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IDENTIFICATION OF RADIOACTIVITY IN SPECIAL SAMPLES

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Telephone conversation between Mr. Robert O. Ryan, AFOAT-1 and
Dr. R. W. Spence, Los Alamos Scientific Laboratory, 18 October 1949

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Identification of Radioactivity in Special Samples

A sample of filter paper (Type 5) was sent to us on 10 September 1949 by Dr. William D. Urry of AFOAT-1 with the request that we make an independent analysis of the sample for fission product activity. The paper sent to us was labelled: Station No. 70265; Paper No. AG370. The wrap-around net count was 2401 c/m, and the sample was collected on 7 September 1949 at 0240 GMT.

The sample was received by us about 0100 MST, 12 September 1949, and was dissolved that night using our regular procedure for Type 5 filter paper. (This procedure was developed for Sandstone samples.) Analyses for molybdenum, zirconium, silver, barium, cerium, and lead were carried out, using analytical procedures which we have adopted as standard. The only variation from our usual procedure was that instead of taking separate aliquots for each analysis we were forced, because of the low activity, to carry out all analyses on the same aliquot. The resulting changes in chemical procedure might affect quantitative results, but could have no effect on qualitative analyses. In any case we could not supply quantitative results without a great deal of additional work, since we had to set up special low background counters (which were uncalibrated) in order to measure the low activities we expected to get.

The sample solution was divided into two portions and analyses carried out on both portions. Two blank analyses were also carried out for each element to check on background and accidental contamination. The most significant results were obtained from the barium and cerium analyses. The initial activities of the two barium samples were 261 c/m and 301 c/m.

(No correction was made for chemical yield, since fresh carrier solutions were used, and these carrier solutions were not standardized.) The barium activities were followed with time, and the activities grew, then decayed, (See Fig. 1) just as one would expect if the activity were due to 12.8 day Ba^{140} with the daughter 40 hour La^{140} growing in. The final half-life was about 13 days, in excellent agreement with the above activity assignment. The two blank barium analyses showed no activity (less than 5 c/m above background).

The two initial cerium activities were 86 c/m and 83 c/m. An absorption curve on one of the cerium samples showed the presence of a hard and a soft beta component. (See Fig. 2) The hard component showed the general characteristics of Pr^{144}, the daughter of 275 day Ce^{144}. The softer component had a half-thickness of $\sim$ 12 mg/cm^2, in excellent agreement with the assignment 28 day Ce^{141}. The cerium activity therefore, could be definitely attributed to a mixture of Ce^{141} and Ce^{144}, both well-known fission products. Blank cerium activities were practically at background.

Molybdenum activities were low, 28 c/m and 23 c/m for the two aliquots. These activities decayed roughly with a 67 hour half-life, corresponding to 67 hour Mo^{99}, the chief molybdenum activity from fission product.

Zirconium and silver activities were low ($\sim$ 10 c/m) and the most that could be said of them was that their half-lives were fairly long; this result is not inconsistent with the results expected from 65 day Zr^{95} and 6.8 day Ag^{111}.

Lead activities were very close to background, indicating that the fraction of the total activity coming from natural radioactive sources was very small.

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A second filter paper (R1, 0392 AEB) was later supplied to us by Dr. Lloyd Zumwalt of the Western Division of Tracerlab, Inc. Analyses of this paper gave essentially the same results as the first sample, although the activities were lower. For example, the initial barium activities were 101 and 107 c/m, and the initial cerium activities were 33 and 31 c/m. A neptunium analysis was run on this sample; the activity found was about 200 c/m. We do not trust this result, primarily because the analyst had prepared a very hot neptunium sample for Tracerlab, Inc. just a few days earlier, and probability of contamination was rather high.

Each of the above activities reported was the net count after the counter background of ~ 15 c/m had been subtracted.

The following conclusions can be drawn:

1. In view of the relatively high counting rates of barium and cerium (and low barium and cerium blanks) it can be stated definitely that the samples supplied to us contained cerium and barium radioactivity.

2. The growth and decay of the barium activity characterizes it beyond doubt as 12.8 day Ba^{140} with 40 hour La^{140} growing in.

3. The absorption curve obtained from the cerium activity characterizes it beyond doubt as a mixture of 28 day Ce^{141} and 275 day Ce^{144}.

4. The decay of the molybdenum activity makes it probable that it is due to 67 hour Mo^{99}; however the activity level was too low to make this absolutely certain.

