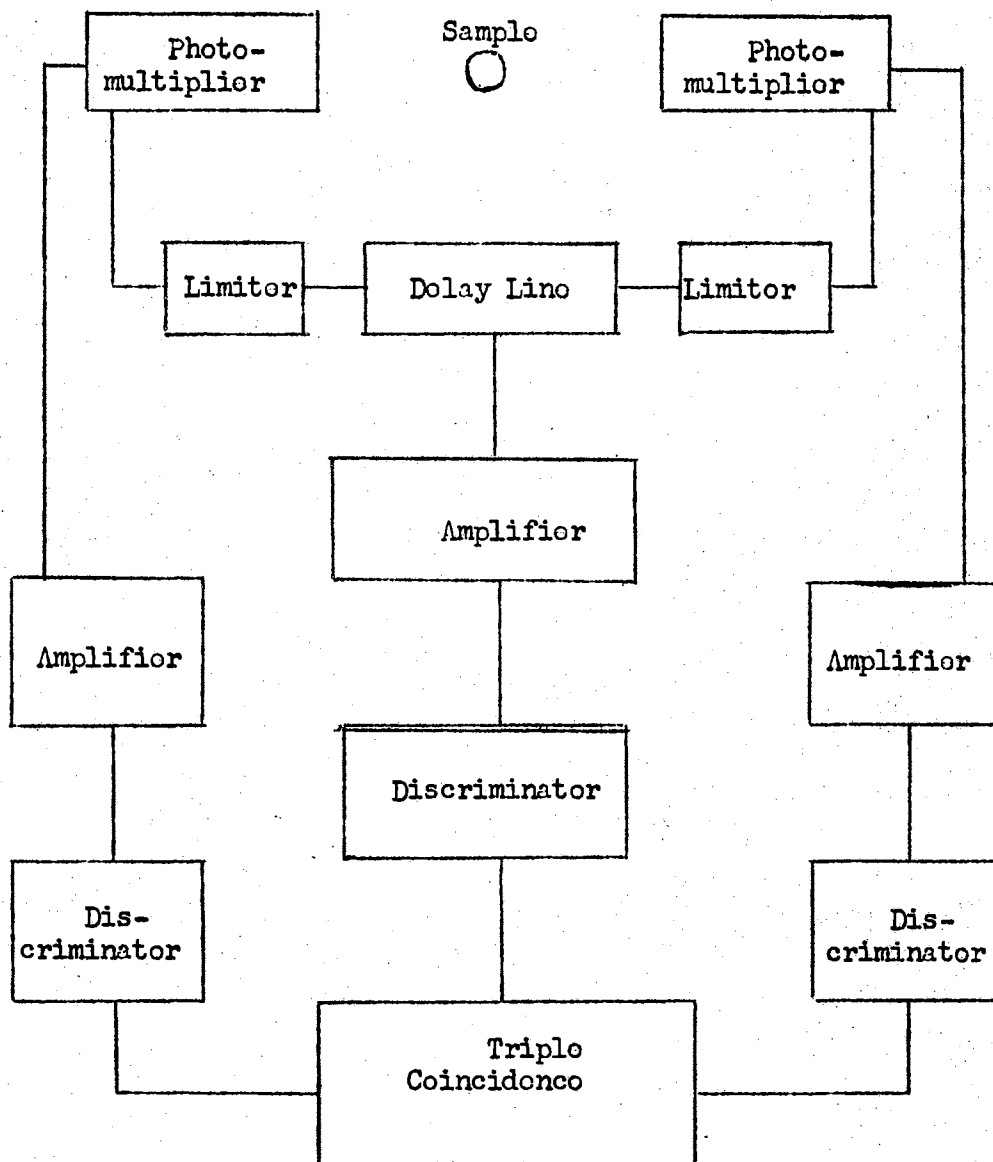


Figure 2. Block Diagram of Electronic Circuitry Used by Boll and Graham (4).

