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DATE April 17, 1951

SUBJECT Radiation Studies 234-5

Building (I)

To File

FROM Wm. C. Roesch, J. S. Reddie,
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April 17, 1951

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From: Wm. C. Roesch, J. S. Reddie and E. C. Watson

RADIATION STUDIES 234-5 BUILDING (I)

Introduction and Summary

The purpose of this memorandum is to clarify the nature of the sources of neutrons and gamma rays which are of interest to the Health Instrument Divisions in the 234-5 building. It is shown that the chief source of neutrons is the alpha-neutron reaction in fluorine. The chief sources of photon radiations are x-rays associated with the alpha decays and the passage of the alphas through material, gamma rays from product, and inelastic scattering of alpha particles by fluorine.

Neutrons

An examination of the sources of neutrons to be expected in the 234-5 processes indicates only two important possibilities. One would expect occasional fissions from cosmic rays and possibly spontaneous fissions. The only other probable nuclear reaction resulting in neutron emission is the alpha bombardment of the compounds associated with the product. A search of the literature 1) concerning this type of reaction reveals only one of interest, F19 (alpha,n) Na22.

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Verification of the presence of these sources of neutrons can be found in measurements made by H. I. Operations personnel and the H.I. Physics Group. These surveys show some neutrons, all along the process line but in especially great numbers at that stage where the product is combined with fluorine. An especially striking proof of the presence of this reaction is provided by measurements with a recoil proton counter at hood #10 during the reduction process.²⁾ The product is here initially combined with fluorine only; the neutron intensity is quite high. When reduction is complete the product is in the form of pure metals and the neutron intensity has dropped to nearly zero. The relatively few neutrons found elsewhere along the line can be attributed to random fission and to scattered neutrons originating in the fluoride sources.

The neutrons from the $F(\alpha, n)$ reaction are the most numerous. An important point regarding these neutrons is that their energy spectrum is different from that of the sources used in calibrating neutron instruments. Fig. 1 shows approximate spectra of Po-B and Po-F, which should be quite similar to that of Pu-F. The spectrum of Ra-Be or Po-Be extends to much higher energies.

Gamma Rays

An examination of the sources of gamma rays (or X rays) to be expected in the $234\ 5$ processes indicates several possibilities. (1) One expects uranium L X-rays³⁾ produced by plutonium atoms which have just undergone decay and been left in an excited electronic state. These X rays will have energies in the range 12 to 21 Kev. (2) L X-rays of plutonium are expected from alpha particle excitation of plutonium atoms⁴⁾; these L X-rays will be similar to those above. Apparently⁴⁾ very few K rays of plutonium from this method of excitation are to be expected. X rays excited in other atoms than plutonium would be expected at various parts of the process but will be of too low an energy to be of interest. (3) Plutonium emits gamma rays in some of its disintegrations. Energies of 200 and 420 Kev have been reported in one case and 50 and 300 Kev in another⁵⁾. (4) Alpha bombardment of some of the atoms associated with the plutonium can be expected to lead to cases of inelastic alpha scattering with accompanying gamma emission; such reactions are denoted $(\alpha; \alpha, \gamma)$. Such a reaction has been reported for fluorine⁶⁾ as giving several gamma rays of energies 0.5 Mev and higher.

To determine which of these possible gamma rays are actually present in $234\ 5$ we made a series of absorption measurements at various places along the line. A Victoreen 1B65 geiger tube was used. This tube has aluminum walls 30 mg/cm² thick and is filled to 10 cm with argon and quench gas. It was operated with a cathode follower at the tube; an I.D.L. #161-G

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scaler was used as power supply and scaler. The absorbers were thin cylinders of polyethylene, aluminum, and copper which fit snugly around the geiger tube. Such absorption measurements can be expected to give valid results for the materials used, from about 10 to 100 Kev; consequently thicknesses of absorber sufficient to determine energies in this range were used.

