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A FIVE-MILLISECOND GAMMA EMITTER FROM PROTONS ON TANTALUM

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(Part B of Thesis)

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ABSTRACT

A search for gamma emitters in the millisecond range of half life was conducted, using the 32-Mev Berkeley proton linear accelerator. Foil targets of eighteen common elements were bombarded, and the activity made in them was followed between beam pulses with a gated scintillation counter. The only activity found of measurable yield was from Ta, and it exhibited a half life of 5.5 ± 0.3 milliseconds. Using a gated nine-channel pulse-height analyzer with the NaI(Tl) crystal counter, the excitation curve of this activity was measured and its gamma spectrum deduced. Gammas of 0.35 Mev and 0.22 Mev are indicated by the pulse spectrum, and no β's are present commensurate with this half life. Comparison of the excitation curve for this activity with those of Ta^{180} (8 hour) and W^{179} (30 m) leads to the tentative assignment of this decay to W^{180m}. As far as can be determined, this is the first known isomer with half life in the millisecond range.

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INTRODUCTION

Nuclei of the same mass and charge with different radioactive properties are known as nuclear isomers. Since isomerism was first experimentally demonstrated by Hahn[1] in 1921, over a hundred examples of it have been found among the known radioactive nuclei and a sound theoretical explanation for isomerism has been developed. Briefly, the existence of two nuclei of the same Z and A is explained by hypothesizing an excited state of this nucleus, of measurable lifetime, in addition to the ground state. This excited, or metastable, state can decay independently with a different half life and different radiations. The original difficulty with this explanation was that one of these states usually decayed by emitting only gamma radiation, and the half lives calculated for simple dipole emission were several orders of magnitude too short to explain the results. Weizsacker[2] overcame this difficulty by showing that higher multipoles in the nucleus would exhibit longer half lives, and one had only to assume that the nucleus could not radiate as a lower order multipole. His results yielded only an order-of-magnitude answer for half life as a function of gamma energy and multipolarity, but the variation of half life with these parameters was so strong that the calculations were useful. With the aid of this picture and corrections to it for the internal conversion of gamma rays[3] in the nucleus, it has been possible to arrive at a semiempirical classification of nuclear isomers[5] according to changes in nuclear spin and parity involved in the transition.

As larger numbers of isomers became known, it became obvious that there was a complete lack of isomers with half lives of the order of one millisecond. Considering the possibility of obtaining such an isomer by means of a parent-daughter relationship from a known beta-decaying nucleus, and the experimental problems associated with such

a short half life, it was a moot question whether this absence of millisecond half lives was merely due to difficulty of detection. However, within the last five years, methods[4] of electronic detection have so improved that almost a dozen examples have been found of isomeric half lives that are less than 10^{-4} sec, yet the search which yielded these activities brought no millisecond activities to light. Goldhaber and Sunyar[5] suggested that the absence of any half lives of this magnitude might actually be equivalent to their improbability on an energy basis. On the basis of their semiempirical classification of isomers, they found that the only combinations of spin change and gamma energy that could correspond to millisecond half lives were E_γ about 50 kev for $\Delta I = 2$ and E_γ about 800 kev for $\Delta I = 3$. Since the variation of half life with energy is a power law, these possible energies fairly sharply define the half life and make the rarity of isomers with these decay energies simply equivalent to a rarity of millisecond half lives.

A way of searching for these activities that had not been extensively used until the last few years was made available by pulsed heavy-particle accelerators. This method consists of bombarding a target that is near a counter and gating the counter so that it can count only during the time between beam pulses. Providing the target did not get too active with longer-lived activities, one could detect by this means any activity whose half life was of the order of the time between beam pulses or much shorter than this. This technique was successfully used by Alvarez[6] to find and identify the twelve-millisecond N^{12}, which emits a very energetic beta, but had not been used to find gamma emitters, perhaps because only in the last few years have high-efficiency gamma counters been available. The advent of scintillation counters, with their large effective volume for gamma detection as well as very short dead times, made such experiments as this practical.

If, now, an activity is detected which exhibits a very short half life and yet emits only gamma radiation or low-energy electrons which could be conversion electrons or photoelectrons, this activity must almost surely be a nuclear isomer, since a K capture or beta emitter would have to be of very high energy indeed in order to have a half life in the millisecond region. For example, the 27-m sec B^{12} emits a β^- of 13.4 Mev, and the 12-m sec N^{12} emits a β^- of 16.6 Mev; a K capture

among even the heaviest of elements could hardly be expected to occur with energies of this magnitude without appreciable positron emission.