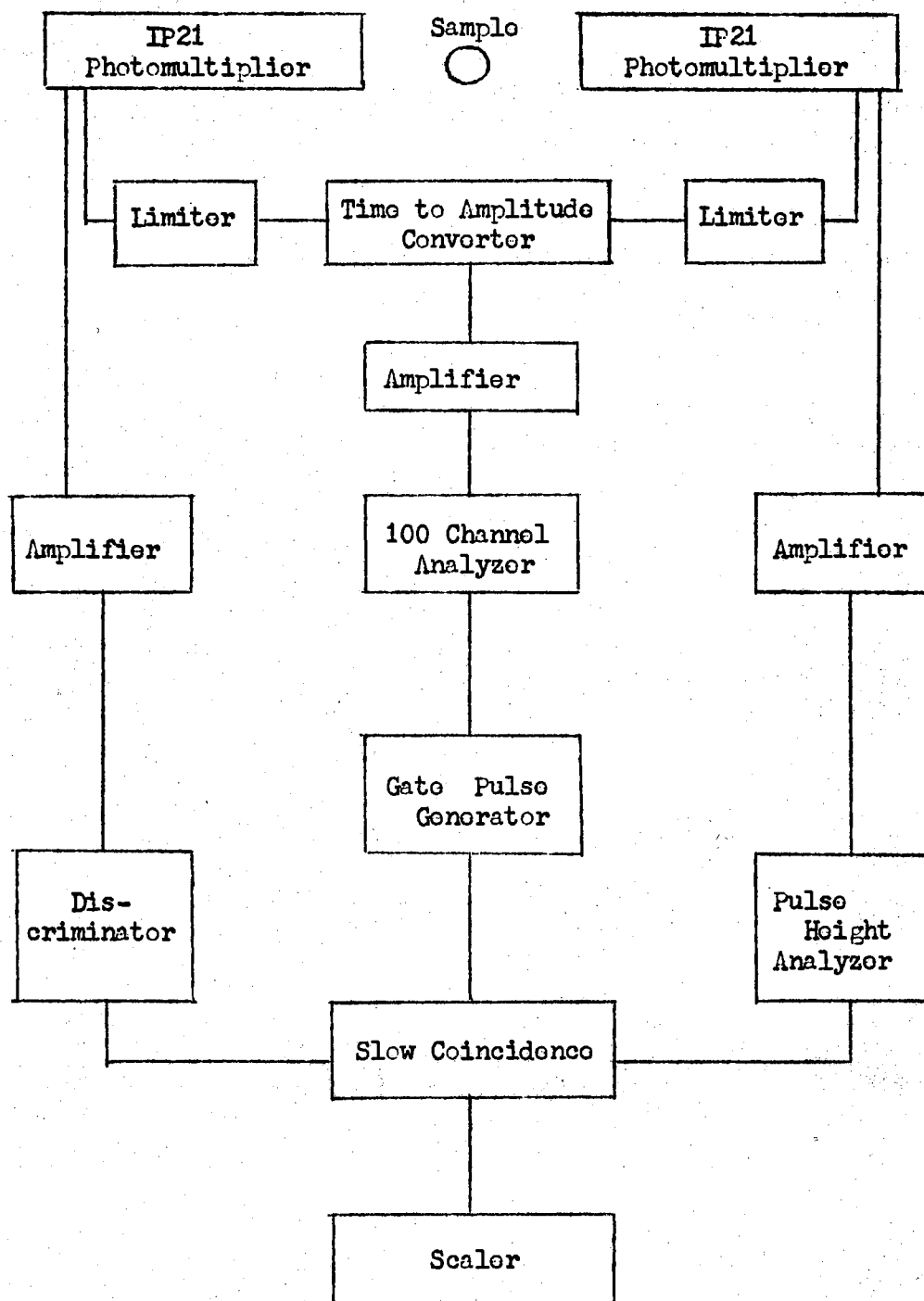


Figure 3. Block Diagram of Electronic Circuitry Used by Korr and Hogg (21).

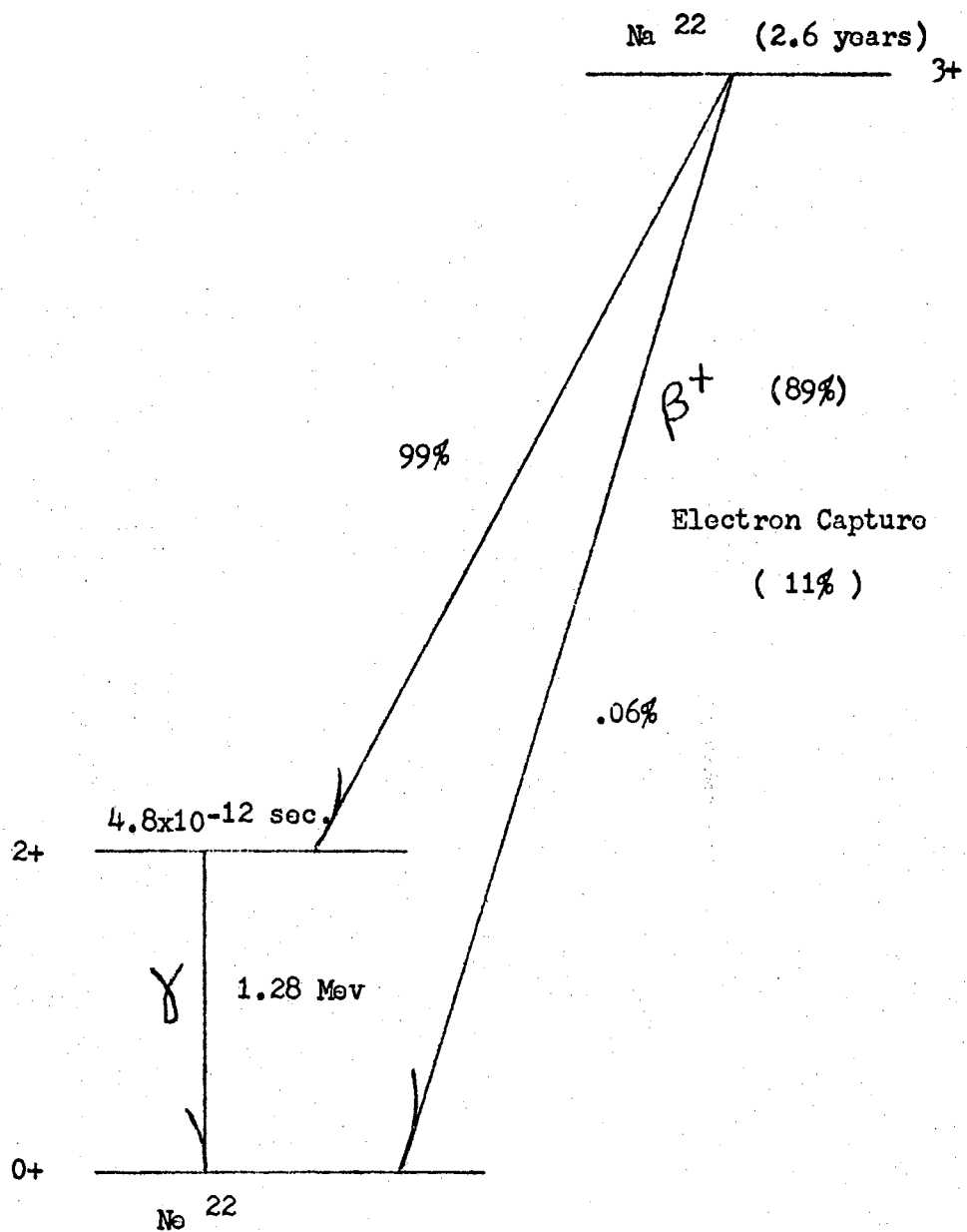


Figure 4. Decay Scheme of the Isotopo $\text{Na } 22$.

for the experimental set up considered in those studies, artificial delays are inserted in each side of the circuit so that the "prompt" coincidence resolution curve $P(x)$ occurs when there is no natural delay, i.e. when the 1.28 Mev radiation with a lifetime too short to be measured by the coincidence circuit occurs.

A block diagram of the electronic equipment used to obtain the data reported in the final chapter of this work is illustrated in figure 5. The center channels together with the time to amplitude convertor (T.A.C.) constitute the fast part of the circuitry and the side or "energy" channels including the triple coincidence unit, form the slow part of the circuitry.

The operating principles of the equipment are as follows:

The gamma rays or annihilation quanta produce photoelectrons in the detector which is a scintillation crystal assembled head-on with a photomultiplier tube that converts extremely weak photoelectronic signals into larger output pulses whose amplitude corresponds to the energy of the gamma incident upon the crystal. Thus the 1.28 Mev nuclear gamma pulses are roughly $2 \frac{1}{4}$ times the size of the 0.51 Mev annihilation quanta pulses.

