NYO-702
AT(30-1)-655

NEUTRON-DEFICIENT MERCURY ISOTOPES*

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ABSTRACT

High energy bombardments with protons on gold and alpha-particles on platinum have been performed to investigate neutron-deficient mercury isotopes. These were separated from the targets both by liquid microextraction techniques and by a carrier-free vaporization method. Two new mercury isotopes have thus been definitely identified as follows:

Hg^{192}, $T_{\frac{1}{2}}$ = 5.7 ± 0.5 hours, 1.18 Mev β^+, 0.18 Mev conversion electrons, K, L X-rays, and 1.39 Mev Gamma-ray. Parent of 4.0-hour Au^{192}.

Hg^{193}, $T_{\frac{1}{2}}$ = 10.0 ± 0.5 hours, No β^+, K,L X-rays, 0.18 Mev conversion electrons. Parent of 15.3-hour Au^{193}.

Tentatively identified was Hg^{195} of 31-hour half-life, but this value may be in error owing to considerable masking from the 24-hour, 65-hour isomeric pair at Hg^{197} which is present in all bombardments. Decay of 31-hour Hg^{195} gives rise to a gold daughter of 180 days half-life, ~~and~~ although this ~~long-lived gold was observed as a daughter product, the~~ genetic relationship has not yet been established quantitatively. No positrons are emitted by 31-hour Hg^{195}, but K-, L X-rays and conversion electrons are observed.

No activity was observed which can be allocated to Hg^{194}, and it is possible that Hg^{194} is beta-stable. No evidence of 39.5-hour, positron-emitting gold daughter was observed among the gold daughter products.

In proton bombardments of gold from 65 to 96 Mev, an unidentified 2.0-hour mercury activity was observed. This must have mass number 191 or lower.

Finally, the genetic relationships between Hg^{192}--Au^{192} and between Hg^{193}--Au^{193} have been quantitatively established.

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*This investigation was performed under contracts with the U.S.Atomic Energy Commission, and was carried out partly at the University of California Radiation Laboratory, Berkeley, California, and partly at the Department of Chemistry, University of Rochester, Rochester, New York.

NEUTRON-DEFICIENT ISOTOPES OF MERCURY

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Neutron-deficient isotopes of mercury have been investigated in the Berkeley 184-inch cyclotron and in the Rochester 130-inch cyclotron.

Irradiations of platinum with 65 Mev alpha-particles for three hours on the Berkeley 184-inch cyclotron were performed, as well as with 30 to 60 Mev protons on gold for periods of from one to three hours. Bombardments of gold with 55, 65, and 96 Mev protons were made in the Rochester 130-inch cyclotron.

A 0.005-inch thick platinum target, irradiated with 65 Mev alpha-particles was dissolved in hot aqua regia, 10 mg each of gold and mercury carriers added, and after evaporation to expell HNO_3 and dilution to 6N HCl concentration, the solution# extracted with five successive portions of isoamyl acetate saturated with HCl just before use. The aqueous layer remaining was treated with excess $SnCl_2$ and the resulting precipitate of Hg_2Cl_2-Hg^0 washed free of the red platinous chloride color and until the washings exhibited little residual radioactivity. The precipitate was then dissolved in dilute aqua regia and utilized as the mercury fraction. From this bombardment the mercury half-lives observed were 5.5 hours, 9.6 hours, and considerable 24-hour, 64-hour activity due to the isomeric pair at Hg^{197} which is well studied[1].

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[1] K. Way, L. Fano, M.R. Scott, and K. Thew, National Bureau of Standards Circular 499 January 1, (1950).

No alpha-emission was detected from these mercury activities.

A Berkeley bombardment of 0.003-inch gold foil with 30 Mev protons for one hour gave a mercury activity of 31 hours half-life plus a long-lived activity of several months duration (probably 180 day Au^{195} daughter) as shown in Figure 1. This long-lived tail was subjected to repurification experiments which demonstrated chemically that it was gold activity and not mercury. It is thus rather certain that the long-lived tail is indeed 180-day Au^{195} reported in the literature[2,3].

[2]G.W. Wilkinson, Phys.Rev., 75, 1019, (1949).
[3]R.M. Steffen, O. Huber, and F. Humbel, Helv.Phys.Acta, 22, 167, (1949).