The Berkeley 32-Mev proton linear accelerator is a pulsed machine which delivers 300-μsec-long pulses fifteen times per second. This makes it almost ideal to use for the production of short-lived activities, and so a search was undertaken for gamma-ray emitters with half lives in the range between one millisecond and 50 milliseconds which could be made by 32-Mev protons incident on a number of different elements.

METHOD

The geometry used for bombardment of the various targets is shown in Fig. 1. The gamma-ray counter consisted of a NaI(Tl) crystal, 3/4 inch in diameter by 3/4 inch high, permanently packaged in a 1/32-inch-wall aluminum can with MgO powder around three sides of the crystal as a diffuse reflector and a glass window glued to one end of the crystal. This is a packaging scheme developed by Swank[7], Borkowski[8], and others which has given the best light collection from the crystal of any method tried. The window of this package was kept in optical contact with the photocathode of a DuMont 6292 photomultiplier tube by using Aroclor resin sealed in with Duco cement. The photomultiplier was protected from stray magnetic fields by a Mu Metal shield, and was plugged into a voltage-dividing network which gave equal voltages between dynodes. The total voltage used across the tube was generally about 800 volts, but this was varied from one run to the next, as calibration was always done against the gamma ray of 0.662 Mev energy from Cs^{137}. The output of the photomultiplier was fed intoto a preamplifier immediately near it, and this preamplifier output was fed into a conventional linear pulse amplifier through a coaxial cable. The gain of the entire system was adjusted to a value that put the pulse height corresponding to photoabsorption of the 0.662-Mev gamma from Cs^{137} at approximately 50 to 70 volts. The pulse spectrum observed due to a Cs^{137} source is shown dotted in Fig. 4, the strong peak being the photoabsorption line, which is clearly visible on an oscilloscope screen as a bright line in the pulse spectrum.

In order to be able to measure half lives by counting immediately after the beam pulse any activity made by the beam, an electronic gating unit was built which consisted of a series of five multivibrators. These multivibrators are arranged so that the fall of the output pulse of one triggers the next in tandem, and the first multivibrator is triggered by the master gating unit, which is variable in delay with respect to the beam pulse. Together with the master gate, this unit gives six gate pulses which (a) follow upon the heels of one another in time, (b) are adjusted so that the first of them appears about two milliseconds after the end of the beam pulse, and (c) are adjusted to be of the same

width in time. These gate pulses are fed separately to each of six scale-of-1024 scalers so that the only time a scaler can accept input pulses from the scintillation counter is within the duration of the gate. Thus the only time Scaler 1 can count is during Gate 1; the only time Scaler 2 can count is during Gate 2, etc. Figure 6 shows a block diagram of this part of the electronic arrangement. To insure that the scalers accept pulses only from the same part of the spectrum, their discriminators are set at the same pulse height and checked frequently during a run. This minimum pulse height is generally equivalent to about that of a 100-kev gamma ray.

Thus the detection of an activity consists of the following sequence: A beam pulse of 32-Mev protons hits the foil target, making the activity; the activity continues to decay during the time following the beam pulse; during this time, Scaler 1 counts for several milliseconds, then it stops counting and Scaler 2 counts for the same number of milliseconds, then Scaler 3 counts, and so on, so that each beam pulse causes a separate counting experiment, and the counting rates on the scalers form a decay curve for the activity after background is subtracted away. Background consists of two counting rates: the first is simply that due to the beam alone with no target in place; the second is due to the long-lived activities made in the target, and was measured by counting for the same length of time as an actual measurement but with no beam and beginning 30 seconds after the beam had been turned off. It was this background which was high for certain elements, making it impossible to say if any short-lived activities were made in them. The experiment is actually possible only because equal numbers of atoms of activity exhibit counting rates inversely proportional to their half lives.