Figure 2 shows the absorption curves made at the entrance to Hood 8 through the lucite window. No sources other than contamination of the hood interior were present; this contamination should not contain any fluoride. Energy components of 17 Kev, 40 Kev, and about 200 Kev or higher can be identified; our equipment was not designed to actually measure energies of the latter value. The 17 Kev component is an average of the L X rays of uranium and plutonium. The 40 Kev component is probably the low energy plutonium gamma ray. We believe that the high energy component is due to plutonium gamma rays since it seems to be too high an energy for K X-rays and no gamma from the fluorine reaction is to be expected.

Figure 3 shows the curves for the exit end of Hood 8 taken through the lucite. Several sources of fluorinated product were present. The same energy components as above can be identified. The high energy component is about six times as intense as it was above. This increase can be attributed to the (alpha; alpha¹ gamma) reaction. During the reduction process in hood 10 the fluorine is removed to leave pure metal; this process is monitored with a geiger counter and the counting rate, due to high energy component only, drops by a factor of six when the reduction is complete.

Figure 4 shows curves taken at the two above positions but through the steel walls of the hoods. These walls are quite effective in removing the low energy component. What remains of it in these measurements may very well be scattered radiation that has passed through the lucite.

Figure 5 shows curves taken through the lucite at hood 22 both with and without a shape in the hood. The same energy components as those encountered previously are present. It will be noticed that the relative number of counts with and without the shape present is not the same for the different energy components. This is due to the fact that for the low energy component both the shape and the contamination are thick sources while this is not true for the higher energies. As the thickness of a source is increased, the gamma intensity will increase until the source is about one mean free path thick after which the increase in gamma intensity for that energy will be small.

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Figure 6 shows curves taken through the lucite window from a shape wrapped in a surgical glove at hood #27. No low energy components are visible. At this point in the process the shapes have been plated with nickel. The thickness of nickel is sufficient to stop all of the lower energy components.

In early 1950 one of us (TCW) prepared some step wedges of different materials to be used with X ray film to determine the energies present by the absorption method. These were exposed by H. I. Operations personnel and interpreted by us. Confusion resulted and the best use was not made of the method. These data have been re examined and found to agree roughly with the counter data reported above. The following table summarizes the energy determinations made from those films:

Energy Measurements at 234-5 Using Film

Energy Component	Hood 8 Through Lucite	Hood 8 Through Steel	Hood 9 Through Lucite	Hood 22 Through Lucite	Shape
Low	22 Kev	18 Kev	N	U	Indicated
Medium	80-130	80-100	100* Kev	75-120 Kev	40 ? Kev
High	230	U	----	180	210

N not detected
U wedges of improper thickness for detection
* density too low for reliable measurement

Reference 5 gives two possibilities for the low energy gamma ray of plutonium, 200 Kev and 50 Kev. The former was determined by absorption measurements with lead; the latter, by the same method using lead and aluminum. The discrepancy is possibly due to the fact that one is very close to the K absorption limit in lead for this gamma ray and accurate interpretation of results is difficult. Where aluminum was used this difficulty is avoided; this gave the 50 Kev result. Our measurements of this component with the counter were based on copper absorption which should also eliminate the difficulty; our result was 40 Kev. The film measurements were made with lead and gold and encounter the same uncertainty.

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Conclusion

Hanford gamma measuring devices are calibrated at much higher energies than the 17 Kev of considerable interest here. We are proceeding with a check of the calibration at this energy.

