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April 19, 1956

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Dear Mr. Taschek:

As a result of the crash review program carried out in Oak Ridge in connection with the release of information to the Civilian Applications Program, the report LA-211 entitled "Radio-active Threshold Detectors for Neutrons"

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January 26, 1945

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RADIOACTIVE THRESHOLD DETECTORS FOR NEUTRONS

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ABSTRACT

Of nine (n,p) and (n,α) reactions studied, only $Be^9(n,\alpha)He^6$, $P^{31}(n,p)Si^{31}$ and $S^{32}(n,p)P^{32}$ gave measurable activities of the residual nucleus after bombardment by neutrons below 2 Mev, and of these three only $P^{31}(n,p)Si^{31}$ had the combination of cross section and half life to make a very practical threshold detector for neutrons. The 4.1 hour non-capture excitation of In was investigated as a function of neutron energy. The threshold and variation of cross section with energy of the $P^{31}(n,p)Si^{31}$ reactions were measured in detail.

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RADIOACTIVE THRESHOLD DETECTORS FOR NEUTRONS

The possibility of using thin foils of elements which become radioactive under neutron bombardment above a threshold energy for investigating neutron sources of unknown number and energy distribution led to a study of the following reactions:

$Be^9(n,\alpha)He^6$

$Al^{27}(n,p)Mg^{27}$

$Si^{28}(n,p)Al^{28}$

$P^{31}(n,p)Si^{31}$

$S^{32}(n,p)P^{32}$

$K^{39}(n,p)^{39}$

$Ti^{49}(n,p)Sc^{49}$

$V^{51}(n,p)Ti^{51}$

$Ga^{69}(n,p)Zn^{69}$

These reactions seemed most likely to prove easily usable detectors since the half lives of the activities were neither too short nor too long and the calculated thresholds ranged from thermal neutron energies to about 2 Mev. Special effort was made to find reactions with very low threshold energies.

The above reactions and their dependence on neutron energy were studied by use of the Li(p,n) source for the most part. No chemical separations were made. Only the reaction $P^{31}(n,p)Si^{31}$ was studied in great detail since it appeared most useful. Except for phosphorus, which will be discussed in detail later, the results were as follows:

$\underline{Be^9(n,\alpha)He^6}$ - calculated threshold 780 kv. The half life of 0.8 sec proved too short to use without special transfer methods from source to counter. With neutrons of 1.2 Mev incident of Be the activity could be observed by quickly

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shutting off the source of neutrons and watching the decay inactivity of a large block of Be in front of the Geiger counter. The activity and counting efficiency are high and special methods could be used for detection if necessary.

$Al^{27}(n,p)Mg^{27}$ - calculated threshold 1.95 Mev, half life 10.2 mins. An aluminum foil bombarded with 2.2 Mev neutrons from the D-D source in the backward direction showed no activity. No further investigations were made since Bretscher has measured the yield up to 4 Mev and found his observable threshold at about 2.3 Mev.

$Si^{28}(n,p)Al^{28}$ - calculated threshold 2.7 Mev, half life 2.4 min. A small activity was observed at thermal energies and none at 2.2 Mev. No further investigations were made.

$S^{32}(n,p)P^{32}$ - calculated threshold 1 Mev, half life $\sim$ 14 days. 500 mg samples in the form of thin foils were exposed for four hours at 1.7 Mev and 1.8 Mev neutron energy. The activity was only about one twentieth of background. If this activity is real then the cross section at 1.7 Mev is approximately 1×10^{-27} cm^2, i.e. about the same as that for phosphorus, which seems reasonable. The long half life, however, makes it impractical to study in more detail with the Li(p,n) source, although perhaps large quantities of sulfur could be used.

$K^{39}(n,p)A^{39}$ - calculated exothermic by 9 Mev, half life 4 mins. No activity at all was found up to a bombarding neutron energy of 1.2 Mev. This reaction is misassigned or the masses are incorrect since at 1.1 Mev the barrier penetration of the protons is already 0.1 percent which should give a detectable activity.

$Ti^{49}(n,p)Sc^{49}$ - calculated threshold 1.1 Mev, half life 57 min. The metal was investigated up to E_n = 1.6 Mev; no activity of any kind was detected due probably to the facts that this isotope is present only to $\sim$ 5.5 percent and

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the barrier against proton emission is relatively high.