Foils approximately 10^{21} atoms/cm^2 thick of Pb, Pt, W, Mg, Al, Ni, C, Cu, Zn, Mo, Ag, Cd, In, Sn, Ta, Bi, Be, and U were bombarded in the manner described, and the only activity observed that indicated a short-lived decay was from Ta as a target. The gate widths and delay were such that an abundant activity of half life from about 2 m sec to 25 m sec would probably have been detected. Thick targets of W, Ta, and Pb were obtained, and the ratio of activities of the Ta to the W target was observed as greater than 50:1; i.e., the W activity was approximately equal to the total background, as was that of Pb. This ratio was

obtained by monitoring the beam with an argon-filled ion chamber and running each target for the same amount of beam. The incident beam energy was varied with Al absorbers, and the yield and half life for twelve energies down to about 14 Mev were measured for the thick Ta target. This gave ten good measurements of the half life, the average of which is plotted in Fig. 2. The gates were 2.5 m sec wide for this measurement and began 5 m sec after the beam pulse ended. The total spread of these ten measurements of half life is less than 5 percent, and the average value is 5.5 ± 0.3 m sec for the half life. The indicated error is not based on the spread in the measurements but represents the extreme decay curves which can be drawn through the six points in Fig. 2.

In order to verify that the activity found was indeed an isomer, its gamma-ray spectrum was investigated and a thin-target excitation function was obtained. For this purpose a nine-channel pulse-height analyzer developed by Harry Bowman, Robert Thomas, and William Gantz[9] of the Counter Maintenance and Development Group was used. This analyzer consists of ten scale-of-1024 scalers modified so that nine of them are used as differential channels of variable voltage width, and the tenth as a surplus register. (Provision is made for the nine channels to be moved about in voltage over a spectrum, but this feature was not used.) The analyzer has a pulse-shaping circuit with a window amplifier to effectively narrow the channel widths so that drift in channel width due to various scaler discriminators is minimized. The pulse spectrum due to the 0.662-Mev gamma of Cs^{137}, as measured by this analyzer in calibrating for the run, is shown dotted in Fig. 4. A beam monitor devised by Sumner Kitchen[10], consisting of a DuMont 6292 photomultiplier looking at air ionized by the beam, was used to measure equal increments of integrated beam, since this type of monitor is linear with beam intensity whereas an ion chamber is not. The ion chamber is not linear because the highly focused beam of the linear accelerator "uses up" all the available atoms of gas in the ion chamber by ionizing them, so that further increases in beam intensity do not give equal increases in ion current.

With the pulse-height analyzer gated so that it only accepted pulses during the time interval from 7 m sec after the beam till 13 msec

after the beam, Ta foils 0.004 inch thick were bombarded so that the beam passed through the target at 45°, as shown in Fig. 1; foils were changed frequently to keep background of long-lived activities down. Use of a pulse-height analyzer actually made the measurements from thin targets possible, since most of the total background was due to either very small or very large pulses. The pulse spectrum measured is shown as the solid histogram of Fig. 4. It is the average spectrum of five runs at different proton energies, and Fig. 3 shows the separate spectra measured at three of these energies. It is obvious that the gamma-ray spectrum does not change with proton energy. Even though the total background due to both causes mentioned changed both in spectrum and in magnitude, averaging 20 to 30 percent, the net spectrum did not. That no beta rays were present in the activity was shown by the fact that the pulse spectrum from the NaI crystal consisted mostly of relatively low-energy pulses. If betas were present with this half life, they would have to be well above 5 Mev in energy, and betas this energetic traversing a crystal this large would have caused very large pulses. Also the pulse spectrum from the crystal, were it due to betas, would certainly have been changed in shape by the introduction of one inch of Be between target and crystal, and it was not. The effect of the Be was simply to smear the spectrum slightly and to attenuate it as much as would be estimated for gamma rays of this energy.

The excitation curve consists of the total net yield measured at seven different proton energies down to threshold, which is between 13 and 14 Mev. The beam energy was changed by Al absorbers and was calculated according to the curves of Aron et al.[11] for the middle of the Ta target. It should be pointed out that it was necessary to have a long air path to the target in order to keep activity from the Al absorbers from raising background, and since the beam was scattered somewhat by the absorbers, undoubtedly for the thicker absorbers some of the monitored beam never reached the target. This means that the measured excitation curve should be multiplied by some correction factor which monotonically increases for decrease in proton energy. The effect of this correction would be to make the true curve rise less rapidly from threshold than the measured one, to make the true peak at about 22 Mev more marked than the measured one, and

to make the threshold as obtained by extrapolation lower than if obtained from the measured curve. Since this correction would not markedly change any conclusions drawn from the observed curve, it has not been made, and the observed curve is shown in Fig. 5.