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J. S. Reddie
J. S. Reddie

E. C. Watson
E. C. Watson

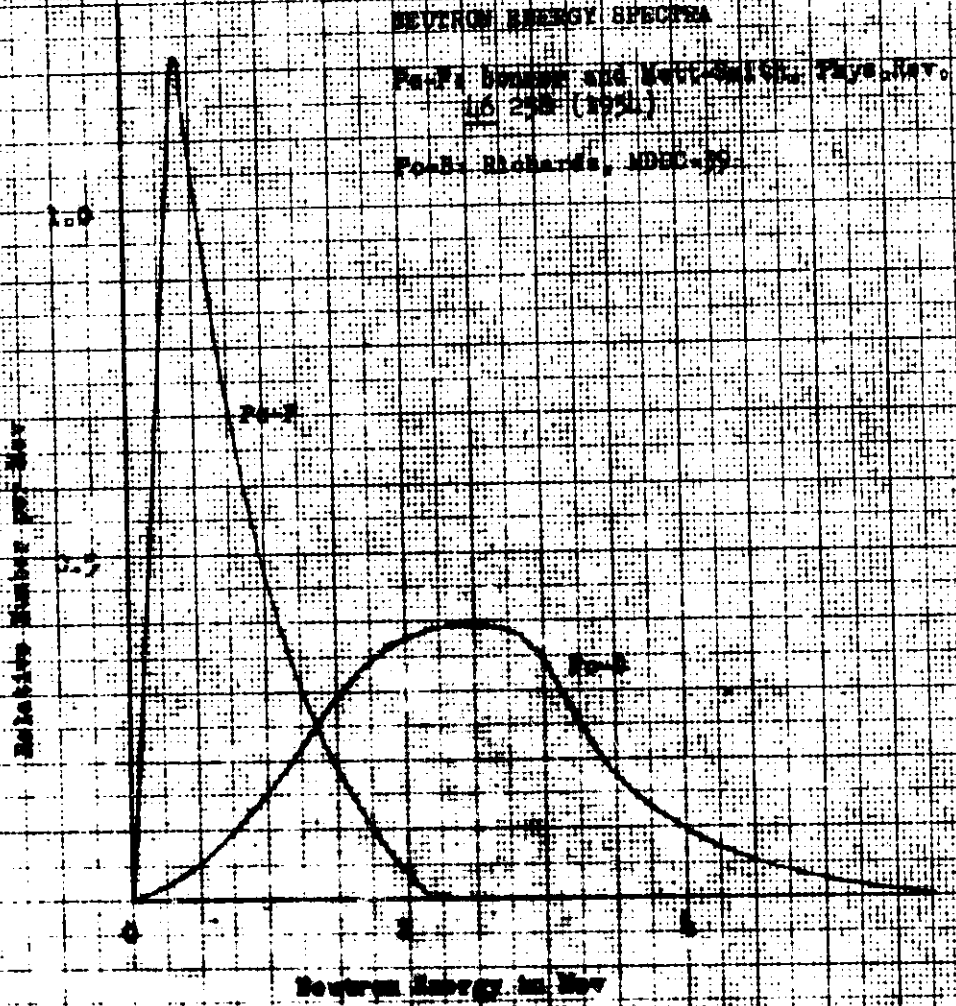
References:

1. Livingstone, M.S. and Bethe, H.A., "Nuclear Physics," Rev. Mod. Phys. 9 303 (1937).
2. Memo, "Neutron Measurements in the 234-5 Process," G. H. Whipple to A. R. Keene, no date. Whipple, G. H. and Reddie, J. S., "Fast Neutron Measurements - 234 5 Building," Sept. 15, 1949, HW-14440, Secret.
3. Barton, G. W., Robinson, H. P., and Perlman, I., "The L X-ray Spectra from Radioactive Decay of Transuranium Elements," Phys. Rev. 81 208 (1951).
4. Bothe, W. and Franz, H., "Untersuchungen uber die durch Alpha-Strahlen erregtes Rontgenstrahlung," Zeits. f Physik 52 466 (1929).
5. Seaborg, G. T., and Perlman, I., "Table of Isotopes," Rev. Mod. Phys. 20 585 667 (1948).
6. Speh, K. C., "Gamma Rays of Lithium and Fluorine under Alpha Particle Bombardment," Phys. Rev. 50 689 (1936).

