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THE USE OF THORIUM AS A THRESHOLD DETECTOR FOR FAST NEUTRONS

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ABSTRACT

Counters have been built and tested in which thorium fission is used as the detector of fast neutrons. At first a large background fission rate was observed when the chamber was irradiated by slow neutrons. This background was shown to be largely a photofission effect produced by gamma rays which were emitted when the materials of and around the counter captured slow neutrons. Iron, copper, and cadmium appear to be the worst offenders. For a particular counter an 83-gm iron cylinder placed around the counter multiplied the background by five. A similar copper cylinder raised the background by a factor of 3.3.

With a suitable arrangement and choice of materials, it was possible to build a counter which had an efficiency of about 2×10^{-6} for fast neutrons ($E > 1.2$ MV) yet gave a background fission rate of but 0.5 counts per minute when in a slow neutron flux, $nv \sim 10^7$.

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THE USE OF THORIUM AS A THRESHOLD DETECTOR FOR FAST NEUTRONS

Since the thorium fission threshold appears to occur in the neighborhood of 1.2 MV neutron energy, a thorium fission chamber might have many applications for the detection of fast neutrons in the presence of large numbers of slow and thermal neutrons.

EXPERIMENTAL TESTS

The first chamber constructed consisted of three interleaved Pt cylinders coated with 1 mg/cm^2 of ThO_2, and was tested in the presence of a slow neutron flux $nv \sim 10^7$. An unexpectedly high background fission counting rate (0.2 count per sec) was observed. If this background was produced by the slow neutrons themselves, thorium would be almost useless as a threshold detector for fast neutrons. Hence tests have been conducted with the object of locating and eliminating this background fission rate.

Several hypotheses could account for the background rate:

(A) fast neutrons were present from the source,

(B) alphas or gammas were piling up to give large pulses,

(C) thorium does not have a sharp threshold at 1.2 MV but simply an exponentially decreasing cross section,

(D) the ThO_2 was contaminated with small amounts of 25,

(E) some isotope of thorium (RdTh, Ionium, or 09) is a slow-neutron fissionable material and present in small amounts,

(F) photofission of thorium is produced by the photons from the target or by photons emitted when slow neutrons are captured in the vicinity of the counter.

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As a neutron source thick lithium metal was bombarded by magnetically analyzed 2.0 MV protons which had been acc rated by a pressure Van de Graaff. The maximum energy of the neutrons from the target is 0.26 MV, but a small deuteron component hitting in the analyzer can give fast neutrons from the C + D and D + D reactions. These fast neutrons were shown to be negligible by the following tests:

1. The magnet current was raised and lowered so that the proton beam was just off the target (the effect on the deuteron beams is slight). In each case the background fissions disappeared.

2. The slowing-down material around the counter (15 cm radius sphere of paraffin) was removed. This reduced the background rate by a factor of six.

3. The proton energy was set below the threshold value for the production of neutrons from $Li^7(pn)Be^7$ reaction. The background rate was then cut to ~.02 counts per second (i.e. by a factor of ten).

Thus hypothesis A is excluded.

Hypothesis B was excluded by observing visually the alphas on an oscilloscope. Many more small pulses (source unknown) are present when the chamber is irradiated by slow neutrons but the individual pulses were still observable on the oscilloscope and there was no piling up to give large pulses. The pulses were very sharp since the voltage between cylinders was 2000 and the distance between adjacent cylinders was 3 mm. The calculated capacity of the counter was 5 mmfd. and with the 5-megohm grid resistor this gives a CR of 2.5×10^{-5} sec. resolving time. The small separation of the cylinders discriminated strongly against alpha pulses but did not greatly affect the size of the fission pulses.

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Hypothesis C is inconsistent with the reduction of background which occurs when the slowing-down material is removed.

The slow-neutron flux inside the chamber was calibrated by using a known amount of uranium (0.2 μgm.) on the cylinders instead of the thorium coating. It was then calculated that the previous thorium coating would have to have 75 parts per million (ppm) of uranium if hypothesis B was to be tenable. However, the thorium oxide had been carefully purified by the ether extraction method by S. I. Weissman. This extraction technique has been checked by adding small known amounts of uranium and was also checked by a sensitive fluorescent technique capable of detecting 2 or 3 ppm of uranium.[1)] The test was extended by Weissman and Duffield until they concluded that the repurified thorium contained less than 0.04 ppm of uranium. (Ordinary Eimer and Amend thorium nitrate contains 8-10 ppm of uranium). Hence uranium contamination of the thorium was roughly a factor of 2000 too low to explain the observed slow-neutron fission background.