In an attempt to identify the 5.5-m sec activity, thin-target excitation curves were obtained for the 8-hour Ta^{180} and the 30-minute W^{179} from protons on Ta. Stacks of 0.002-inch Ta foils were bombarded with 31-Mev protons and the activities in each foil followed with the scintillation counter biased low enough to detect K x-rays from the decay. The composite decay curves were resolved for these two half lives, and exhibited no activities in the range of 2 minutes to 30 minutes, nor measurable amounts of activities longer than 8 hours. This means that the 5-min activity assigned to W^{179} and listed in Ref. 12 does not exist, since it was supposedly made by 20-Mev protons on Ta.

A range-energy curve for Ta was obtained by interpolating between those for Ag and Pb given in Aron et al.[11], e.g., by assuming that

$$\frac{R_{Ag}(E_1)}{R_{Pb}(E_1)} = K_1 \frac{(Z/A)_{Pb}}{(Z/A)_{Ag}}$$

and using this to get a K_2 also for E_2. For E_1 = 32 Mev, K_1 = 0.92; for E_2 = 13 Mev, K_2 = 0.88. Then, with K = 0.90, it was assumed that

$$\frac{R_{Ta}(E)}{R_{Pb}(E)} = K \frac{(Z/A)_{Pb}}{(Z/A)_{Ta}}$$

The resulting excitation curves for Ta^{180} and W^{179} are plotted in Fig. 5 on the same energy scale as the 5.5 m sec activity.

DISCUSSION OF RESULTS

In accordance with the properties of a nuclear isomer, the above experiments show the existence of an isomer which has a half life of 5.5 m sec and a threshold for formation from protons on Ta of about 13 Mev. That it is an isomer is shown by the fact that it emits no betas of energy appropriate to this short half life, and also that all the mass numbers to which it could possibly be assigned already either have activities of other half life assigned to them or are stable. Table I shows the Segrè Chart for this region of Z and A.

That this activity is either Ta^{180} or W^{179} is ruled out because its excitation curve is noticeably different from the curves of those activities, and in accord with the mechanism for formation of isomers, it would have to be the same to be isomeric with either of them; i.e., isomers result from the falling down into a metastable state of a residual nucleus of high excitation, and it is this state of which we are actually measuring the excitation curve. That the activity is Ta^{179} is ruled out because its threshold would have to be about 8 Mev higher than that of Ta^{180}, which already has too high a threshold. Two assignments which cannot be ruled out on the basis of threshold and shape of the excitation curve are W^{181} and W^{180}. These would be possible by $Ta^{181}(p,n)W^{181}$ and $Ta^{181}(p,2n)W^{180}$, respectively. The observed threshold corresponds well with the Coulomb barrier height $Z/A^{1/3}$(Mev) for protons penetrating the Ta nucleus, and the peaked shape of the excitation curve is what one might expect from a (p, xn) reaction. Of these two, $Ta^{181}(p,2n)W^{180}$ seems more likely because it is peaked where one would expect it to be relative to the measured $Ta^{181}(p,3n)W^{179}$ and to other known (p, 2n) reactions. All other nuclides except Ta^{181} itself can be ruled out on the basis that the energetic threshold plus the Coulomb barrier to protons within the compound nucleus add up to a total threshold that is too high to fit the curves obtained. By this means all such reactions as (p, 2pn) or (p, pxn) are ruled out. The assignment of the activity to W^{180} is also consistent with the fact that it was not observed from protons on W, since to make it from W^{182} would require a (p, p2n) reaction that is energetically of too high a threshold to see, and W^{180} (stable) is only 0.13 percent abundant, so the (p, p')

Hf Z = 72	Ta Z = 73	W Z = 74
Hf^{180} 5.5 hr γ's of 0.057 0.093 0.214 ↓ Stable 35.4%	Ta^{181} 22 μsec γ's of 0.345 0.136 0.481 ↓ Stable 100%	W^{182} Stable 26.3%
Hf^{179} 19 sec γ 0.15 ↓ Stable 13.8%	Ta^{180} 8 hr β^- 0.6 γ 0.09 x-rays	W^{181} 140 d γ 1.83 e^- 0.07 x-rays (Threshold ~ 13 Mev)
Hf^{178} Stable 27.1%	Ta^{179} 600d γ 0.7 e^- 0.01	W^{180} Stable 0.126% (Threshold ~ 13 Mev)
Hf^{177} Stable 18.4%	Ta^{178} 9 min β^+ 1.06 3% γ 1.5 3% e^- 0.08 50% x-rays	W^{179} 30 min no γ x-rays

TABLE I

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reaction would make little of it. Perhaps a further experiment could be done on separated or enriched W^{180} to verify the assignment.