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Figure 1



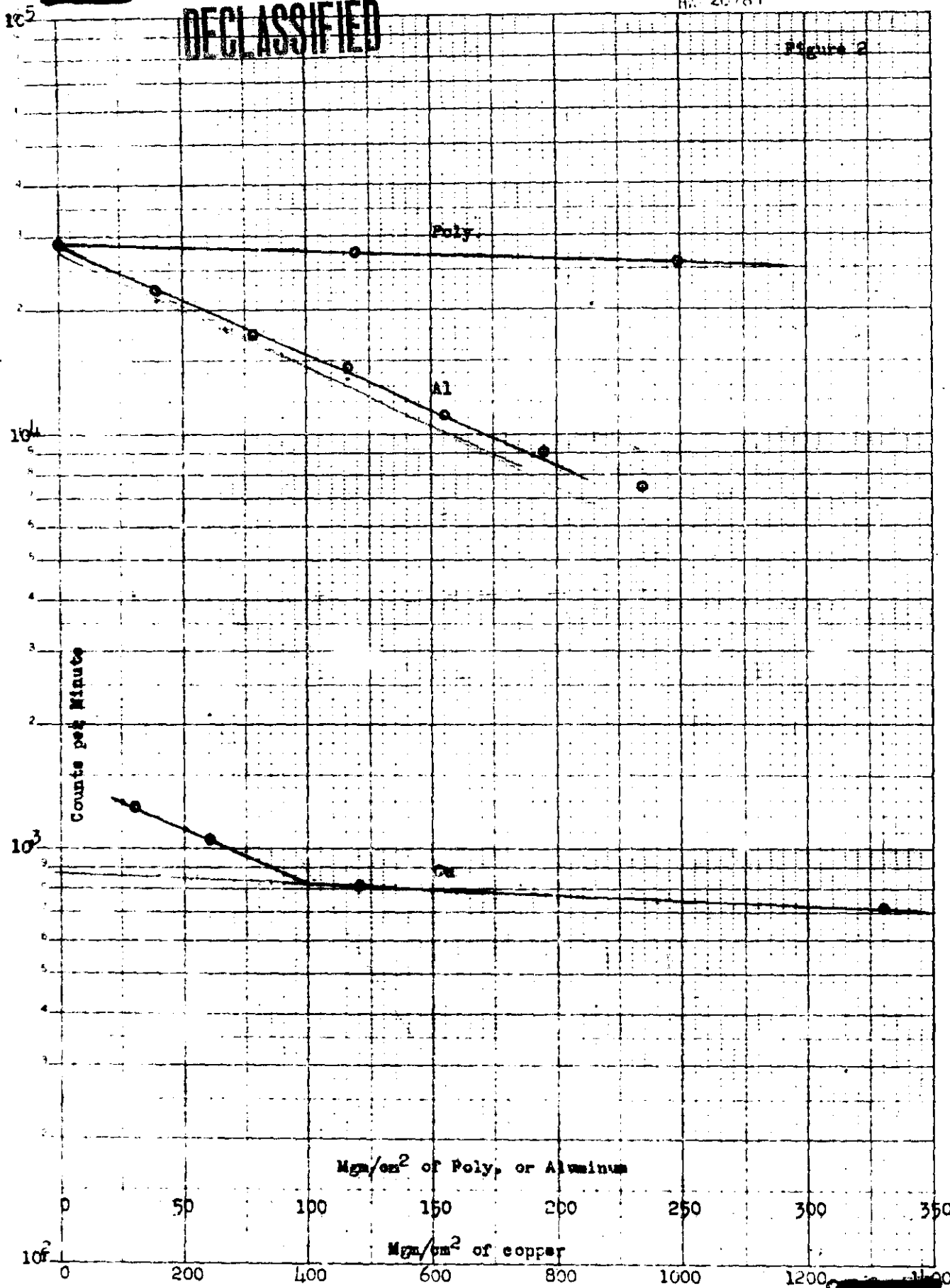