$V^{51}(n,p)Ti^{51}$ - calculated threshold 180 kev, half life 3 mins. Samples of the metal were studied in detail to E_n = 1.6 Mev. Between about 600 kev and 1.6 Mev the activity is easily observable and practically constant; below 600 kev the activity rises very approximately as 1/v as shown in Fig. 1. All, or at least that part of the activity up to 600 kev, is probably due to $V^{51}(n,\gamma)V^{52}$ which decays with approximately 4 min half life and which thus, due to its large intensity at high energies even though Cd covered, masks the threshold of the n,p reaction.

$Ga^{69}(n,p)Zn^{69}$ - calculated threshold 250 kv, half life 57 min. Studies in detail but found no 57 min activity. Strong 20 min (n,γ) from Ga^{70} and strong 14 hr (n,γ) from Ga^{72} even at high energies make this useless as a threshold detector.

Elements higher in Z do not appear practicable since the barrier is too high for appreciable activity at low neutron energies. Possible elements below Z = 12 have too short half lives for practical use.

$P^{31}(n,p)Si^{31}$ - The threshold of this reaction as calculated from the β-ray energy[1] of 1.8 Mev should be at about 1 Mev neutron energy. The half life has been reported as 150 to 180 min[2] and 170 min[3]. The half life observed from six active foils was 145 ± 5 minutes. The neutron energy at which activity in a 500 mg foil could just be measured was 1.4 Mev. At this energy the activity was about 20 percent of background after a two hour exposure to the Li(p,n) source. A Chicago type thin walled counter was used to measure the activity.

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1) Seaborg, Rev. Mod. Phys. 16, 1(1944)

2) Fermi, Amaldi, D'Agostino, Rasetti and Segre, Proc. Roy. Soc. A146, 483 (1934).

3) Newson, Phys. Rev. 51 624(1937)

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Relative data were rapidly obtained with 1 gr samples of red phosphorus spread evenly between two sheets of scotch tape to an area of 15 cm^2. These foils were activated at a distance of 2" from the Li target and the neutron flux was monitored by means of the long counter[4]. Activities were measured by wrapping the activated foils around the Geiger counter and following the activity from 3 to 4 hours. The zero time activities were converted to infinite exposure activities by use of exposure time and the 145 minutes half life. In this way activities per unit neutron flux were obtained in the energy interval from 1.4 to 1.9 Mev.

In order to determine the cross section for the above reaction, foils were made of 500 mg of phosphorus evenly dispersed in a thin film of flexible collodion of total area 15 cm^2. The total weights of collodion and phosphorus were about 700 mg making a weight of about 46 mg/cm^2 which corresponds to a <u>maximum</u> stopping power of about 80 kev for 1.8 Mev electrons. During irradiation the foils were folded into an area 3 x 2.5 cm and placed on the front of a fission chamber containing a foil of 763 γ of 25. At two inches from the target there was 1/4" between foil and phosphorus; the $1/r^2$ correction has been applied to the cross-section calculations. During approximately two hours' irradiation the fission count was taken at frequent intervals and the flux kept as constant as possible in time. From five to ten minutes usually elapsed between measuring activity and stopping the exposure. From this activity and the fission cross section the $P^{31}(n,p)Si^{31}$ cross section was found after determining the Geiger counter sensitivity. Three separate exposures of three different collodion foils were made at 1.8 Mev and one measurement at 1.4 Mev neutron energy. When the relative activities measured with the larger foils were normalized to 1.8 Mev, the absolute 1.4 Mev measurement checked well with the normalized one so the curve was considered established.

4) LAMS-66.