Nuclear isomers of even Z and A are rather rare, there being five in the newest isotope table available[12]. These are all in this region of Z = 72, being Hf^{180m}, Os^{186m}, Pb^{202m}, and two of Pb^{204m}. They are unlikely simply on the basis of having both neutrons and protons all paired, but the assignment of the 5.5-m sec activity to an excited state of stable W^{180} is not particularly difficult to reconcile with this scarcity, since those which are known are scattered over this very region of Z and A.

From the relatively crude pulse-height spectrum of Fig. 4, it can only be inferred that there is a gamma ray of about 0.35 Mev in the decay of this activity, and very probably one of about 0.22 Mev corresponding to the lower peak. There could easily be lower-energy gammas present that were not counted. Goldhaber and Sunyar[5] predicted on the basis of their semiempirical classification of isomers that the only possibilities for an isomer to have a half life in the millisecond range would correspond either to a spin change of two for which $E_\gamma \simeq$ 50 kev, or spin change of three, for which $E_\gamma \simeq$ 800 kev. They also show a distribution in spin and parity for the first excited states of even-even nuclei, which overwhelmingly favors I = 2, +. On the basis of these facts the most comforting guess one can make about the 5.5-m sec transition is that it is $\Delta I = 2$, no change in parity, which would make it electric quadrupole. Then one would have to hypothesize a low-energy gamma in the spectrum, which was the half-life-determining transition.

CONCLUSIONS AND SUMMARY

From the bombardment of several common elements with 32-Mev protons, only one activity with half life between 2 m sec and 25 m sec was found. On the basis of thresholds for neighboring activities, and shapes of their excitation curves and that of the new activity, this activity from p + Ta has been tentatively assigned as a metastable state of stable W^{180}. No sure assignment can be made to the transition concerning spin and parity. As far as can be determined at this writing, this is the first* experimentally demonstrated isomer with half life in the millisecond range, its measured half life being 5.5 m sec.

*Work is now being done on an isomer of Nb^{90} by Earl K. Hyde et al. at UCRL, which indicates another activity of about 20-m sec half life.

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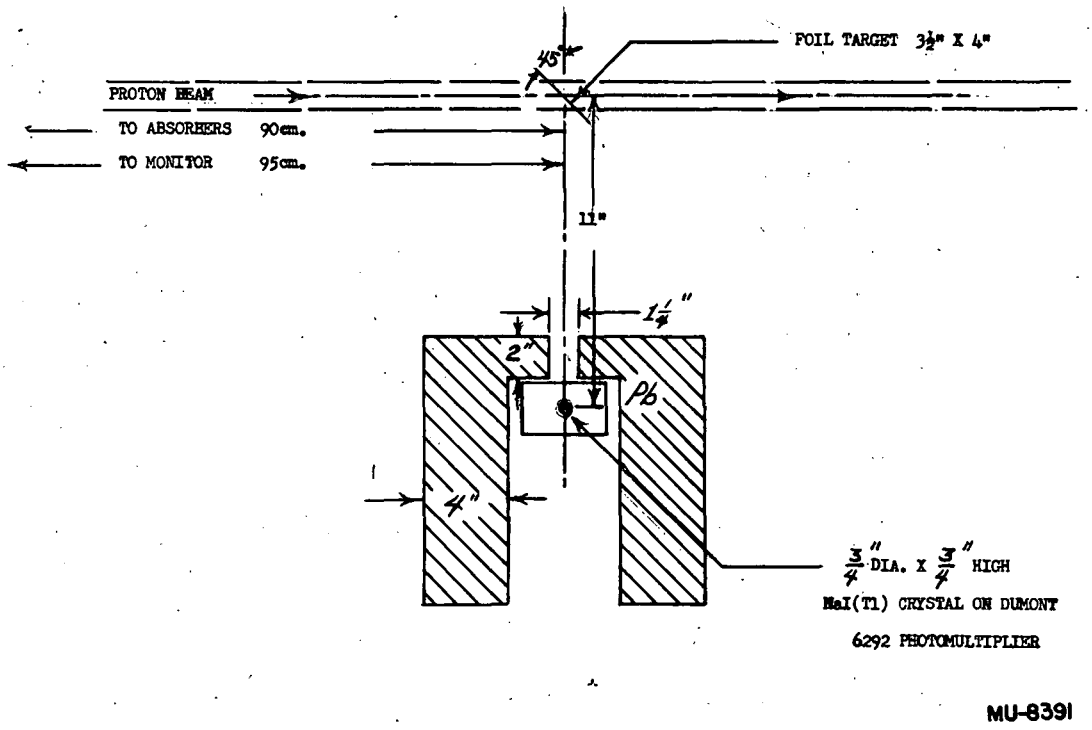