If hypothesis E is assumed and if RdTh is the offender it would have to have a fantastically high cross section since its half life is but 2.02 years. Ionium ($T_{\frac{1}{2}} = 6.9 \times 10^4$ years) has a long enough half life to account for the background provided (a) that it is a slow-neutron fissionable material, and (b) that the thorium was extracted from ores containing several percent of uranium. Condition (b) is quite probable but the known evidence[2)] is against (a). If the unknown isotope 09 were a slow-neutron fissionable substance, and if it were present as 0.5 ppm of ordinary thorium it would account for the observed background. 09 has an even number of protons and an odd number of neutrons similar to the other two known slow-neutron fissionable substances, 25 and 49, so if it

1) S.I. Weissman and R.F. Duffield, L. A. 15
2) Jentsehke, Prankl, and Hernegger, S. B. Akad. Wiss. Wien. Abt IIa, 151, p. 147-157, 1942.

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exists, one might expect it to undergo slow-neutron fission. Also noteworthy is the fact that 09 bears the same relationship to 02 that 25 does to 28. Since 28 and 02 have such similar fission characteristics, one might expect the same for 25 and 09. Hence it is conceivable that part of the fission background could be caused by this unknown isotope. However, as we shall see below most of the background can be explained by hypothesis F.

The below-threshold voltage test described earlier indicated that less than 10% of the background could be photofission effect of the 17 MV gammas from the Li target. Hence if the rest of the background is to be attributed to photofission, it would have to be caused by the photons emitted when slow neutrons are captured in the vicinity of the counter. The binding energy of a neutron may be 2-10 MV so that quite energetic gammas are possible if the energy is all concentrated in one photon. The first evidence that the slow-neutron background effect was in reality a photofission effect was the fact that surrounding the counter with cadmium gave quite different results than surrounding the chamber with pyrex glass or boron carbide. The cadmium cover cut the slow-neutron flux in the chamber (as measured by the activity of a copper wire in the center of the counter) by a factor of ten, but reduced the background fissions by less than a factor of two. However, when the chamber was surrounded by pyrex glass the slow-neutron flux and background fissions were each reduced by a factor of 3. The difference between the cadmium effect and the boron effect may be attributed to the difference in the gamma rays emitted when cadmium and boron capture slow neutrons. Kikuchi, Husini, and Aoki[3] (KHA) report that cadmium upon slow-neutron capture seems to give off both soft gamma rays (2 MV) and hard gamma rays of 10 MV energy. Since 10 MV

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3) S. Kikuchi, K. Husini, and A. Aoki, Proc. P.M.S.J., 18, 192, 1936.

is well above the photofission threshold (~ 6MV) the gamma rays could contribute to the observed background fissions. Boron, however, has no known gammas of energy greater than 440 KV and hence could not give a photofission effect. These observations indicate that photons emitted after slow-neutron capture by materials of the counter could account for much of the observed fission background.

The first counter was constructed chiefly of brass so its effect was tested by placing additional amounts of brass about the counter. A larger background resulted. Hence a new counter was constructed of pure aluminum since a similar preliminary test showed no effect from extra aluminum around the chamber. Also the ratio of the aluminum to brass cross sections for gamma-ray excitation by slow neutron capture is 1/18.[3)]

The new aluminum counter with seven concentric cylinders contained approximately ten times as much thorium. Hence it was easier to detect small changes in background fission rate. A 25 fission chamber was located in the paraffin to monitor the slow neutron flux, and the brass target tube was also replaced by an aluminum tube.

These changes reduced slightly the background fission rate although the amount of thorium had been increased by a factor of ten. The effect of various materials on the fission background was then tested by surrounding the counter with a cylinder of the substance in question. A 83 gm copper cylinder increased the background fission rate by 3.3. A similar 83 gm iron cylinder multiplied the background fissions by 5. One hundred seventy grams of aluminum piled around the counter increased the background by less than 1.5.

These results are compatible with the gamma ray energies and cross sections for gamma ray excitation by slow neutrons reported by KHA.[3)] For aluminum they find the gamma ray energy to be 4.05 ± 0.29 MV (presumably too low to produce

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photofission) and the cross section to be 0.18 ± 0.18 barns. Copper and iron have roughly eighteen times as large a cross section (σ_{Cu} = 2.9 ± 0.8, σ_{Fe} = 3.1 ± 0.4), and the gamma ray energy is reported as 6.6 ± 0.24 MV from iron, and 6.15 ± 0.36 MV from copper, which energies certainly should be sufficient to produce photofission. It is also noteworthy that KHA report that for both iron and copper there is some evidence of the existence of hard gammas, $E > 10$ Mv. The larger background effect from the iron may perhaps be attributed to a larger number of the higher-energy gammas.

The boron tests were repeated with the new counter and monitor arrangement and with much better boron shielding of the counter. The slow-neutron flux, as measured by the monitor in the paraffin, was halved, while the flux inside the counter itself (measured by the activity of a copper wire) decreased by a factor of 32. At the same time the background fissions were reduced by a factor of 215/50. Hence we may calculate the fraction of the background effect attributable to the slow-neutron flux in the counter itself. Let f be the fraction of the background fissions which are produced by a slow-neutron effect in the materials of the counter itself. Then

$$50/215 = (f/32) + (1 - f)/2$$

and f = 0.57. Hence approximately half the background effect could still be attributable to slow-neutron effects external to the counter. The materials external to the counter consisted chiefly of the 15 cm radius sphere of paraffin, its sheet-iron container and stand, and the brass housings of the preamplifiers for the monitor and fission chamber.