For each gamma detected, the photomultiplier tube gives two output pulses, one out of the tenth dynode going to the slow circuitry through the side or energy channel, and the other out of the anode going to the fast circuitry through the center channel. The side channels have discriminators that block all pulses which are not necessary, such that a discriminator is set to pass only

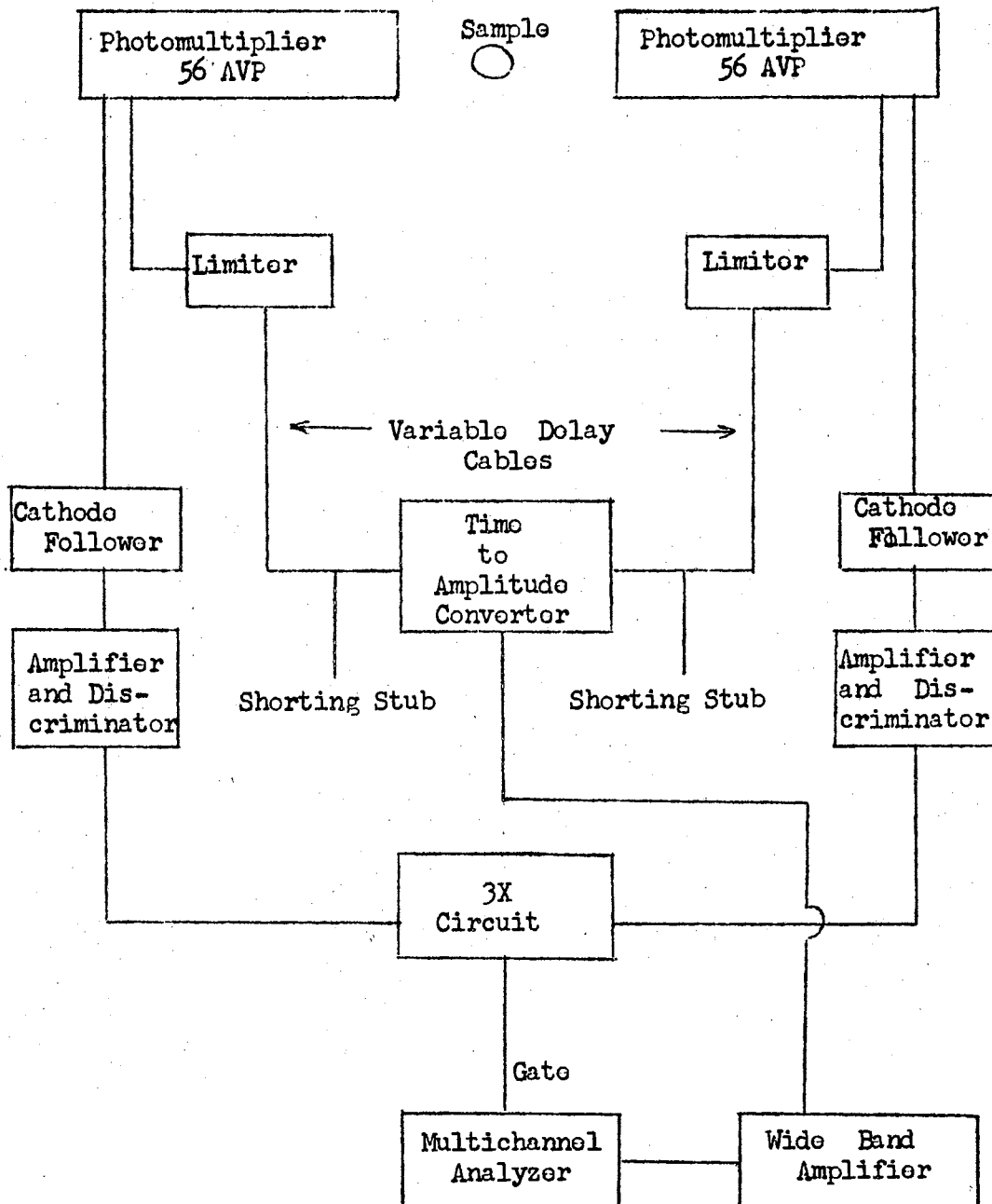


Figure 5. Block Diagram of Electronic Circuitry Used in This Thesis.

1.28 Mev gamma pulses, and the other is set to pass both 1.28 Mev and 0.51 Mev gamma pulses. This eliminates false coincidences between two 0.51 Mev pulses.

The two pulses then go into a coincidence circuit, with a resolving time of one microsecond. When pulses from each side channel arrive at this coincidence circuit within one microsecond of each other, a positive pulse approximately four microseconds wide appears at the output and it serves as a "gate" pulse for the multichannel analyzer by turning it on to record the T.A.C. pulses.

These T.A.C. pulses are produced when the signals out of each photomultiplier anode enter into its respective limiter circuit, which produces flat top pulses with short rise times and of the same height, (rise times are estimated to be less than one nanosecond and flat plateaus of about five microseconds) regardless of whether the pulse was due to the detection of a 1.28 or 0.51 Mev gamma ray.

The pulses are then transmitted by variable lengths of RG-7/U coaxial cables with a characteristic impedance of 97 ohms, to T-junctions where they split. The clipping junction consists of 183 centimeters of shorted RG-8/U coaxial cables with an impedance of 50 ohms. Part of each limiter pulse flows down the shorting stub to be reflected at the short circuited end and returns to the T-junction, reducing in this way the width of about five microseconds in the original pulse to a width of $2t$, where t is the time required for the pulse to travel the length of the shorted RG-8/U cable.

With 183 centimeters lengths of shorting stubs $2t$ is about 15×10^{-9} seconds, thus only the fastest rising portion of the limiter pulses are used as inputs for the T.A.C.

When the "fast" pulse from one limiter is produced within the time of activation of the other limiter, a superposition of the two signals occurs at the input of the T.A.C. and with this unit properly biased, only the overlapping portion of the two wave forms will be transmitted, resulting in an output pulse with its height being proportional to the overlap in time of the two limiter pulses. Then, in order to be counted, a pulse from the T.A.C. must represent a true coincidence, satisfying both the time and energy conditions of the circuit, such that the analyzer accepts output pulses from the T.A.C. only when they are allowed by the coincidence unit which "checks" the pulses for the correct energy in the side channels and the time separation in the center channel.

In the Multichannel Analyzer, the pulse height of an input pulse from the T.A.C. is converted to channel number by an analog-to-digital converter. The magnitude of the channel number is a linear function of the peak amplitude in the input pulses. A count is then recorded in the proper channel, and the time difference between the appearance of the 1.28 Mev and the 0.51 Mev quanta is obtained from the conversion factor of the time increment per channel number in the T.A.C. - Analyzer system.