A Rochester bombardment with 50-55 Mev protons on 0.003-inch gold foil, of 99.999% spectroscopic purity obtained from Johnson-Matthey & Co.Ltd. (London), showed mercury half-lives of 10.0 hours and the 24-hour, 64-hour isomeric pair at Hg^{197}, as illustrated in Figure 2. In this and all other gold bombardments, mercury was separated from the gold foil target by carrier-free volatilization. A portion of the gold target was placed in the chamber of a stainless steel vaporizer. A clean platinum collecting plate was cemented to a stainless steel cold-finger which was cooled with flowing cold water. The bottom of the apparatus was heated with a Fisher burner to not more than about 400°C. Since the vapor pressure of pure mercury is 1574.1 mm of Hg at 400°C., of pure molten gold at 1292°C. is only 0.001 mm, and of pure thallium at 413°C. is only 0.001 mm, this volatilization procedure affords a mercury separation which is quick, carrier-free, and excellent with respect to purity. Gross decay curves, Figure 2, of the vaporized mercury fraction were always identical with those from the chemically-isolated aqueous mercury fraction.

As the energy of proton bombardment is increased to 65 Mev or greater, a new 5.7-hour mercury activity appears, Figure 5, which has a 4.0-hour gold daughter. These do not appear at lower bombardment energies. Through identification of 4.0-hour Au^{192} daughter, the 5.7-hour mercury is shown to have mass number 192. With 96 Mev protons a greatly increased yield of 5.7-hour activity relative to 10.0-hour mercury is observed and a new 2.0-hour mercury activity appears in high yield. The assignments and genetic relationships are discussed below.

MERCURY 195

The 31-hour activity from 30 Mev proton bombardment of gold, Figure 1, is assigned to Hg^{195}, although the half-life value may be in error by as much as 20% owing to considerable masking by the 24-hour, 64-hour isomeric pair at Hg^{197}. A long-lived gold daughter activity is observed which is presumed to be Au^{195}.

But the genetic parent-daughter relationship has not been quantitatively proved. No positrons are detected on a 4.0-cm magnetic spectrometer, but L X-rays of 10.5 Kev energy and conversion electrons of 0.19 Mev were observed in aluminum-beryllium absorption measurements.

MERCURY 194

Although the interesting nucleus Hg^{194} was certainly produced in these bombardments, no activities were observed which can be allocated to it, nor was its 39.5-hour, positron-emitting Au^{194} daughter ever observed from it. These findings are consistent with the statement that Hg^{194} is probably stable.

MERCURY 193

A 10.0± 0.5-hour mercury activity which is produced ~~as~~ with 55 Mev, but not with 30 Mev, protons and which diminishes in yield as bombardment energy is increased to 96 Mev, is assigned to Hg^{193}. The quantitative relationship between 10-hour mercury and 15.3-hour Au^{193} daughter[2,3] ~~is shown in Figure 3,~~ has been demonstrated. ~~Curve B.~~ Parent-daughter separations were performed by extracting the 6N HCl aqueous mercury fraction to which 6 mg gold carrier was added with isoamyl, or in some experiments with normal-amyl, acetate saturated with HCl just before extraction, and then stripping the organic layer with 1N NH_4Cl to remove any traces of mercury isotopes from the gold daughter fraction.. The validity of this separation was studied with carriers and found to be of excellent quality. Light from a carbon arc ~~has been~~ was found not to reduce $HAuCl_4$ photochemically in presence of mercury, so that this procedure affords a clean separation of mercury and gold. Isoamyl acetate is superior to ethyl acetate in microextractions because the latter is ~~considerably~~ somewhat soluble in water. Use of a small rheostated stirring motor fitted with flexible micro-stirring rods fashioned out of thermoplastic Kel-F (poly-trifluorochloroethylene) quarter-inch rod greatly facilitates microextractions on quantities as small as 0.030 ml. Curve B of Figure 3 shows the decrease in yield of 15.3-hour Au^{193} daughter from the mercury fraction, the slope being 10.6 ± 0.5 hours, in agreement with the 10.0 ± 0.5 —