In a new counter design these external background sources were minimized by using aluminum-shielded leads 50 cm long from the chambers to the preamplifiers, and by replacing the paraffin and iron container by a composition container

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filled with water. The new counter (Fig. 1) also contained less material and the cylinders in particular were coated with but 1/15 the mass of thorium used in the previous counter. Also a thin Li target was used to avoid any effect from the 17 MV gammas coming from the 440 KV resonance. For the new arrangement the background was reduced by a factor of somewhat greater than 30. Since a factor of 15 was to be expected because of the smaller amount of thorium, the other factor may be taken as evidence that most of the background effect attributable to slow neutrons external to the counter had been eliminated.

In the final arrangement, the counter had an efficiency for counting fast neutrons ($E > 1.2$ MV) from a source in the center of the counter of approximately 2×10^{-6}. This counter had three layers of ThO_2 coating (3.5 mg/cm^2) and in a thermal neutron flux of $nv \sim 10^7$ it gave a background fission rate of 0.5 counts per minute.

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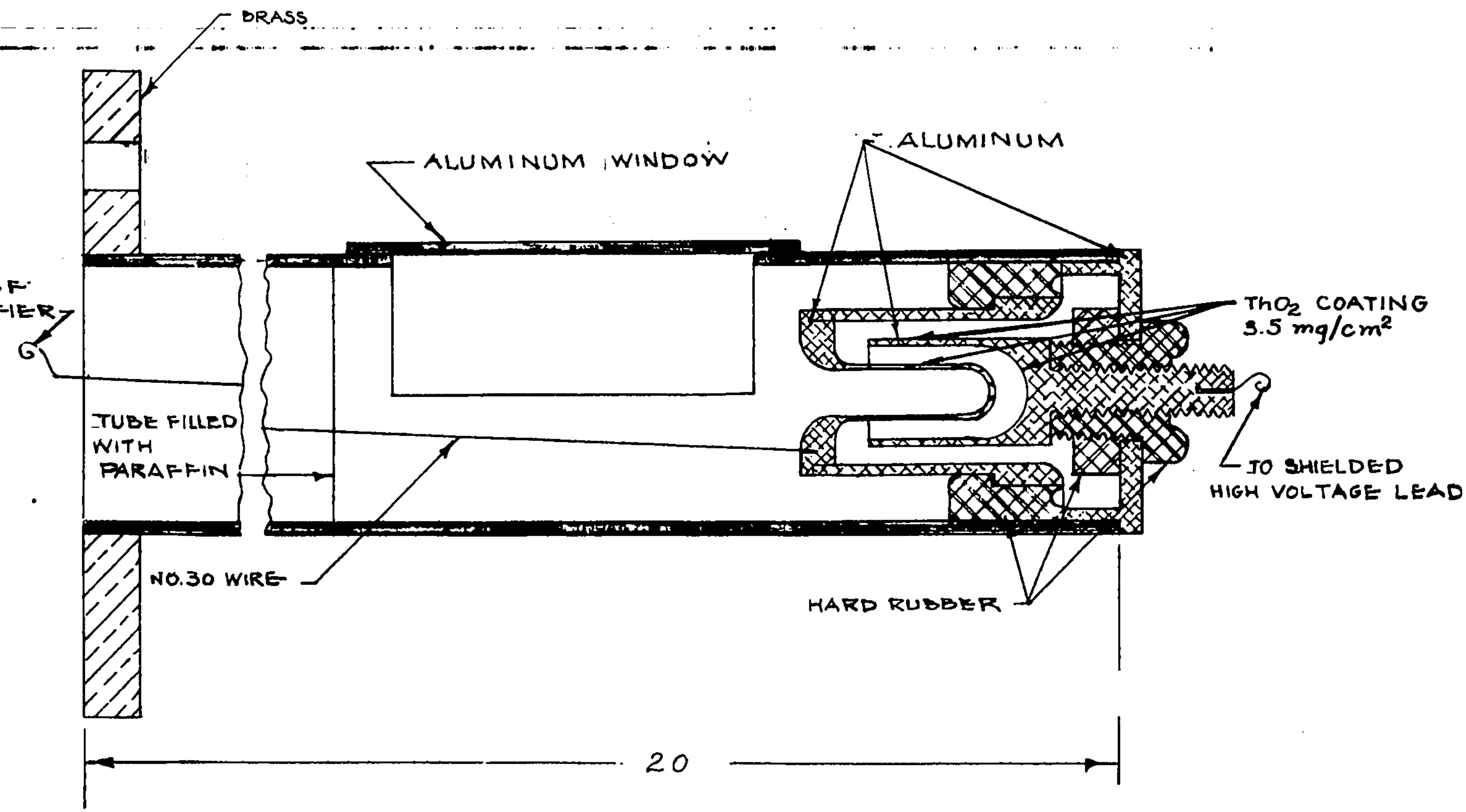

FIG. 1

SCALE - TWICE FULL SIZE

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