Fig. 1. Bombardment Arrangement.

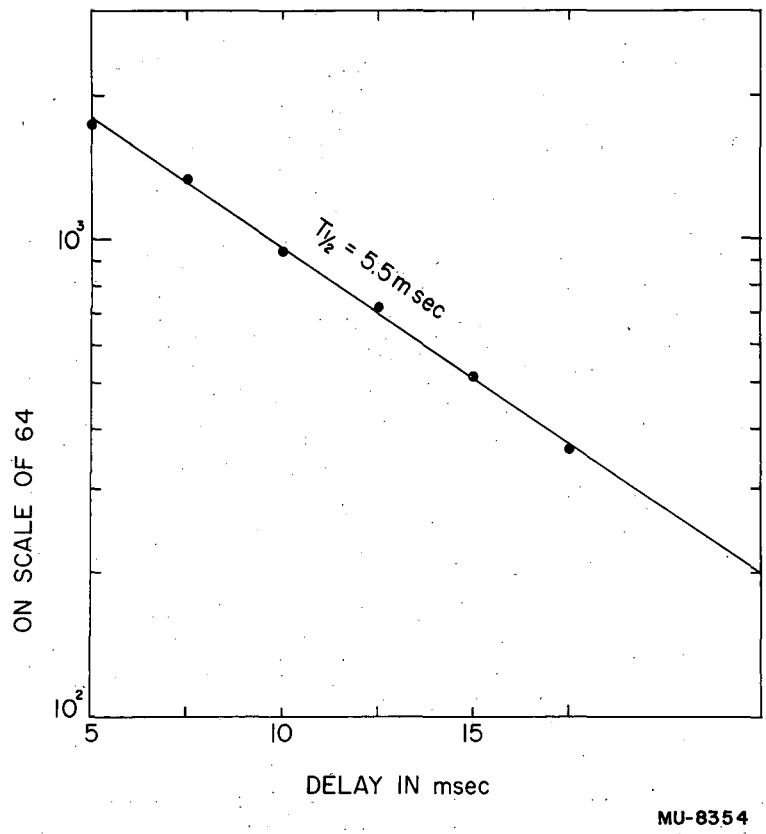

Fig. 2. Decay curve of 5 m sec activity.