Apart from some minor adjustments and appropriate setup for this experiment, the electronic circuitry and additional equipment

used, was basically the same as those previously tested by Loper (25) and Pigg (26) whose theses were of great use and value in the completion of this work.

The scintillators used were Nuclear Enterprises NE 102 with a decay constant of 3.5×10^{-9} seconds, specially designed for room temperature experiments and with a maximum emission peak at 4,200 Angstroms. The scintillators were attached head-on to the photomultipliers. Dow Corning silicon grease was used for better optical coupling.

The photomultipliers used were Amperex 56 AVP, with 14 dynodes, high gain of approximately 10^8 at 2000 volts, rapid rise time of output pulse (2×10^{-9} seconds) and short cathode time difference (3×10^{-10} seconds at 2000 volts). The tubes were positioned horizontally at 180 degrees to each other, properly protected, socketed into a fixed chassis that contained also the cathode follower and limiter circuits, and were usually operated at about 1850 volts supplied to the anode by a Hamner High Voltage Power Supply, Model N-4035.

The limiters used Amperex 5847 pentodes and the cathode follower used 6AK5 pentodes.

The energy discriminations were accomplished with Non-Overloading Linear Amplifiers, Baird Atomic, Model 215.

The two output pulses from these amplifiers were transmitted to a Coincidence-Anticoincidence Analyzer, Advance Radiation Engineering Corporation, Model 401.

The T.A.C. was a copy of the one designed by Simms (27), and the positive pulses out of it were inverted and further amplified by a Hewlett Packard Model 460 ER wide band amplifier. These negative pulses were then fed into the Multichannel Analyzer.

The Multichannel Analyzer employed was a Nuclear Data 512 channel Analyzer, Model ND 130, in which data were collected only in 128 channels. The data recorded by the analyzer were displayed on a Tektronix Oscilloscope, Model 503, and typed out by an IBM electronic typewriter.

Calibration of the Time Amplitude Converter.

For a given length of delay cable between limiters and T-junctions there is a determined overlap at the T.A.C. allowing an output pulse from it.

This effect relates delay cable length, which can be expressed more conveniently as delay time by knowing the pulse velocity in centimeters per nanosecond in the RG-7/U cable, to the peak channel number (read out from the Multichannel Analyzer) of a time distribution curve for a source of "prompt" gammas.

Thus, when the cable length is varied in one side of the limiter-to-T.A.C. circuit, and the resulting peak channel number of a time distribution curve of a known source is recorded, the plot of a straight line called the Apparatus or T.A.C. unit Calibration Curve is obtained. See figure 6.

The range of linearity of the apparatus is easily seen in this

graph, and the calibration factor k is readily obtained in units of seconds per peak channel number which is used in the calculation of lifetimes from the final data stored in the Multichannel Analyzer when a sample is studied.

The source of "prompt" gammas is one with a delay between emitted gammas which is short in comparison to the resolving time of the apparatus, where the resolving time is defined as the width of a "prompt" curve at half-maximum. See Figure 7.

Aluminium was used as the "prompt" gamma source; it has a single lifetime for positrons of 1.9×10^{-10} seconds as reported by Bell and Jorgenson (28).

Preparation of Samples.

Since the organic materials under study were all liquids, an "open" source technique was used to prepare the samples. This was done by direct deposit of about 5 microcuries of Na^{22} in the form of Na^{22} solution, inside the glass vial containing the liquid sample to be studied. The container was 1.5 cm. in diameter and 3 cm. high, which insured that less than 2% of the positrons emitted by the Na^{22} would annihilate in the walls of the container (29). This direct deposit of Na^{22} into the sample proved to be the best (30). To get the data needed to obtain the aluminum curves, the "sandwich" source technique was used. Na^{22} was deposited directly on household aluminum foil fixed tightly to an aluminum retaining cylinder especially designed to hold around the flours.