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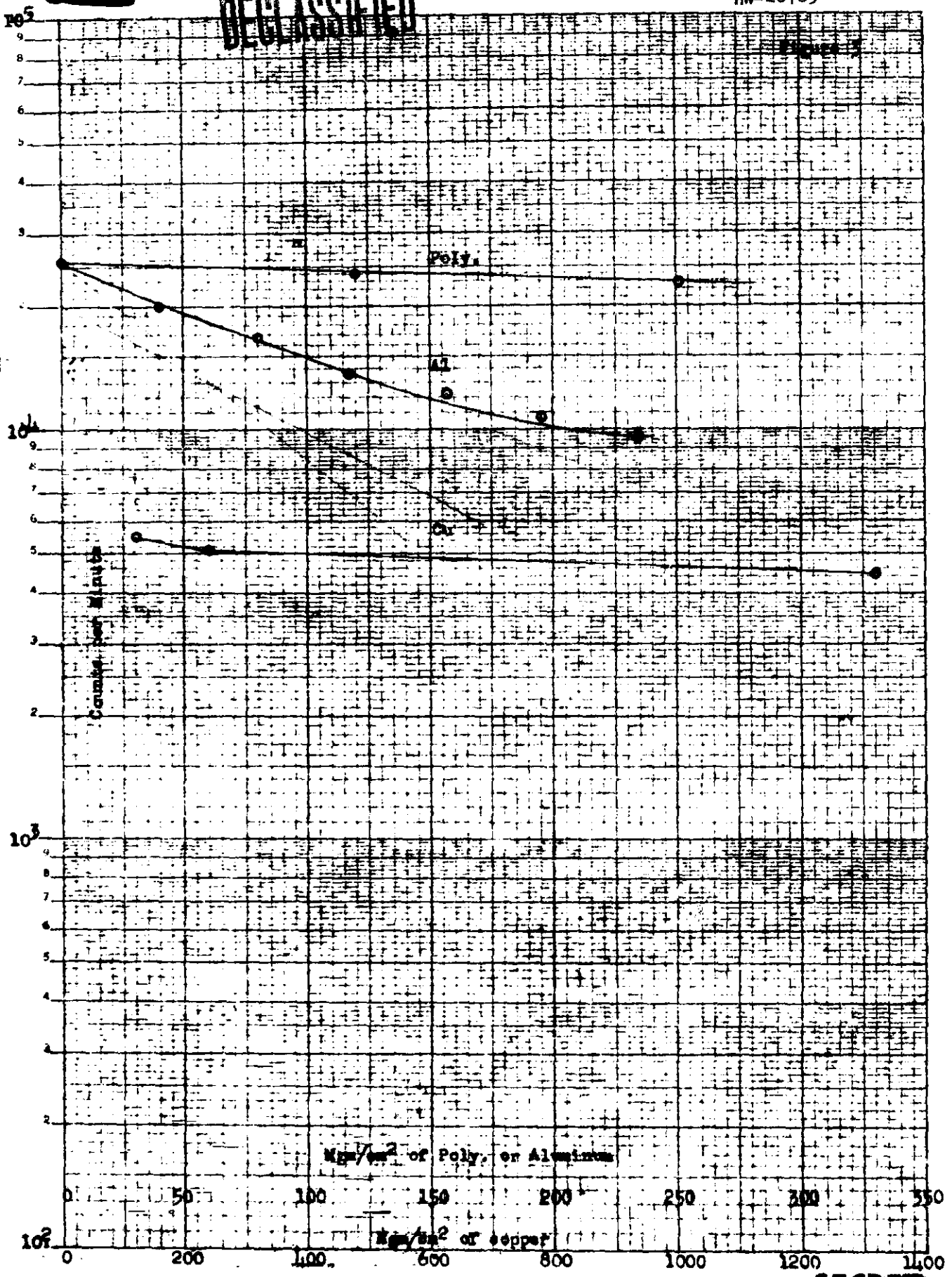