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The Geiger counter sensitivity was obtained in the following way. Uranyl nitrate was dispersed in a thin collodion foil and the counting rate obtained; when this foil was cut into several strips the sum of the counting rates of the strips equalled the counting rate of the whole. Therefore a large foil of this type containing an accurately weighed amount of uranyl nitrate (chosen because of its small fraction of uranium) was cut into strips of identical size and shape with the phosphorus foils and all the activities determined. The fractional counting rate of a single strip then measured the mass of uranium in that strip and therefore the number of β's/sec being counted. From the known 732.5 β's/mg-minute, the counter efficiency was determined as 29 percent for uranium β's under these conditions. Since the phosphorus β's are somewhat softer than those from U, absorption data with thin aluminum foils was obtained from a thickness of 7 mg/cm^2 to 96 mg/cm^2. In this range the P and U absorption curves were linear on a semi log scale of activity as a function of absorber thickness and from the slopes of these curves near zero absorption the efficiency for the β's from the phosphorus foils was calculated to be 25 percent.

The ratio of the $P^{31}(n,p)Si^{31}$ cross section to the 25 fission cross section was calculated from

$$\frac{\sigma_P}{\sigma_f} = \frac{1}{\varepsilon_\beta}\,\frac{C_P}{C_f} \times \frac{N_{25}}{N_P} = \frac{31}{235}\,\frac{1}{\varepsilon_\beta}\,\frac{M_{25}}{M_P}\,\frac{C_P}{C_f}$$

after correcting for the amount of 28 present in the foil.

ε_β = Geiger counter efficiency for phosphorus βs.

$$C_P = \frac{\beta \text{ counts/sec at zero time}}{1 - e^{-.693\, t/T_{1/2}}} = \text{counting rate for infinite exposure}$$

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[redacted]

C_f = fissions/sec - corrected for time variations in flux if any.

N_{25} = number of 25 atoms

N_P = number of phosphorus atoms

The cross section σ_P was obtained from this ratio by using $\sigma_f = 1.33 \times 10^{-24}$ cm^2 in the energy interval used. The average of the three separate exposures at 1.8 Mev gave

$$(\sigma_P/\sigma_f)^{1.8 \text{ Mev}} = 4.77 \times 10^{-3} \pm 3 \text{ percent}$$

where the error is due only to deviations from the mean of the three values. Similarly

$$(\sigma_P/\sigma_f)^{1.4 \text{ Mev}} = 0.137 \times 10^{-3}.$$

Bretscher[5] has obtained relative data on this reaction between 2.05 Mev and 4.05 using D-D neutrons. In order to join his data to that obtained here one of the collodion-type foils was exposed to the 3.1 Mev maximum energy neutrons from the D-D source in Z in the same way as above.

The ratio

$$(\sigma_P/\sigma_f)^{2.9 \text{ Mev}} = 40.7 \times 10^{-3} \text{ was thus obtained.}$$

The cross section obtained from this ratio was then used to normalize Bretscher's data to that found here. The fit near 2 Mev where the two sets of data join is not very good and may indicate that the 2.9 Mev cross section is somewhat low.

In Fig. 2 is plotted the cross section for the reaction $P^{31}(n,p)Si^{31}$ so obtained as a function of energy, Bretscher's data being joined to the present data by means of the normalization at 2.9 Mev. It will be seen that there are some indications of a resonance near 1.5 Mev; this seems to be real since several check runs were made in this short energy interval. Bretscher's original data

5) Bretscher, personal communication.

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which are plotted on a linear activity scale shows a strong resonance near 3.5 Mev which is washed out in this plot. The cross section increases by a factor of several hundred between 1.4 Mev and about 2.5 Mev.