hour half-life found for the parent, Hg^{193}. The parent emits no observable positrons, but manifests 9.6 hour K and L X-rays as followed on an X-ray counter, fitted with a permanent alnico magget of 1600 gauss and xenon-filled Geiger tube of such geometry that all particles of energy less than 2.4 Mev would be bent away and not counted. In addition, the sample was covered with 195 mg/cm^2 of beryllium and surrounded with lead housing. A 4.0-cm magnetic spectrometer confirmed the presence of 0.18 Mev conversion electrons in agreement with aluminum-beryllium absorption curves. Gross decay of a typical gold daughter from this 50-55 Mev proton bombardment is shown in Figure 4, where the half-life observed is 15.3 ±0.5 hours for Au^{193}. The second component in Figure 4 tails out to long-lived gold, presumably 180-day Au^{195}.

MERCURY 192

With protons of ~~from~~ 60 to 96 Mev on 0.003-inch gold foil in the Rochester cyclotron, a new mercury activity of half-life 5.7±0.5 hours ~~was~~ observed, Figure 5, Curve C. The growth of 4.0 hour Au^{192} daughter also manifests itself in Figure 5, Curve A, where the observed appearance of maximum growth occurs at 6.5±0.5 hours from time of separation, whereas the calculated time of maximum growth ~~occurs at~~ is 6.8 hours, assuming a 5.7-hour half-life for Hg^{192} parent and 4.0-hour half-life for Au^{192} daughter. This parent-daughter genetic relation is shown even more strikingly in Figure 3, Curve A, where a decrease in yield of Au^{192}, extrapolated back to instant of isolation, approaches 5.6 hours, half-life in excellent agreement with the half-life of 5.7 hours for the parent. Curve D, Figure 5, represents the small amount of 10.0-hour Hg^{193} formed at 96 Mev proton bombardment, and curves E and F represent the longer-lived 24-hour, 64-hour isomeric pair at Hg^{197}, present in all bombardments. Curve B is an unidentified new 2.0-hour mercury activity, probably of mass number 191 or less. Hg^{192} emits positrons of 1.18 Mev maximum energy detected on a 4.0-cm magnetic spectrometer. These positrons decayed with a half-life of 6 hours. The maximum positron energy was measured by aluminum absorption curves as well and agrees with the spectrometer results. Thus, Hg^{192} is the highest atomic-numbered ~~isotope~~ nuclide to emit positrons

known to date. Lead absorption curves show a 1.39 Mev gamma-ray in addition to 0.5 Mev annhiliation radiation associated with Hg^{192}. The decay curve of the gold daughter fraction isolated from the mercury fraction by isoamyl acetate extraction and followed by a scintillation counter fitted with a NaI(Tl) crystal and RCA 5819 photomultiplier tube is shown in Figure 6. A half-life of 4.13 ± 0.10 hours for Au^{192} is thus obtained.

OTHER ACTIVITIES OBSERVED

In the vaporized mercury fraction from 96 Mev proton bombardment of gold, an unidentified 2.0-hour activity occurs in high yield, Figure 5, Curve B. This yield increases relative to other activities present as the energy of bombardment increases from 65 to 96 Mev, indicating that this activity might belong to a mercury isotope of mass number 191 or less. It could hardly be Tl^{198} on the basis of the volatilization procedure used. The gold fraction from 60 Mev proton bombardments showed the presence of 13.8-hour Au^{196m} and 5.3-day Au^{196} as expected from a (p,pn) reaction. The platinum fraction shows a 4.3-day activity, Pt^{193}, and a 7.0-hour activity which is so far unidentified.

~~We have recently learned of the work by J.H.~~ Moon and ~~[illegible]~~ Thompson[4] ~~et~~ have recently reported results as follows: Hg^{195}, 38 hours; Hg^{194}, not found; Hg^{193}, 14.5 hours and 29.0 hours; Hg^{192}, 8.4 hours; and Hg^{191}, 12.4 hours; all ~~of~~ which are K-capturing isotopes. We feel that the 8.4-hour activity reported for Hg^{192} may be a mixture of 5.7-hour Hg^{192} and 10.0-hour Hg^{193}, and that the 14.5-hour activity reported for Hg^{193} is in error possibly due to masking by Hg^{197}, which may also account for the 29-hour activity reported, or to presence of 15-hour gold daughter in the mercury fraction. The 38-hour value reported for Hg^{195} differs from our 31-hour value, but owing to considerable masking by Hg^{197}, this is not outside of our experimental error. The 12.4-hour activity reported for Hg^{191} leading to an 18-hour Au^{191} daughter is felt to be in reality mass number 193, with the half-lives appearing longer due to presence of Hg^{197} and Hg^{195} in the

[4] Bulletin Am. Phys. Soc., 26, #5, 12 (1951).
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mercury fraction and presence of Au^{196} in the gold daughter fraction. The chemical procedures used by Moon and Thompson are not given in the brief report cited.