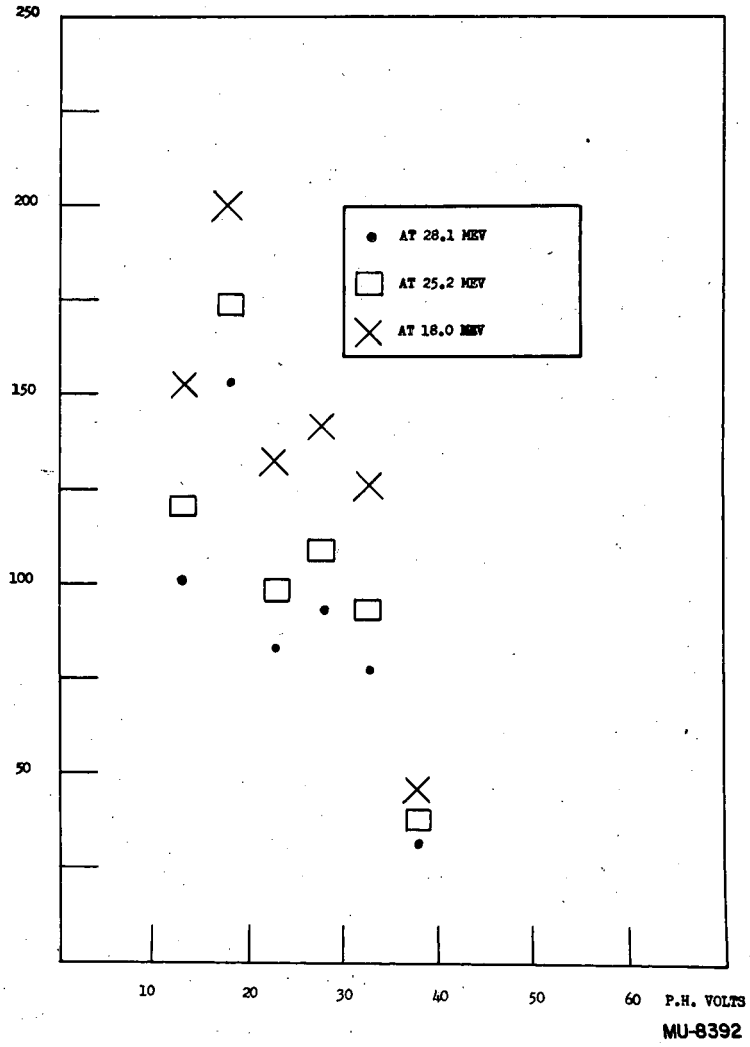

Fig. 3. Pulse height spectrum of activity for three proton energies.

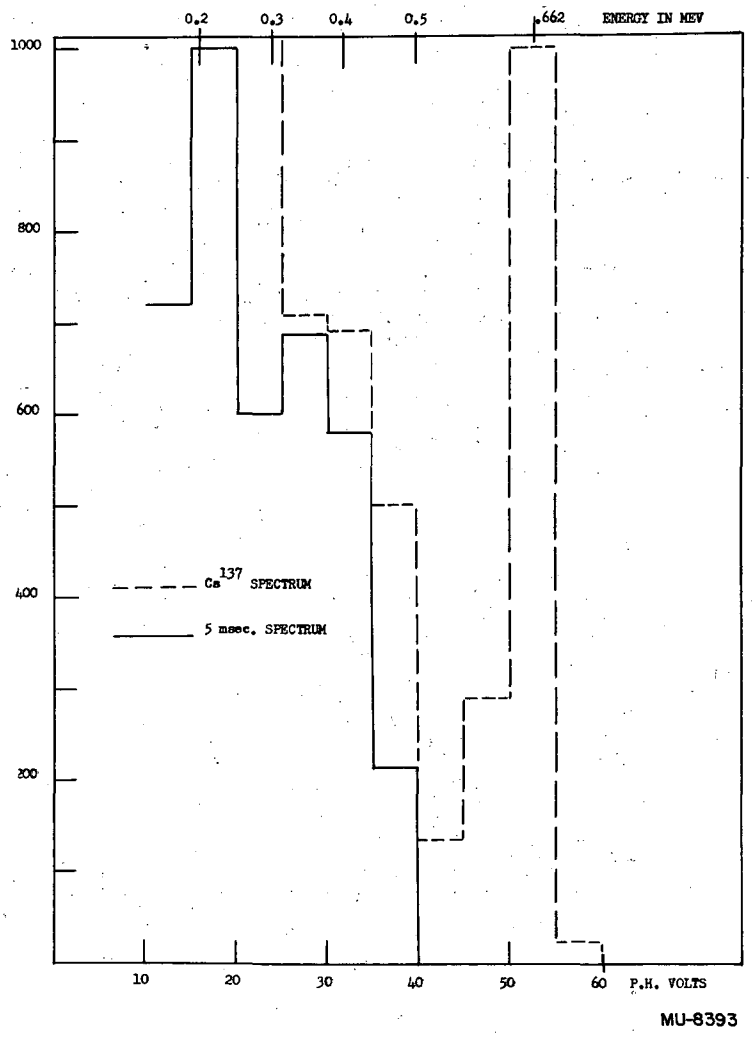

Fig. 4. Pulse height spectra of Cs^{137} and 5.5 m sec activity.