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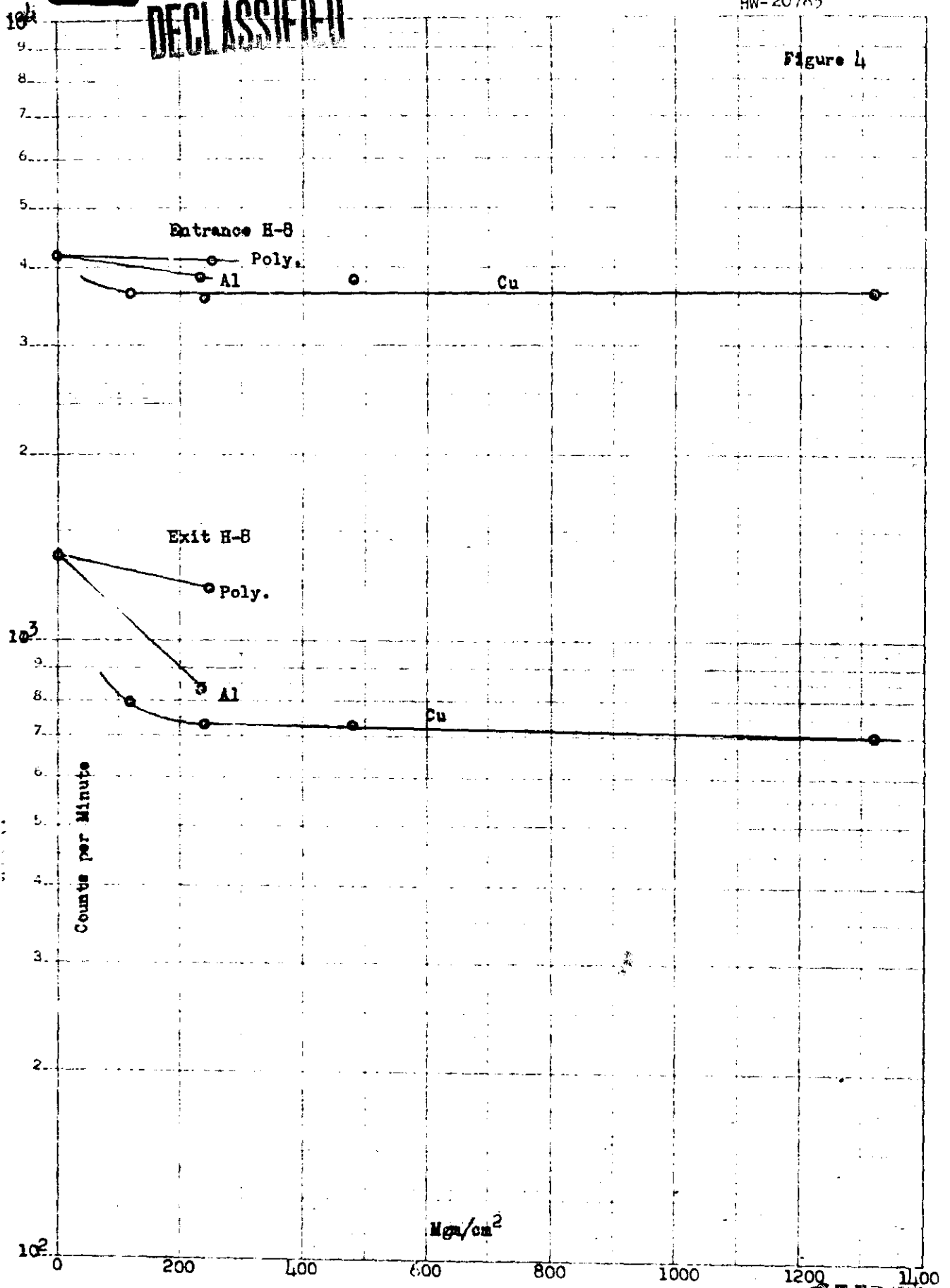