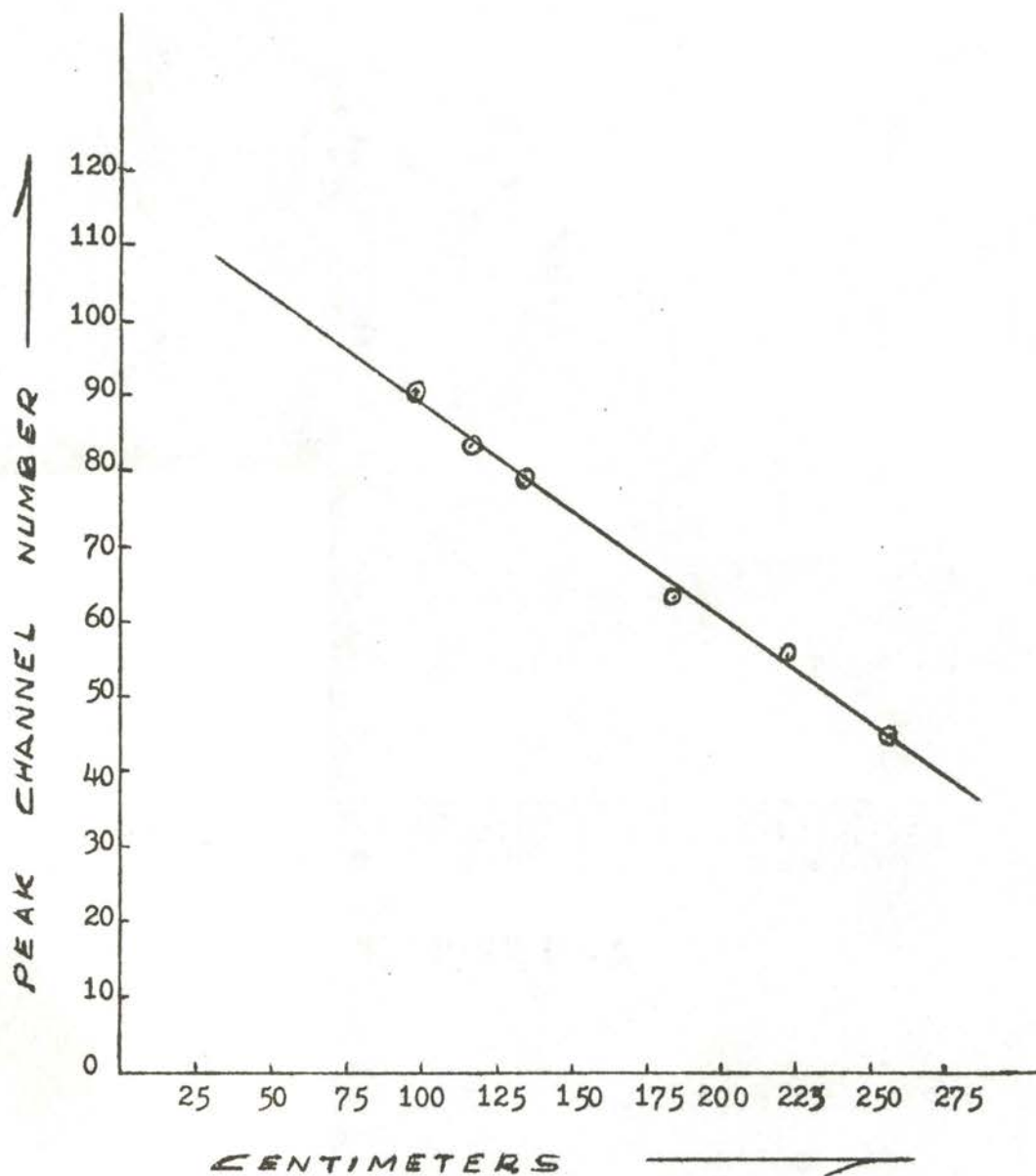


Figure 6. Apparatus or Time to Amplitude Converter Calibration Curve.

- This curve gave a calibration constant of

$$k = 1.46 \times 10^{-10} \text{ sec/channel}$$

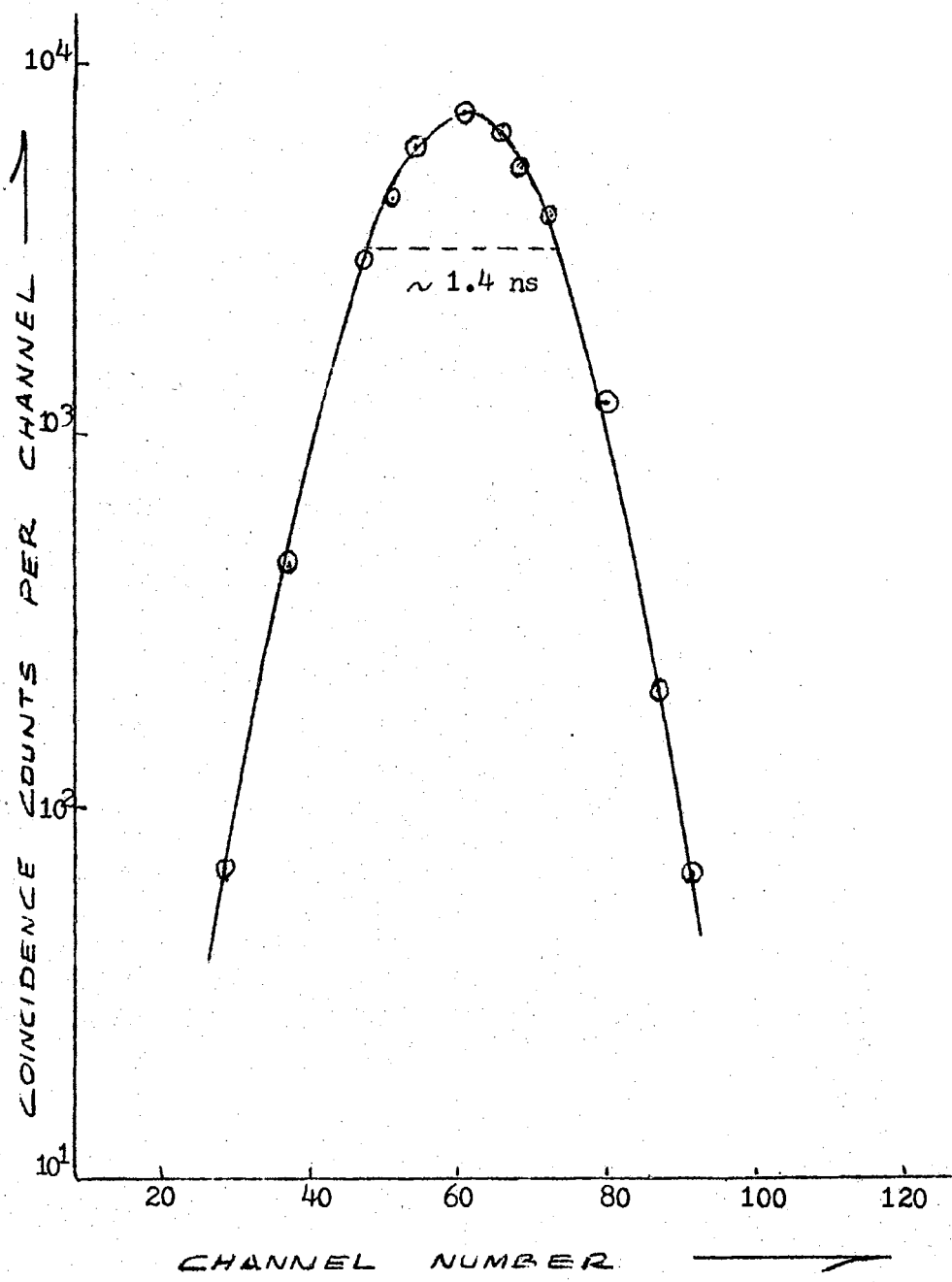


Figure 7. Typical Aluminum Time Resolution Curve.

- The width at half-maximum gives the resolution time.

It was then evaporated and "sandwiched" between two 1/10 inches thick aluminum discs. Household aluminum foil was chosen because it has been degreased.

CHAPTER IV.

RESULTS AND CONCLUSIONS.

Data Techniques.

Once the electronic equipment was calibrated and tested for performance, according to past experience of previous work with the same system (25), (26), collecting the data was completely automatized in the sense that only the "on" and "off" switch position had to be manually controlled to start or cut off the accumulation of data in the Multichannel Analyzer.

Data were allowed to accumulate until a well defined form of the curve (see Fig. 8) was observed on the screen of the Multichannel Analyzer oscilloscope, the time of accumulation varied from sample to sample due to slight differences in the source strengths deposited on each sample, a typical time being around 3 hours. Before or after each run an aluminum calibration curve (see Fig. 6) was taken in order to have a check on the stability of the apparatus, and to get the calibration constant needed to compute the positron long lifetime in the samples. A typical accumulation time for the points of this curve was $1/2$ hour per cable length.