ACKNOWLEDGEMENTS

We are indepted to Dr. David H. Templeton of the Radiation Laboratory, University of California, Berkeley, for valuable suggestions and criticisms; to James T. Vale and the crew of the Berkeley 184-inch cyclotron; to Mr. James Norton of the Rochester cyclotron for his assistance in design of cyclotron probes; and to Dr. Sidney W. Barnes and the crew of the Rochester 130-inch cyclotron for their assistance in bombardments.

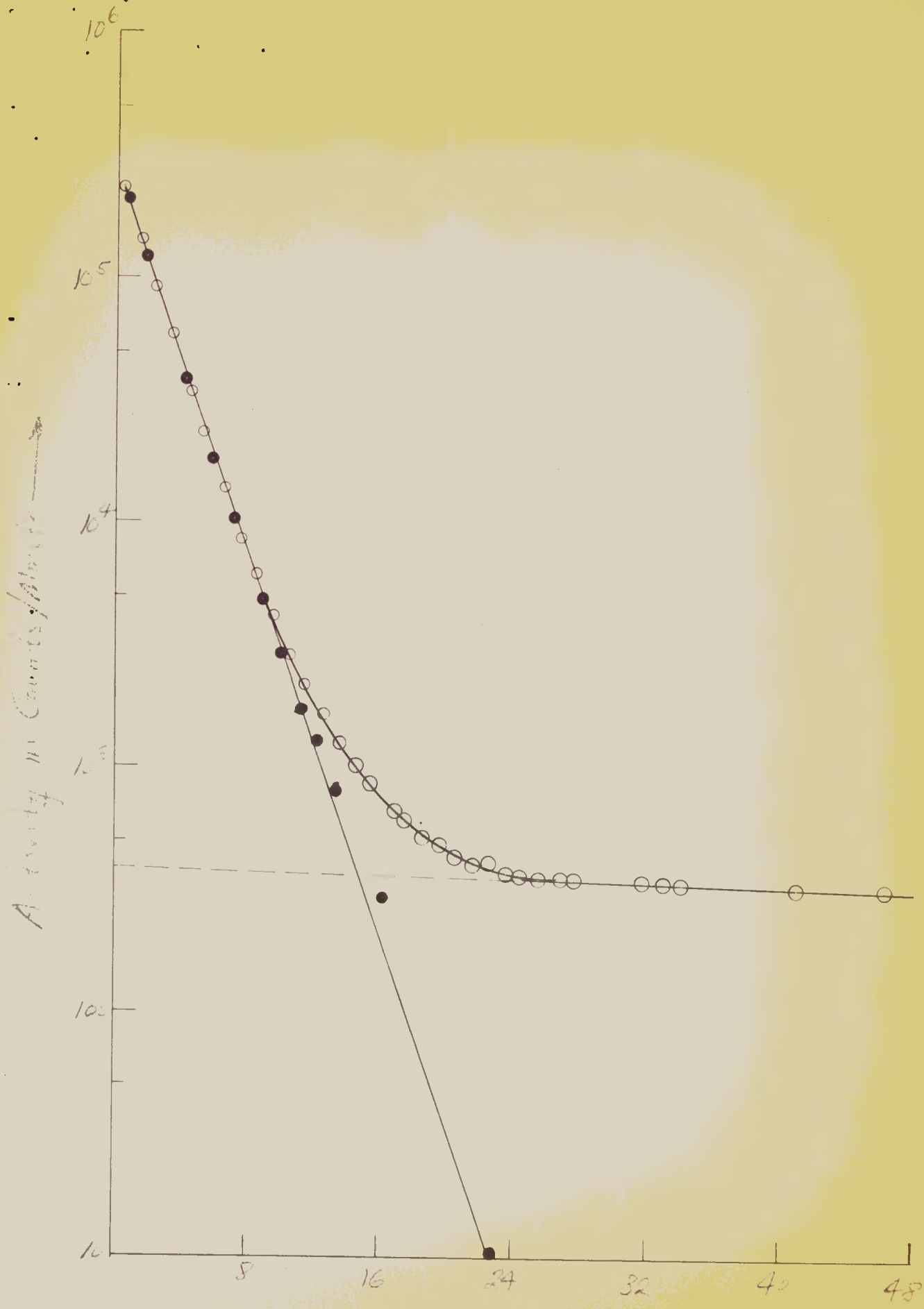

Fig. 1 — Hg^{195} (31-hr.) and Au^{195} (180-day) in Hg fraction.

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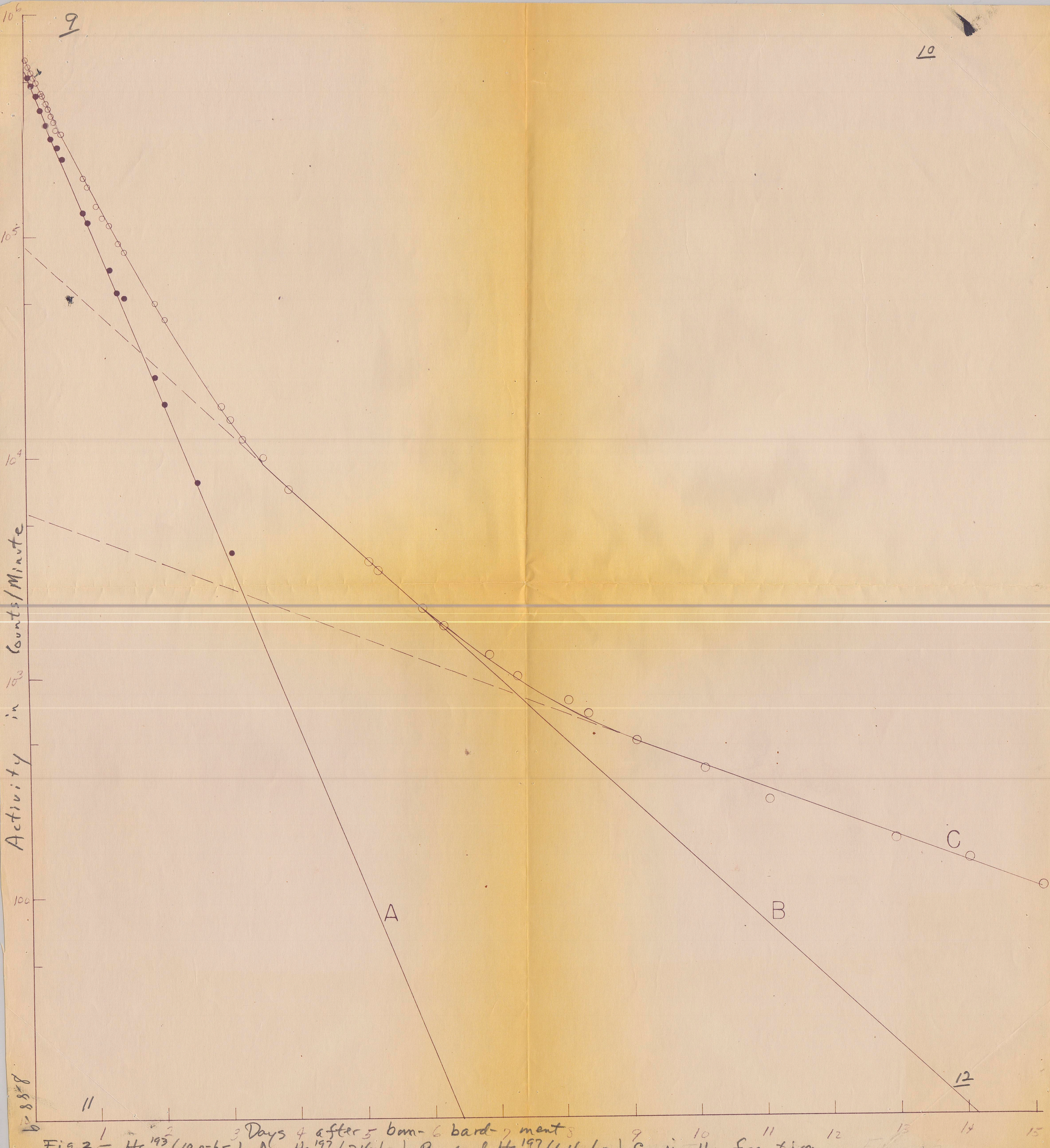

Fig. 2 — Hg^{193} (10.0-hr.), A, Hg^{197} (24-hr.), B, and Hg^{197} (64-hr.), C, in Hg fraction.