In Fig. 3 are plotted the penetration factor $P_o = e^{-2C}$ by the fractional neutron energy above threshold since this quantity should indicate roughly the theoretical expectation of how the cross section should vary with energy below the top of the barrier. $E_{min}\, P_o/E_n$ is shown for the reaction $P^{31}(n,p)Si^{31}$ for elementary nuclear distance $r_o = 1.65 \times 10^{-13}$ and $r_o = 1.3 \times 10^{-13}$ cm assuming that the β-ray energy is about 1.75 Mev, which makes the reaction threshold 1.0 Mev. The same quantity is also shown for $r_o = 1.3 \times 10^{-13}$ and a β-ray energy of 1.45 Mev, corresponding to a reaction threshold of about 700 kv. For purposes of comparison $E_{min}\, P_o/ E_n$ has been plotted also for $P^{31}(n,\alpha)Al^{28}$ assuming a threshold of 900 kv and $r_o = 1.3 \times 10^{-13}$. In all these curves the factor e^{-2C} is calculated as the penetration in percent and then multiplied by the ratio of the minimum energy used to the energy in question. The observations have been normalized at 2.6 Mev to the two calculated curves for $r_o = 1.3 \times 10^{-13}$ cm in the $P^{31}(n,p)Si^{31}$ reaction. It is immediately seen that only the curve using the 700 kv threshold energy gives a reasonable fit at all. Seaborg[1)] gives only Kurie, Richardson, and Paxton's[6)] β-ray energy of 1.8 Mev corresponding to a 1.0 Mev threshold but more recent measurements[3,7)] indicate that 1.5 Mev or even less is more likely correct, which would be good evidence in favor of choosing the 700 kv threshold to get a penetration function fit. However, such a theoretical picture is probably much too simple to explain the variation in cross section of a mediumly heavy element over such a wide energy range. The penetration function for the reaction $P^{31}(n,\alpha)Al^{28}$ becomes appreciable near 2.5 Mev and at 4 Mev is almost equal to that for $P^{31}(n,p)Si^{31}$ indicating that strong competition can be

6) Kurie, Richardson, and Paxton, Phys. Rev. 49 368(1936)
7) Paxton, Phys. Rev. 51, 170(1937)

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[redacted]

expected from the former above about 3 Mev which might reduce the cross section for $P^{31}(n,p)Si^{31}$ and account for a bad fit at high energies. The Al^{28} has a 2.4 min half life and was probably not observed under the present experimental conditions since several of these short half lives would have elapsed before counting began after an exposure.

It is interesting to note here that of 29 possible reactions listed by Uchiyamada[8] which go either by (n,p) or (n,α), for all but three (both positive and negative), the reaction energies are more positive for (n,α) than for (n,p). The listed reaction energy for P^{31} $(n,p)Si^{31}$ is -1 Mev and that for $P^{31}(n,\alpha)Al^{28}$ is given as -900 kev, whereas it appears here that a reaction energy of - 700 kev may be more nearly correct for $P^{31}(n,p)Si^{31}$.

$\underline{In^{115}(n,n)\gamma}$ - At Fermi's suggestion the 4.1-hour non-capture excitation period of In^{115} was investigated at neutron energies of 600, 800, 1000, and 1500 kev. Packets of four standard indium foils inside a tight cadmium box were exposed for periods from two to four hours to the neutrons from the Li(pn) reaction and the activity of the innermost foil measured in each case. The 4.1-hour period was measurably excited at 800, 1000, and 1500 kev but absent or extremely weak at 600 kev. In this energy interval, unfortunately, the 54-minute (nγ) period and a long period, presumably the 48-day (nγ) in In^{113}, are still present, the 54-minute with such strength for these almost saturated exposures that the 4.1-hour period is not obviously apparent for about eight hours after the exposure. The long-period activity is weak but must be measured reasonably accurately since it is the background for the other activities. It was not found possible to distort appreciably the activity-time curves by using Pb absorbers since the 54-minute activity consists principally of γ-rays whose energies lie on both sides of the 350-kev γ-ray from the non-capture excitation.

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8) Uchiyamada, CP-1336

[redacted]

Fig. 4 shows the 4.1-hour activity calculated for infinite exposure in counts/sec on a Chicago type Geiger counter per neutron incident on 100 mg/cm^2 foils of In as a function of neutron energy. Rough values of the corresponding 54-minute and 48-day activities are shown in the same figure for comparison with the 4.1-hour period. The non-capture excitation shows a threshold near 600-kev neutron energy and rises rapidly until its saturated activity at 1.5 Mev is about one-seventh of the activity of the 54-minute activity. Goldhaber, Hill and Szilard[9] have reported that the neutrons from Rn-α-Be (maximum energy 13.7 Mev) excites the 4.1-hour activity more strongly than the 54-minute. A rough estimate would indicate that about 5-Mev neutrons would be required to make the 4.1-hour activity equal to the 54-minute.

The threshold for the 4.1-hour activity appears to be sharp and the cross section rises rapidly with neutron energy but the 54-minute and very long period activities make its use as a threshold detector difficult.