5. The zirconium and silver activities found were too low to make any conclusion valid; their long half-lives are not inconsistent with 65 day Zr^{95} and 6.8 day Ag^{111}.

6. The low lead activities make it certain that only a small fraction of the total activity of the sample could be due to natural radioactivity.

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7. No accurate statement can be made concerning the age of the sample, beyond the statement that the sample could hardly have been more than about one month old, and was probably younger than one month. This conclusion is based upon the relative activities found for radioelements with half-lives varying from 67 hours to 275 days.

Our final conclusion is that the samples supplied to us contained radioactive isotopes and that the bulk of the activity was due to fission products of fairly recent origin, their age probably being one month or less.

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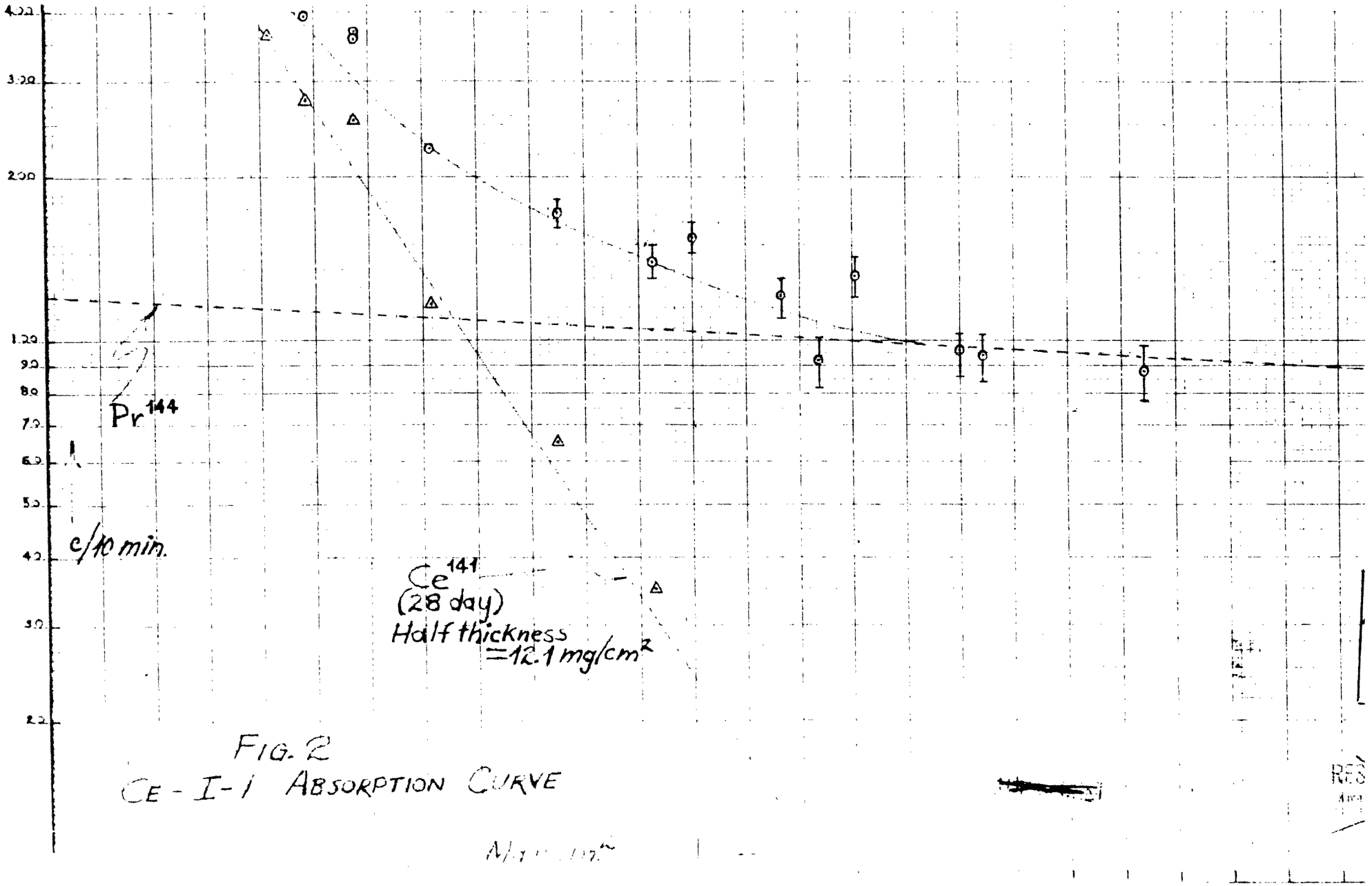