In the curve displayed on the oscilloscope screen, the

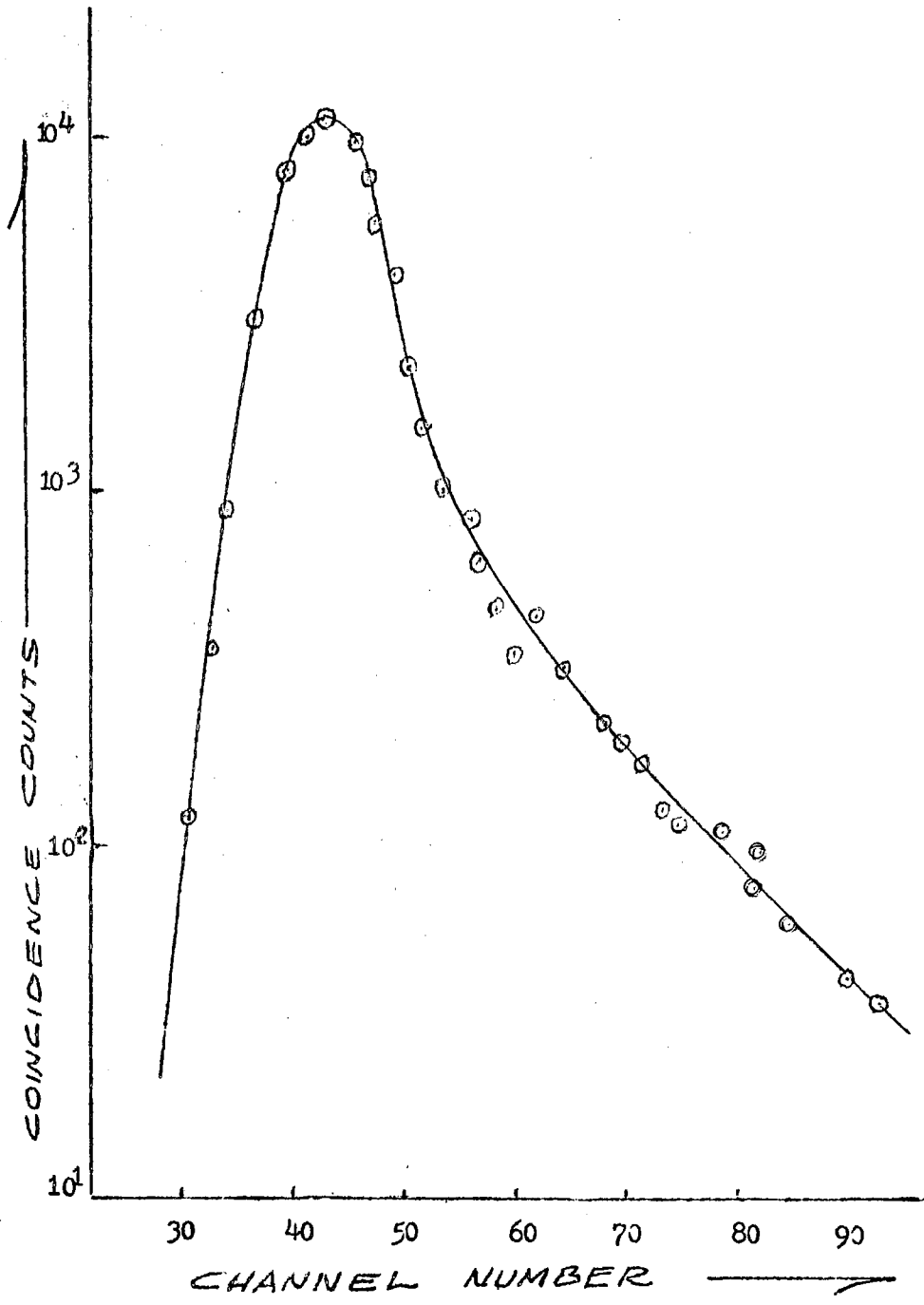


Figure 8. Experimental Time Distribution Curve for Cumono.

ordinates represented the number of coincidences between "prompt" gammas and "annihilation" gammas, and the abscissas represented the channel number where these coincidences occurred. The curve analyzed had a range of 128 channels although to determine the long lifetime only the information contained usually from channel 55 to 85 was used.

This information was printed out by the electric IBM typewriter attached to the electronic system. With the printed data, coincidence delay curves were plotted on semilogarithmic paper and the analysis was carried out as outlined in the next paragraph.

Analysis of Coincidence Delay Curves.

The method used for studying the long positron lifetime behavior based on the delayed coincidence electronic techniques, gives an experimental curve that is a composite of the apparatus electronic resolution curve, or "prompt" curve, and the delay coincidence curve due to the annihilating of positrons in the sample under investigation. This delayed coincidence curve may be due to the long lifetime τ_1 and/or the short lifetime τ_2 depending on the sample being studied.

The mathematical form of the delayed coincidence curves gives simple methods to analyze the experimental data. The

anomalous long lifetime component is easily recognized as being present in the semilogarithmic plotted data by a "tail" in the form of a straight line that breaks off an otherwise normal distribution curve along its side. In order to interpret the resulting experimental curve, it is necessary to define the apparatus resolution curve, or "prompt" curve, which is obtained by plotting coincidences of a source of simultaneous events as a function of channel number. Providing the drop off rate due to the coincidence apparatus is less than the mean lifetime of positrons in aluminum, an aluminum time distribution curve corrected for positron lifetime in aluminum, can serve as a "prompt" curve.

The mathematical expression (24) for the delayed coincidence curve $F(x)$ then is

$$F(x) = \int_{-\infty}^{+\infty} f(t) P(x-t) dt$$

where $P(x)$ = prompt curve (adjusted aluminum curve)

$f(t)$ = probability of a positron annihilating in an interval dt in a time t (time distribution to be measured)

x = artificial delay (i.o. channel number)

This expression is valid under the conditions (31) that (a) the quantities x and t are interchangeable, i.o. the inserted delay mechanism does not affect the pulse shape, and also that

(b) pulse shape distributions in both channels of coincidence circuit are the same for $F(x)$ and $P(x)$. Condition (a) is satisfied by using short delay cables of negligible attenuation. Condition (b) is met either by assuming that the same type and energy of radiation enters the detectors from both sources or by proper pulse shaping or pulse height selection, which is done by the use of limiters set just after the photomultiplier anode.