[redacted]

9) Phys. Rev., 55 47(1939)

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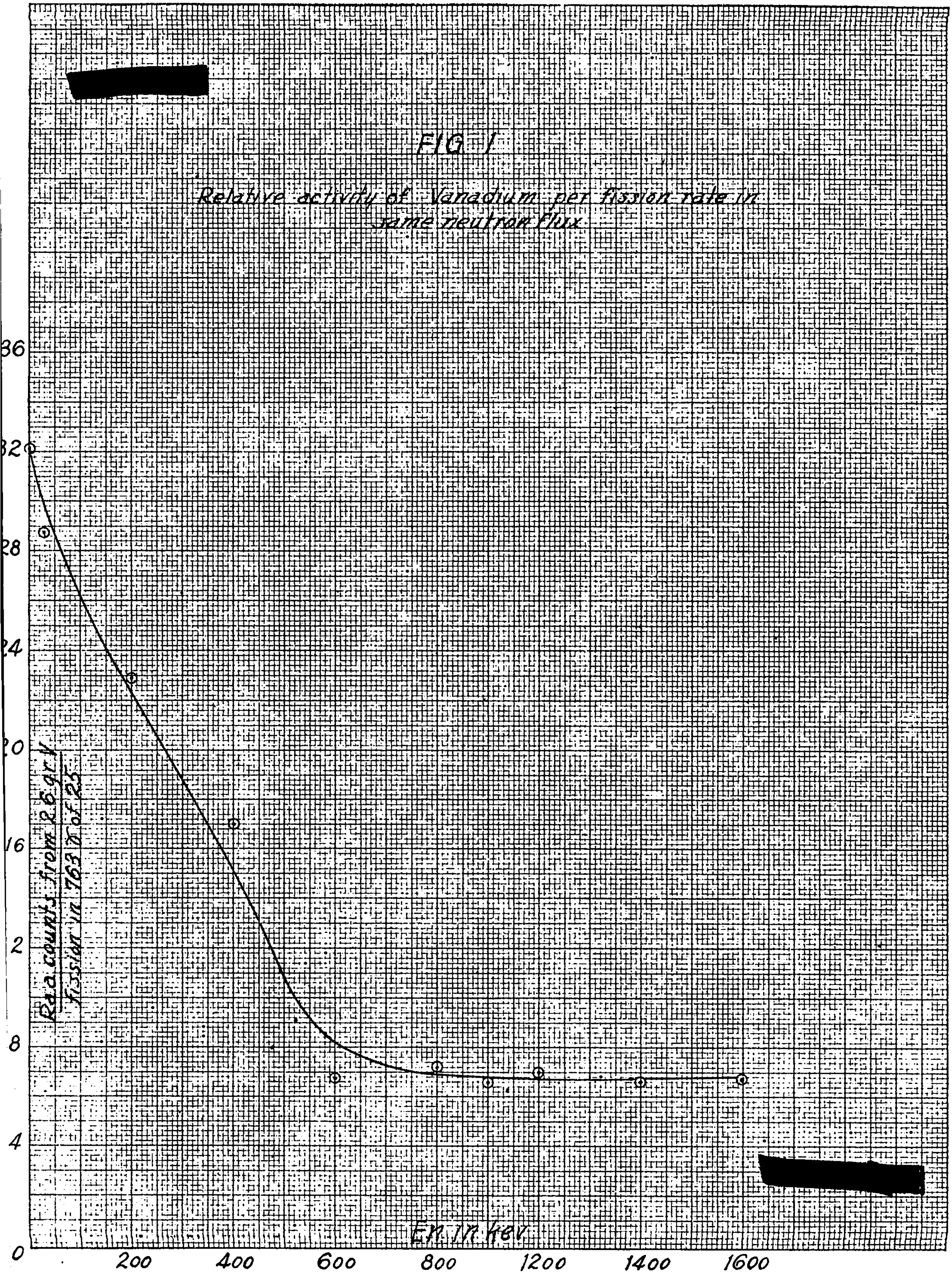
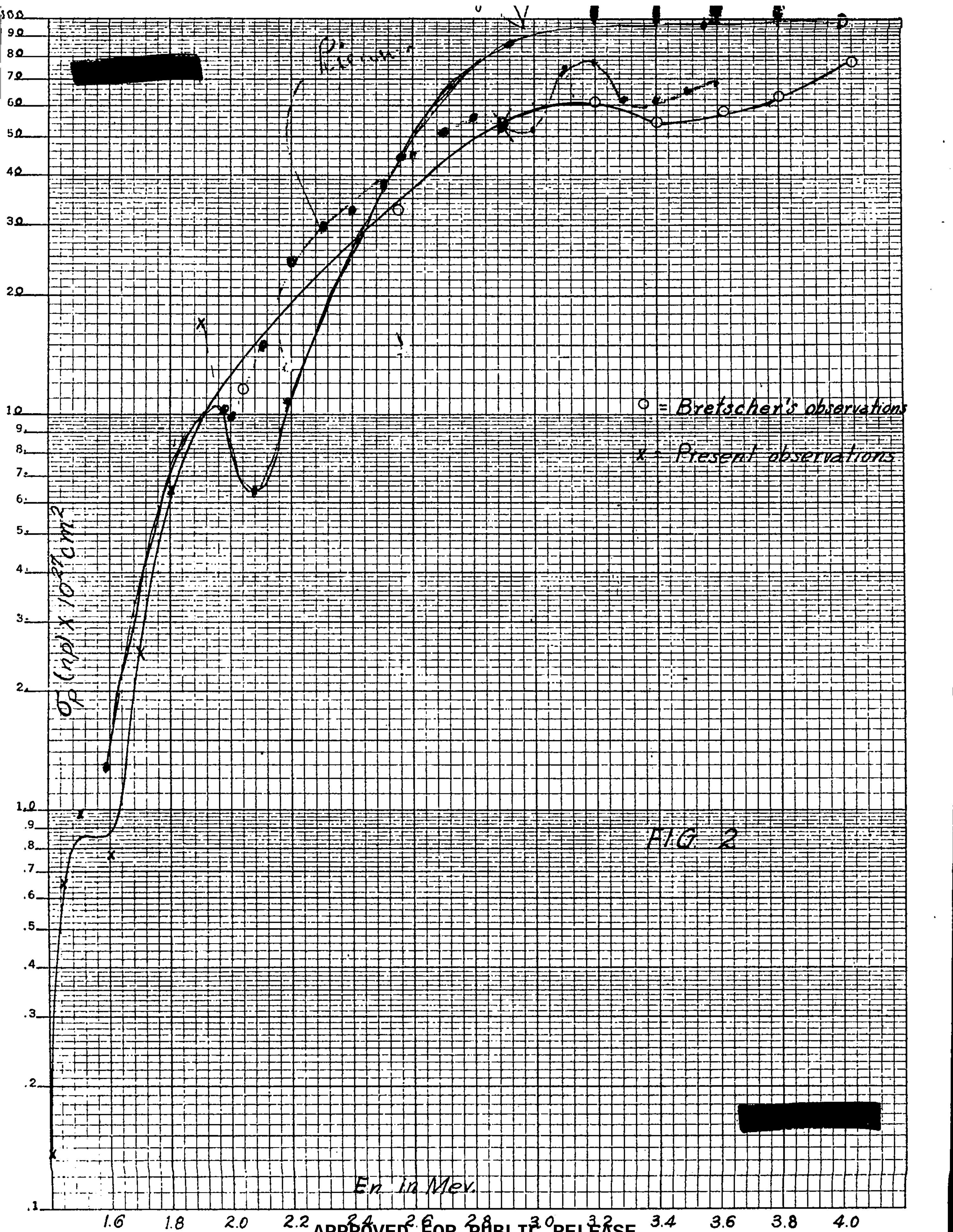