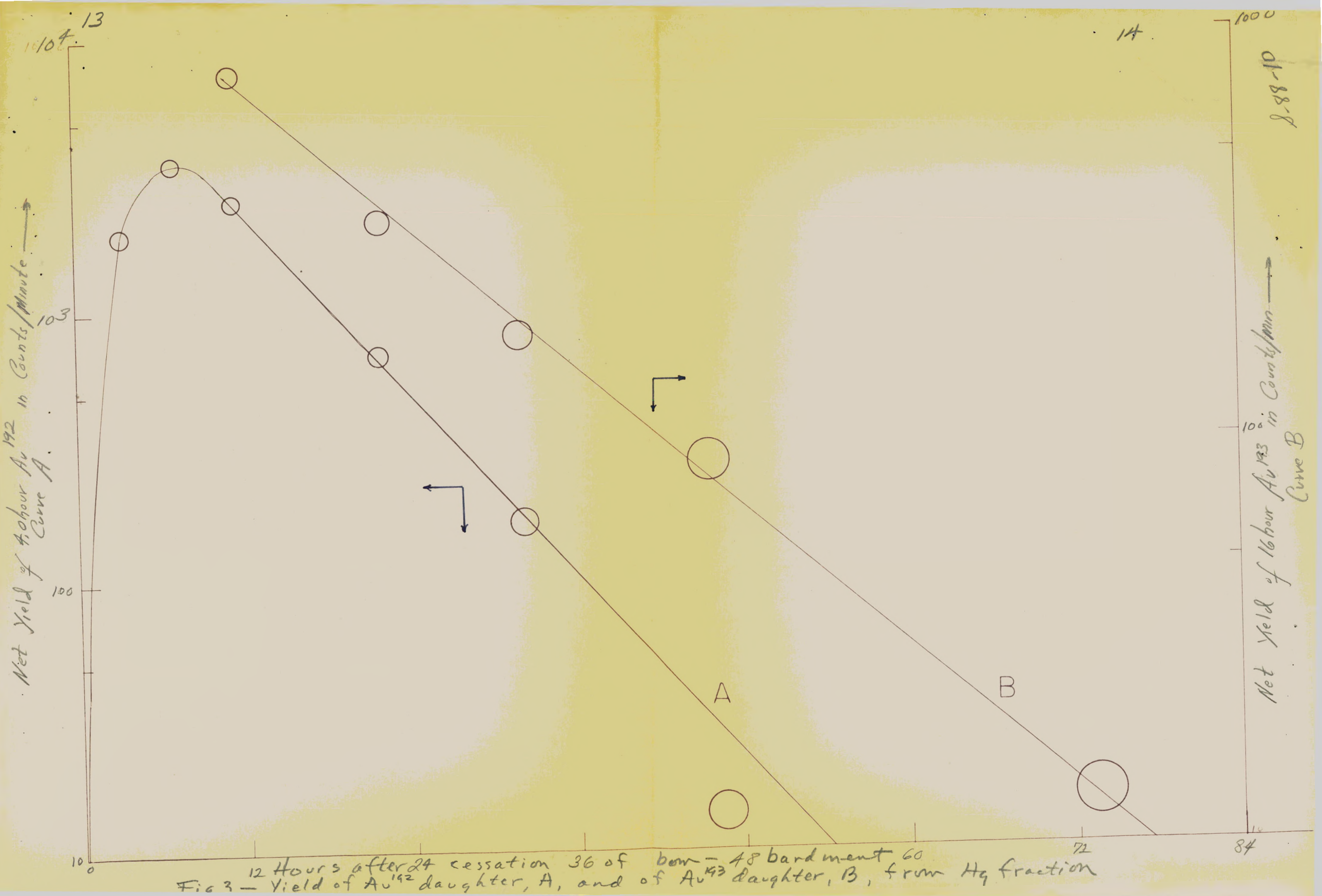

Fig 3 — Yield of Au^{192} daughter, A, and of Au^{193} daughter, B, from Hg fraction