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Figure 4



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S. D. Long...
New York, N. Y.

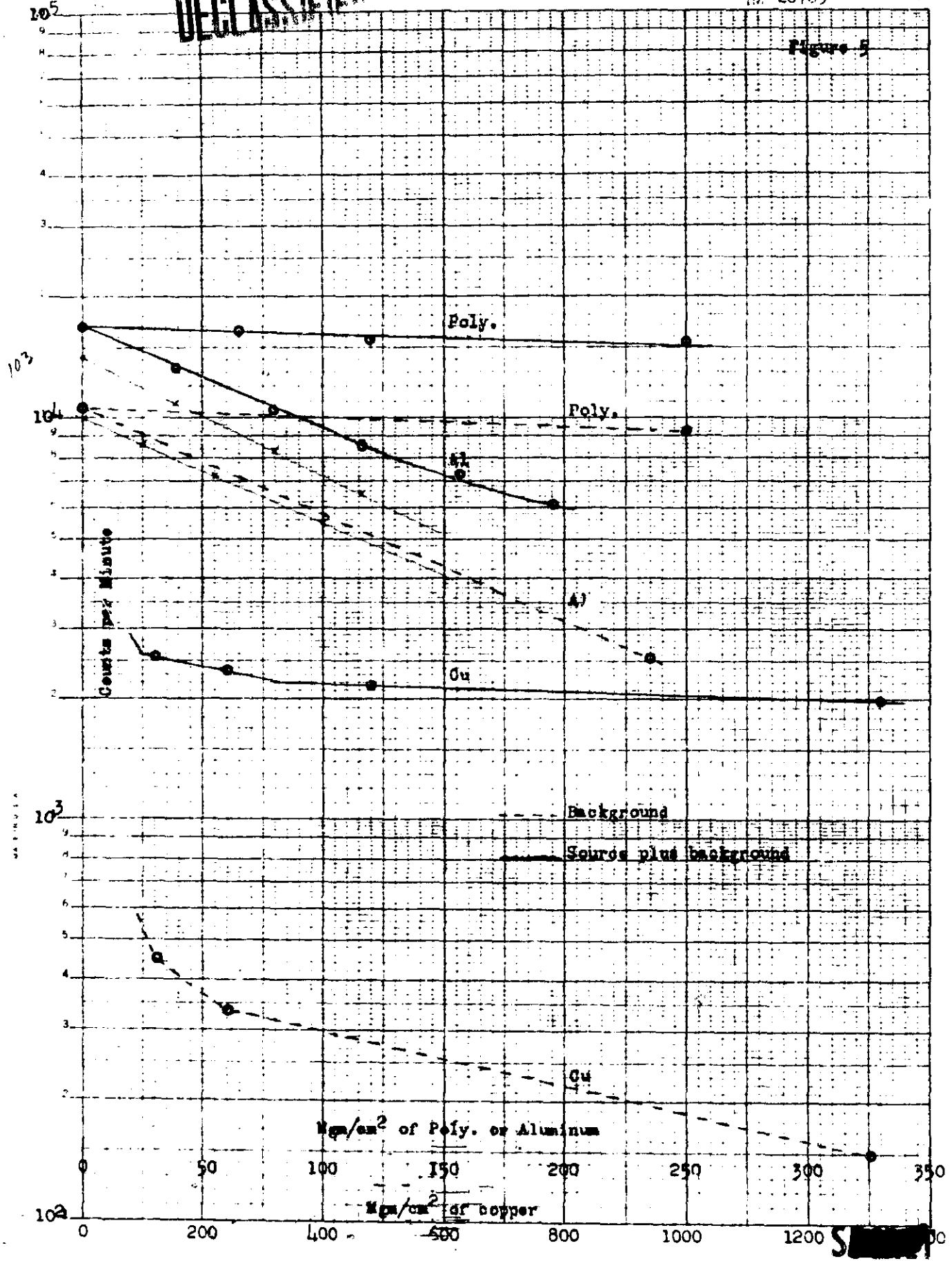
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Figure 5



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Figure 4

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Counts per Minute

- ◆ Polyethylene
- △ Aluminum
- Copper

10³

Mg/cm²

10²

0 200 400 600 800 1000 1200

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KODAK SAFETY FILM CO. N. Y. NO. 33021
1110 1/2" x 8 1/2" for the use of 35 film camera
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