[redacted]
FIG I
Relative activity of Vanadium per fission rate in same neutron flux
Rel. a. counts from 2.6 gr V
Fission in 763 γ of 25
36
32
28
24
20
16
12
8
4
0
200
400
600
800
1200
1400
1600
En in kev
[redacted]



[redacted]
○ = Bretscher's observations
x = Present observations
σp (np) x 10-27 cm2
En in Mev.
1.6
1.8
2.0
2.2
2.4
2.6
2.8
3.0
3.2
3.4
3.6
3.8
4.0
[redacted]

FIG. 2

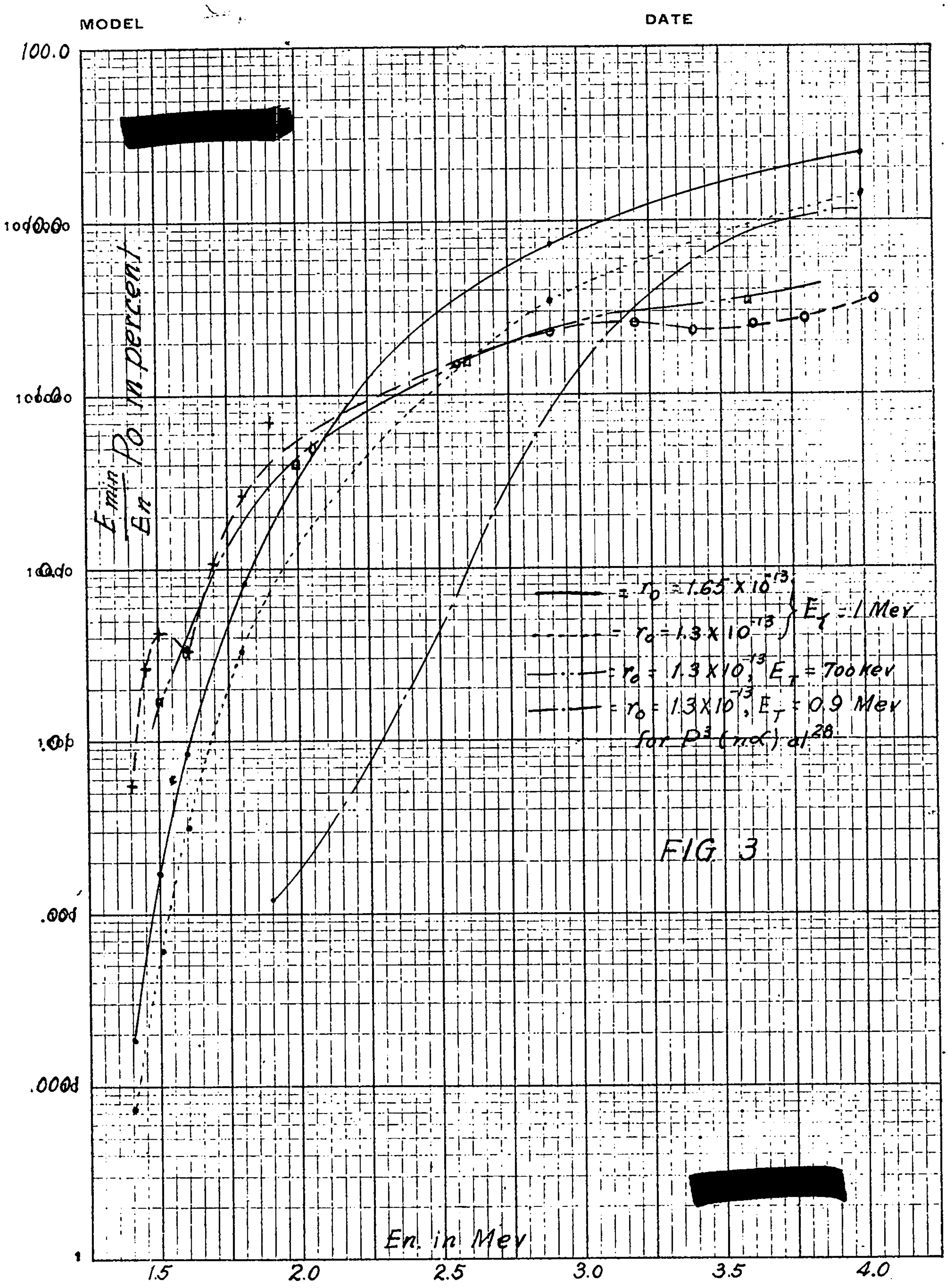