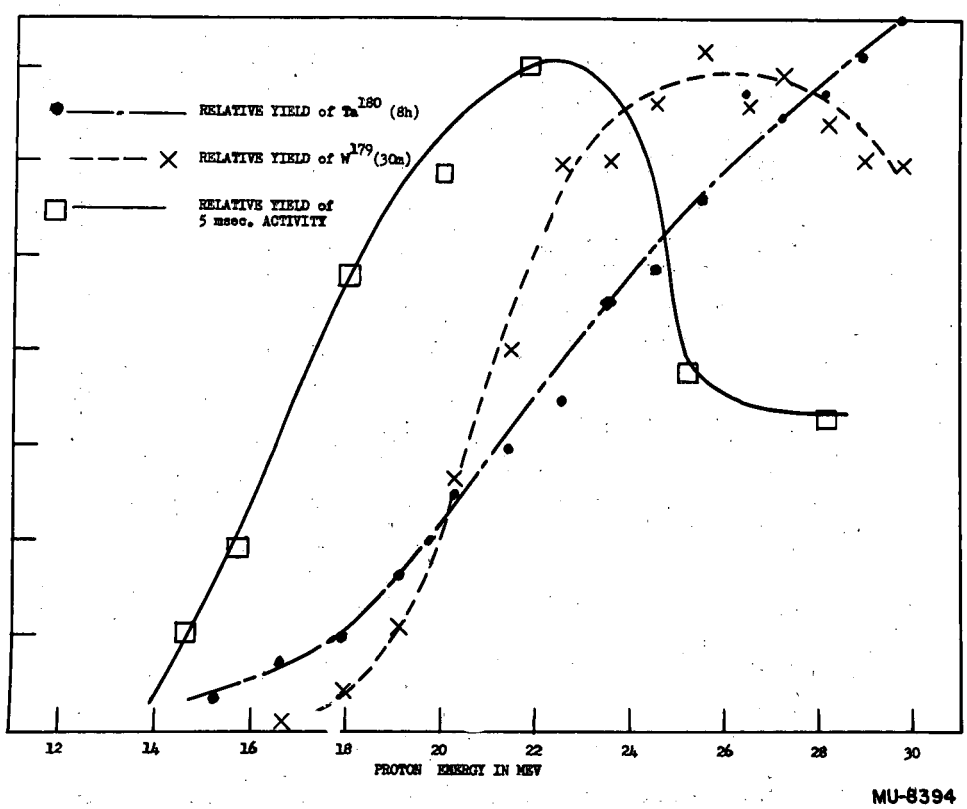

Fig. 5. Excitation functions for three activities from Ta + p.

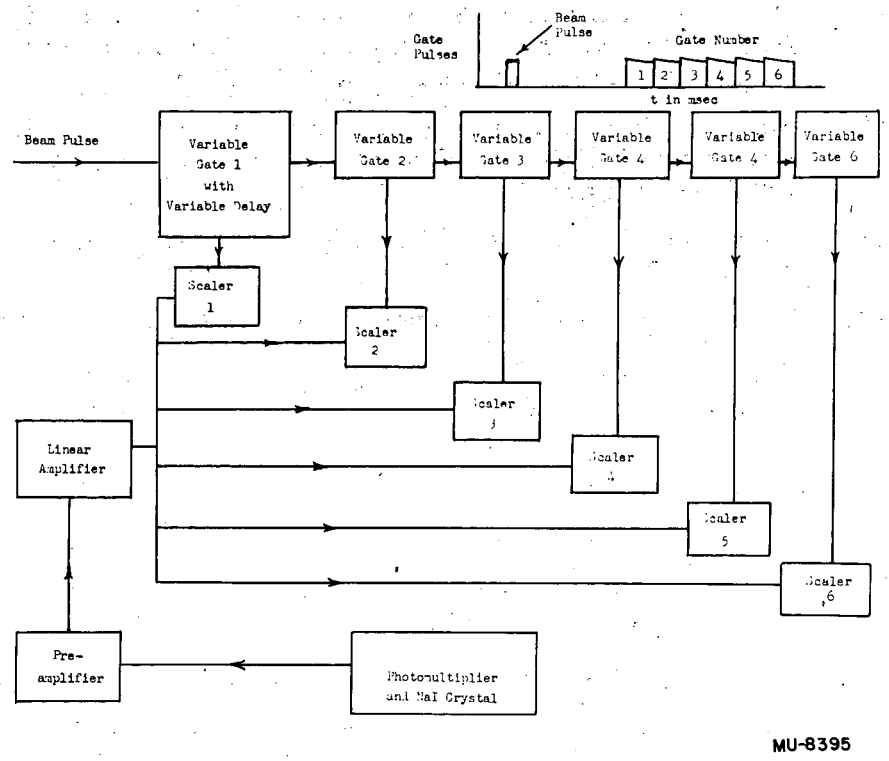

Fig. 6. Block Diagram of Electronics.

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