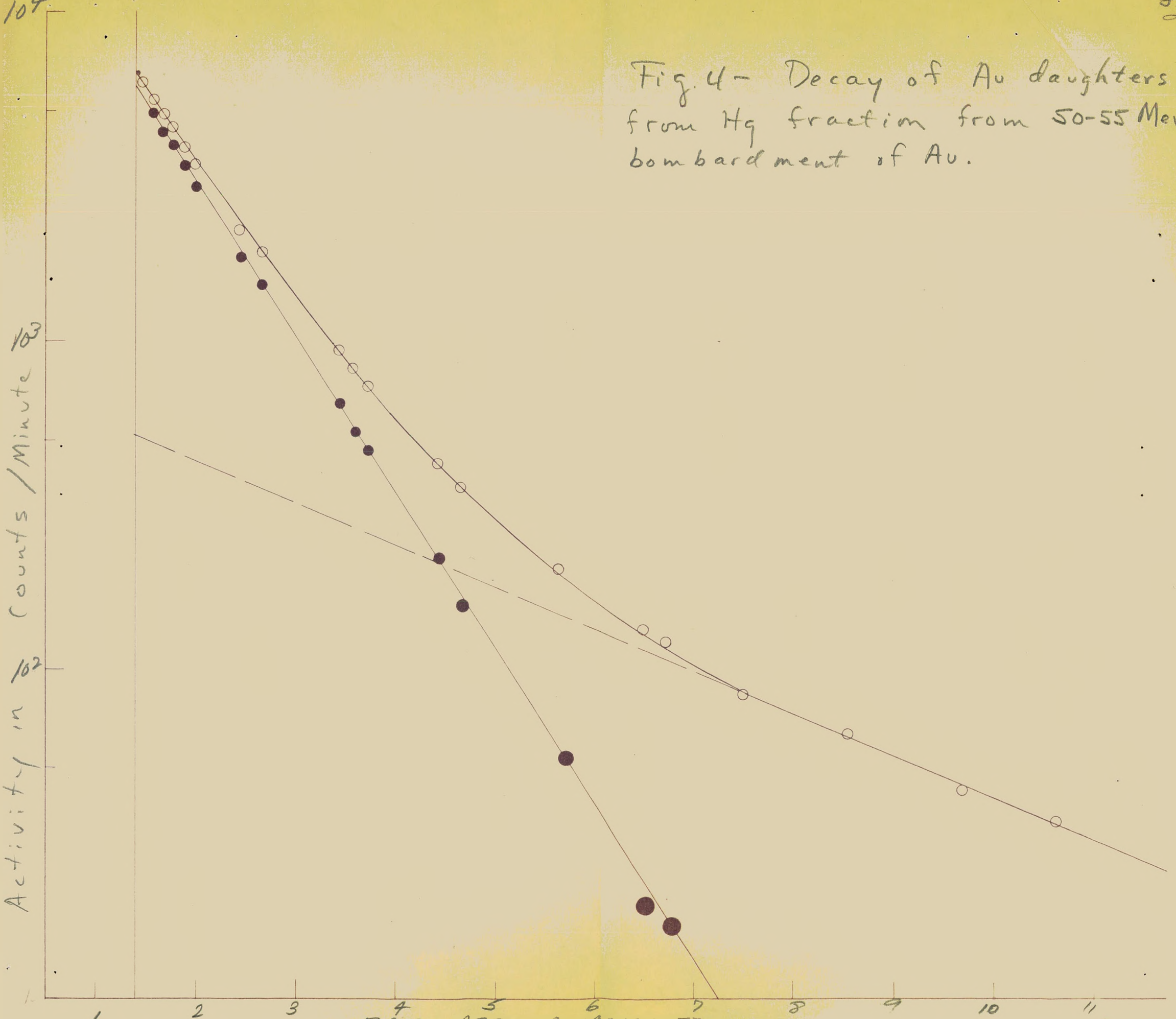

Fig. 4- Decay of Au daughters from Hg fraction from 50-55 Mev bombardment of Au.