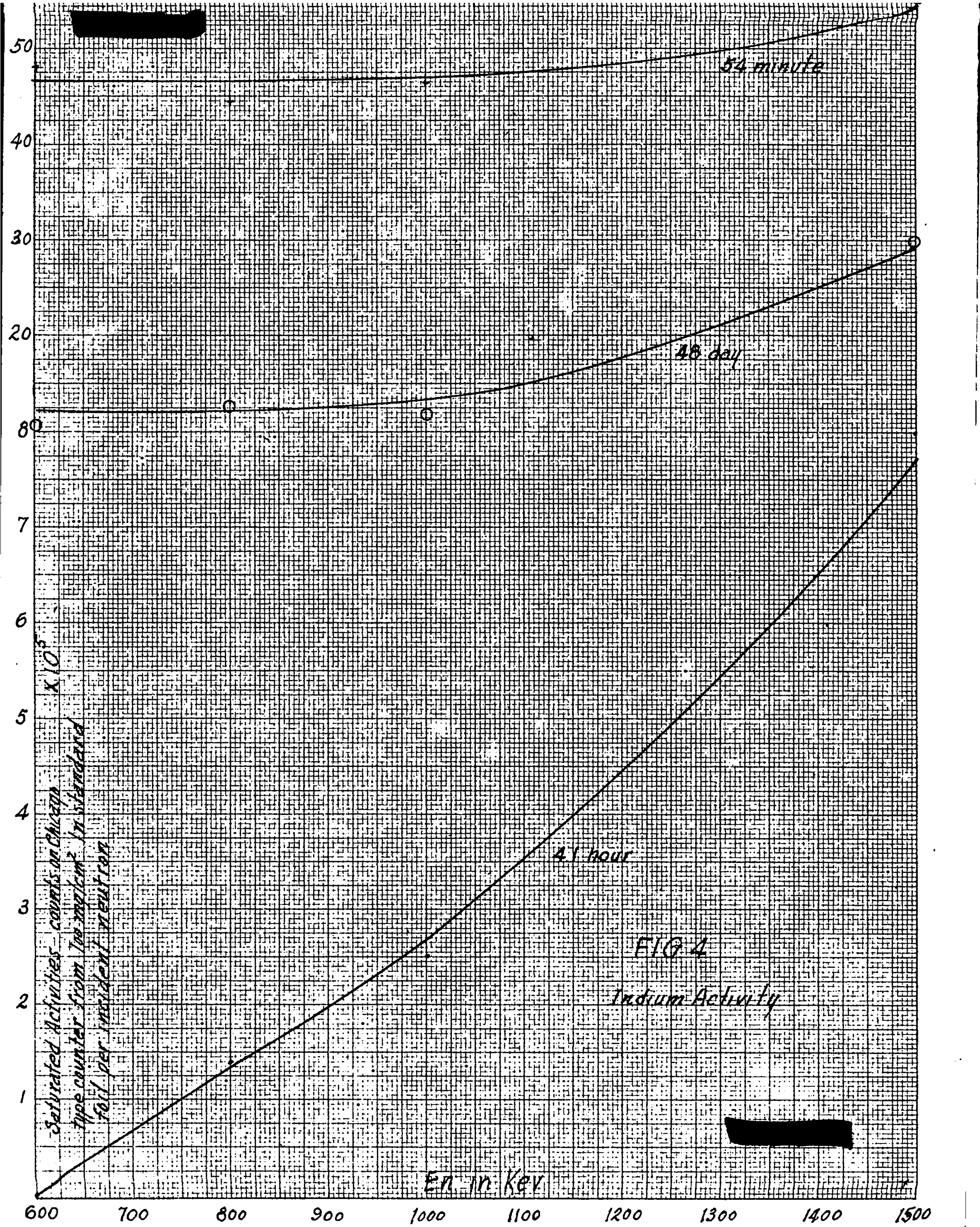

MODEL
DATE
[redacted]
100.0
.001
.0001
1
$\frac{E_{min}}{E_n}$ Po in percent
$= r_o = 1.65 \times 10^{-13}$
$= r_o = 1.3 \times 10^{-13}$
$E_\gamma = 1$ Mev
$= r_o = 1.3 \times 10^{-13}, E_\gamma = 700$ kev
$= r_o = 1.3 \times 10^{-13}, E_\gamma = 0.9$ Mev
for $P^3 (n\alpha) Al^{28}$
[redacted]
En. in Mev
1.5
2.0
2.5
3.0
3.5
4.0

FIG 3



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