For the case of a single decay, which is the case exhibited by the positron lifetime decay, an exponential distribution in time is obtained such that

$$f(t) = \lambda e^{-\lambda t} \quad \text{for } t \geq 0$$

$$F(t) = 0 \quad \text{for } t < 0$$

where $\tau = \frac{1}{\lambda}$ = meanlife of positron

$$\text{then } F(x) = \lambda \int_0^{+\infty} e^{-\lambda t} P(x-t) dt$$

and differentiation with respect to x gives

$$\frac{dF(x)}{dx} = \lambda [P(x) - F(x)]$$

$$\frac{d \ln F(x)}{dx} = -\lambda \left[1 - \frac{P(x)}{F(x)} \right] \approx -\lambda \quad \text{when } F(x) \gg P(x)$$

So, the slope of the $F(x)$ curve on semi-log paper gives the reciprocal of the mean lifetime. The determination of τ_2 from the tail has the advantage of involving only one coincidence curve. There is therefore no need to be concerned with pulse

shape distributions, (condition (b)) but this restricts the practical use of the tail method (31) to $\tau_2 \gg$ resolution time of the apparatus.

The slope of the curve is best obtained by using the method of weighted least squares. In this method the essential problem consists of fitting a straight line to n points $(x_1, y_1), (x_2, y_2) \dots (x_i, y_i) \dots (x_n, y_n)$ for which the standard deviation of y , σ_y varies from point to point and x is known exactly. The data is plotted in semilog paper, so that

$$y_i = \ln N_i$$

$$x_i = \text{ith channel number}$$

$$N_i = \text{number of coincidence counts of ith point}$$

$$\text{Letting } N_1 \sigma_{y_1}^2 = N_2 \sigma_{y_2}^2 = \dots = N_n \sigma_{y_n}^2 = \sigma^2$$

so that the weights N_i , become the ratio of the variance at each point to some convenient variance σ^2 taken as reference, then it can be shown (see Appendix A) that the line among n points such that the sum of the squares of the vertical distances between each point and the line is a minimum, is satisfied by an equation of the form

$$y = \lambda x + b$$

$$\text{where: } \lambda = \frac{\sum_{i=1}^n N_i (x_i - \bar{x})(y_i - \bar{y})}{\sum_{i=1}^n N_i (x_i - \bar{x})^2}$$

$$b = \frac{\bar{y} \sum_{i=1}^n N_i x_i^2 - \bar{x} \sum_{i=1}^n N_i x_i y_i}{\sum_{i=1}^n N_i (x_i - \bar{x})^2}$$

$$\bar{x} = \frac{\sum N_i x_i}{\sum N_i}$$

$$\bar{y} = \frac{\sum N_i y_i}{\sum N_i}$$

finally giving

$$\tau_2 = \left[\frac{1}{\lambda} (\text{channels}) \right] \left[k \left(\frac{10^{-9} \text{ sec.}}{\text{channels}} \right) \right]$$

where k is the calibration constant.

The variance of the mean lifetime, that is of $1/\lambda$ is given by

$$v_{\tau} = \frac{(1/\lambda)^4}{\sum_{i=1}^n N_i (x_i - \bar{x})^2}$$

These computations were programmed to an IBM computer (see Appendix B) which reduced greatly the manlabor. The square root of the variance gives the standard deviation of the reciprocal of the slope and when converted to nanoseconds by multiplying it by k , the calibration constant, gives the uncertainty in τ_2 .

When more than one data run is taken on a particular sample, each yielding a lifetime and uncertainty of $T_i \pm t_i$, the most probable values of lifetime and uncertainty (32) are obtained from

$$T = \frac{\sum_{i=1}^n \left(\frac{T_i}{t_i^2} \right)}{\sum_{i=1}^n \left(1/t_i^2 \right)}$$

$$\bar{t} = \sqrt{1 / \sum_{i=1}^n (1/t_i^2)}$$

Measured Long Lifetimes.

The complete list of the experimental long lifetime measured in the chosen organic materials is given in Table I. The data show the average of four "reference" samples and five "now" samples. The reference samples were used with the purpose to give reliability to the results computed for the "now" or unreported samples, and it is seen from Table II showing a comparison of experimental lifetime results versus published values, that the "reference" samples do fall within the 3-sigma limit of the reported lifetimes (33).

TABLE I
COMPLETE LIST OF EXPERIMENTAL LIFETIME RESULTS.

Sample		τ_2 (10^{-9} sec)
$(C_6H_5)CH_3$	Toluene	$1.8 \pm .1$
$CH_3(C_6H_4)CH_3$	m-xylene	$2.1 \pm .1$
$CH_3(C_6H_4)CH_3$	o-xylene	$2.2 \pm .1$
$CH_3(C_6H_4)CH_3$	p-xylene	$2.1 \pm .1$
$CH(C_6H_5)(CH_3)_2$	Cumene	$1.9 \pm .1$
$(C_6H_5)CH_2CH_2CH_2CH_3$	Butylbenzene	$1.9 \pm .1$
$(C_6H_5)CH_2CH(CH_3)_2$	Iso-butylbenzene	$1.9 \pm .1$
$(C_6H_5)CHCH_2(CH_3)_2$	Sec-butylbenzene	$2.0 \pm .1$
$(C_6H_5)C(CH_3)_3$	Tert-butylbenzene	$1.9 \pm .1$

Calibration curves were run occasionally between "rest" periods of accumulating data. This insured against unnoticed changes in the apparatus which would be reflected directly in the calibration constant k used for computing the lifetimes. All data were taken at room temperature, which was maintained at 75° F.

TABLE II.
COMPARISON OF EXPERIMENTAL LIFETIME RESULTS OF THIS WORK VERSUS
PUBLISHED VALUES

Compound	Reported τ_2 (10^{-9} sec)	Measured τ_2 (10^{-9} sec)
$(C_6H_5)CH_3$ Toluene	$1.9 \pm .1$	$1.8 \pm .1$
$CH_3(C_6H_4)CH_3$ m-xylene	$2.2 \pm .1$	$2.1 \pm .1$
$CH_3(C_6H_4)CH_3$ o-xylene	$2.0 \pm .1$	$2.2 \pm .1$
$CH_3(C_6H_4)CH_3$ p-xylene	$2.1 \pm .1$	$2.1 \pm .1$

Discussion of Results.

The samples chosen were expected to give a different positron lifetime which could be related to some molecular structural property. However, no significant changes were found in the experimental lifetime results of the five organic compounds (see Table III), indicating that the different arrangements and order of the radical in the common molecular benzene ring, do not influence the positron annihilation or positronium formation phenomena, from one sample to the other.