FIG. 2
CE - I - 1 ABSORPTION CURVE

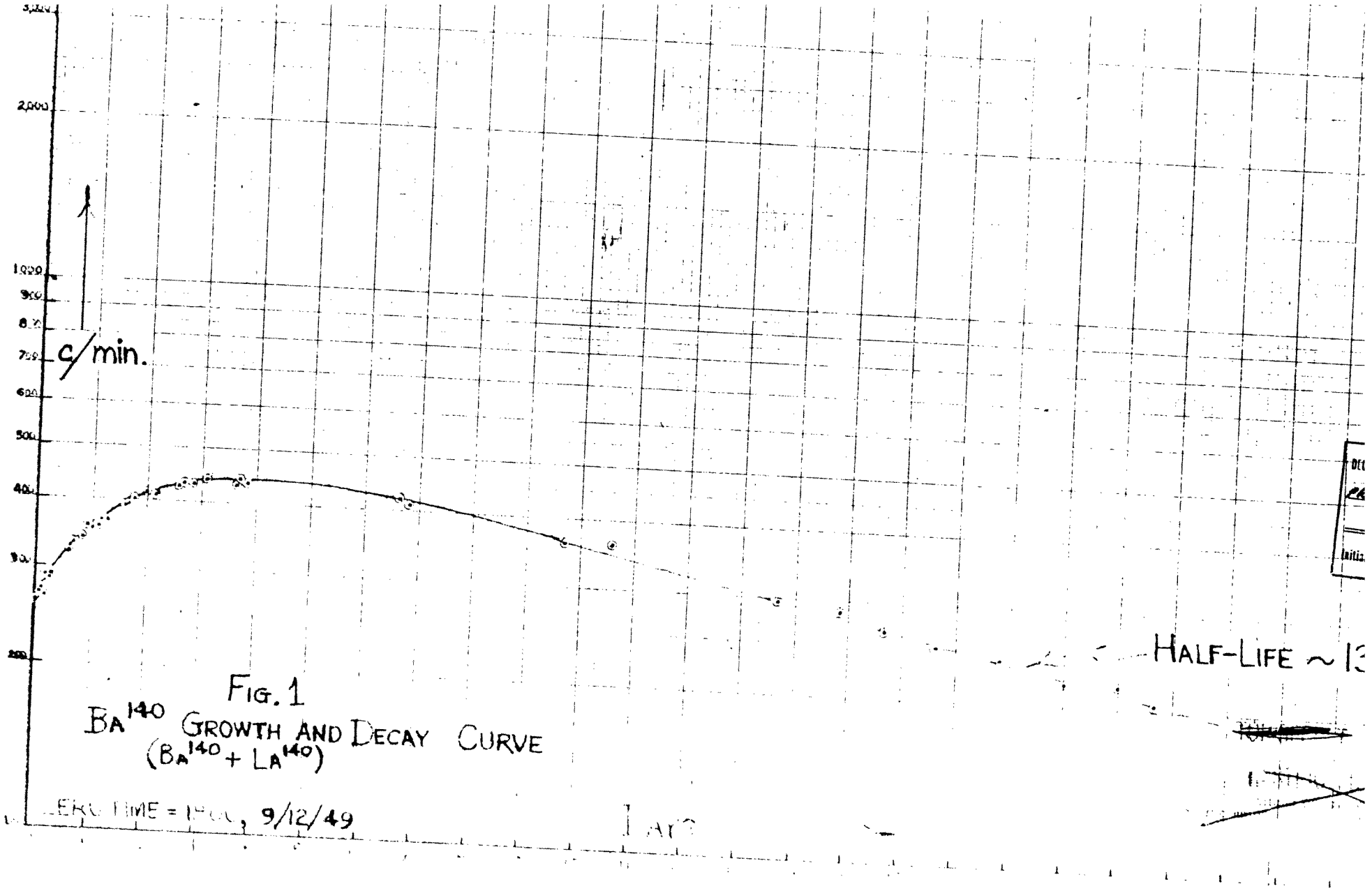

FIG. 1
Ba^{140} GROWTH AND DECAY CURVE
(Ba^{140} + La^{140})

CLOCK TIME = 1400, 9/12/49