TABLE III.
EXPERIMENTAL LIFETIME RESULTS OF MATERIALS NOT FOUND
IN PUBLISHED WORKS.

Compound		$\tau_2 (10^{-9} \text{ sec})$
$\text{CH}(\text{C}_6\text{H}_5)(\text{CH}_3)_2$	Cumene	$1.9 \pm .1$
$(\text{C}_6\text{H}_5)\text{CH}_2\text{CH}_2\text{CH}_2\text{CH}_3$	Butylbenzene	$1.9 \pm .1$
$(\text{C}_6\text{H}_5)\text{CH}_2\text{CH}(\text{CH}_3)_2$	Iso-Butylbenzene	$1.9 \pm .1$
$(\text{C}_6\text{H}_5)\text{CH}(\text{CH}_2(\text{CH}_3)_2)$	Sec-Butylbenzene	$2.0 \pm .1$
$(\text{C}_6\text{H}_5)\text{C}(\text{CH}_3)_3$	Tert-Butylbenzene	$1.9 \pm .1$

There is however, a significant difference between such C_6H_5 series and the xylenes, which have two hydrogens substituted in the benzene ring, leaving a basic C_6H_4 molecular arrangement. See Table I. In that table it can also be seen how toluene having a C_6H_5 basic molecular structure, that is with only one hydrogen substituted, shows statistically the same lifetime for positrons as the 5 last samples seen in the same table. In fact, toluene is reported in literature as having a 1.9×10^{-9} sec. positron lifetime (see Table II).

Thus positron annihilation experiments are promising techniques in molecular structure investigations which with more theoretical and experimental refinements could be made very efficient in determining molecular properties.

Table III gives the separate results for the samples studied which are not found in literature, and it is the hope of the author they can be useful to researchers in this field.

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APPENDIX A.

The Least Squares Method.

On analyzing the plotted data a straight line is "curve fitted" to the tail points of $F(x)$, the delayed coincidence curve, in the region conditioned to $F(x) \gg P(x)$, where $P(x)$ is the prompt resolution curve.

For this region a statistical technique of regression analysis is used, namely the method of least squares.

Such a method has for its object the adjustment and comparison of observations. Adjustments to make measurements agree and comparisons to determine the relative precision of measurements.

This statistical technique (34) assumes the consideration of certain characteristic (η) which is related or depends on certain other characteristics (X_1, X_2, \dots, X_p) according to the relationship

$$\eta = \mathcal{F}(X_1, X_2, \dots, X_p | \theta_1, \theta_2, \dots, \theta_q)$$

In our case we only have one characteristic X , and

$$\eta = y = \text{natural logarithm of the coincidence counts}$$

$$X_1 = x = \text{channel number}$$

$$\theta_1, \theta_2 = \lambda, b = \text{function parameters to be determined}$$

$$\mathcal{F} = \text{a straight curve function}$$

so that

$$y = \lambda x + b$$

The parameters values cannot be determined without error because the observed values of the dependent variable seldom agree with the expected variable; this is expressed by the equation

$$Y = y + \epsilon$$

where ϵ is the error made in measuring η and it is usually assumed that ϵ is a random variable with mean 0 and variance σ_{ϵ}^2 .

Calling \hat{y}_i the estimator of Y_i the necessary differences $(Y_i - \hat{y}_i)$, are formed. The values of the function parameters estimators $\hat{\lambda}$ and \hat{b} are determined by minimizing the sum of squares of the deviations that is, by minimizing

$$S = \sum_{i=1}^n (Y_i - \hat{y}_i)^2$$

This is accomplished by differentiating S with respect to the estimators of λ and b separately and setting each partial derivative equal to zero. Symbolically

$$\frac{\partial S}{\partial \hat{\lambda}} = 0 \quad \text{and} \quad \frac{\partial S}{\partial \hat{b}} = 0$$

Solution of these equations gives:

$$\hat{\lambda} = \frac{\sum_{i=1}^n N_i (x_i - \bar{x})(y_i - \bar{y})}{\sum_{i=1}^n N_i (x_i - \bar{x})^2}$$

$$\hat{b} = \frac{\bar{y} \sum_{i=1}^n N_i x_i^2 - \bar{x} \sum_{i=1}^n N_i x_i y_i}{\sum_{i=1}^n N_i (x_i - \bar{x})^2}$$

where
$$\bar{x} = \frac{\sum N_i x_i}{\sum N_i}$$

and
$$\bar{y} = \frac{\sum N_i y_i}{\sum N_i}$$

where the N_i 's (number of coincidence counts of i th point) are the weight factors subjected to the condition

$$N_i \sigma_{y_i} = \sigma^2$$

APPENDIX B.

The IBM Computer Program.

The facilities of the 1620 IBM Computing Center installed at O. S. U. were used. The symbols used and the FORTRAN program are listed below.

Definitions:

Z(I)	N_i , number of experimental coincidence counts in ith channel
X(I)	x_i , ith channel number
Y(I)	y_i , natural logarithm of N_i
AX	\bar{x} , mean of x
AY	\bar{y} , mean of y
T	τ_2 , mean life
D	σ , square root of variance
S	λ , slope
OI	b, ordinate intersection
CK	k, calibration constant
IDENT	Identity number, or data run number
K;L	Interval for least squares fit.

The program is shown on next page.

```
DIMENSION Z(128),X(128),Y(128)
2  SUM 1=0.0
    SUM 2=0.0
    SUM 3=0.0
    SUM 4=0.0
    SUM 5=0.0
    SUM 6=0.0
    READ,IDENT,K,L,CK
    DO 11=K,L
11  READ,X(I),Z(I)
    DO 24 I=K,L
    Y(I)=LOG(Z(I))
    SUM 1=SUM 1+Z(I)
    SUM 2=SUM 2+Z(I)*X(I)
24  SUM 3=SUM 3+Z(I)*Y(I)
    AX=SUM 2/SUM 1
    AY=SUM 3/SUM 1
    DO 30 I=K,L
    SUM 4=SUM 4+Z(I)*(X(I)-AX)*(Y(I)-AY)
    SUM 5=SUM 5*Z(I)*((X(I)-AX)**2)
30  SUM 6=SUM 6+AY*Z(I)*(X(I)**2)-AX*Z(I)*X(I)*Y(I)
    S=SUM 4/SUM 5
    OI=SUM 6/SUM 5
    T=(1.0/S)*CK
    D=SQR(((1.0/S)**4)/SUM 5)*CK
    PRINT,IDENT,T,D,S,OI
    PAUSE
    GO TO 2
    END
```


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