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COBALT-60 FROM THERMONUCLEAR TESTS IN THE ATMOSPHERE

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COBALT-60 FROM THERMONUCLEAR TESTS IN THE ATMOSPHERE *)**)

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INTRODUCTION

We have detected the presence of Co^{60} in the atmosphere as it comes down with rain water. Its concentration is high enough so that we have been able to measure it fairly accurately. This is the third radioisotope near iron that has been found in rain water, the others, Co^{57} and Mn^{54} were found by MARQUEZ, COSTA and ALMEIDA (1958). For reasons to be discussed at the end of this paper, this Co^{60} must come from thermonuclear tests and possibly from tests of the so called "cobalt bomb" or "hydrogen-cobalt bomb".

EXPERIMENTAL METHOD

The rains that fell on the roof of our laboratory from June

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1957 until March 1958 were collected in several tanks of 1000 liter capacity and processed in several batches. The chemical procedure for processing was as follows:

For every 1000 liters of water, 20 mg of Co carrier were added and the water stirred thoroughly. The rain water was passed through a cation exchange column of 10 cm diameter and 80 cm high, filled with Amberlite LR-120 in the acid form. The cations were retained and the column was regenerated with 4 N HCl.

The 4 N HCl contained the Co and many inert impurities as well as radioactive impurities from other isotopes present in the rainwater. It was evaporated to a small volume, it was made basic with an excess of NH_4OH , and H_2S gas was passed through it. The precipitate was filtered, washed and dissolved in aqua regia, the HNO_3 of the aqua regia was destroyed by repeated boiling with HCl and then it was taken to dryness and kept for one hour at 120°C to dehydrate the silica. The residue was dissolved in HCl, diluted with water, filtered, and washed with dilute HCl. The total amount of HCl used was measured and the solution was made 1 N in HCl.

At this point, carriers were added of the elements La, Sr, Ba, Pb, Zr, Bi, Sb, Cu, Se, Te, Fe and Mn. The amount of carrier added was from 10 to 30 mg, according to the amount needed to make a good scavenging. The solution was saturated with oxalic acid, to precipitate the La. Na_2SO_4 also was added to precipitate the Pb, Ba and Sr. The precipitates were allowed to settle, filtered and washed.

The oxalic acid in the filtrate was completely destroyed by boiling with HNO_3 and then the solution was evaporated to a small volume. After adding H_2SO_4 , it was heated to white fumes to eliminate the HNO_3 . The solution was allowed to cool, diluted to 10% in H_2SO_4 and the Zr precipitated with NaH_2PO_4 . The precipitate was allowed to

settle, filtered and washed.

The filtrate was neutralized with NH_4OH and HCl was added to make it 0.3 N in HCl . H_2S was passed to the hot solution to precipitate the sulphides. These were filtered and washed.

The filtrate was boiled to eliminate the H_2S and bromine water was added and boiled. It was neutralized with NaOH and an excess was added to make it 2 N in NaOH . The precipitate was filtered and washed. It was dissolved in HCl , taken to dryness and dissolved in a mixture of 3 ml of HCl and 100 ml of H_2O .

The solution was passed through a cation exchange column to retain the cations and eliminate the anions. The column was washed with a very dilute solution of HCl and the washings rejected. The column was regenerated with 4 N HCl which contained the Co and other metals.

The 4 N HCl solution was evaporated to a small volume, neutralized with NaOH and an excess was added to make it 2 N in NaOH . The precipitate was filtered and washed. It was dissolved in HNO_3 and the MnO_2 was precipitated by boiling it with KClO_3 . The MnO_2 precipitate was centrifuged and washed.

The supernatant together with the washings were neutralized with NaOH . An excess was added to make it 2 N in NaOH . The precipitate was filtered and washed. It was then dissolved in HCl .

The HCl solution was treated with an excess of NH_4OH to precipitate the hydroxides and complex the Co. The precipitate was filtered and washed. The hydroxides were redissolved in HCl and reprecipitated with an excess of NH_4OH three times. All the filtrates were evaporated to dryness. The residue was dissolved in a mixture of 1 part of HCl and 4 parts of HNO_3 and boiled to destroy ammonium

salts. When all the ammonium salts were destroyed as indicated by a test with Nessler's reagent, the solution was evaporated to a small volume, boiled with HCl to eliminate the nitric acid and then it was taken to dryness.

The residue was dissolved in 5 ml of HCl and diluted to 200 ml and the Co was precipitated with a freshly prepared solution of 1-Nitroso-2-Naftol in acetic acid. The precipitate was allowed to settle, filtered, washed, dried, incinerated in a crucible and weighed as Co_3O_4 . This weight served to determine the chemical yield.

All the washings were done according to the standard prescriptions of Analytical Chemistry.

RESULTS AND DISCUSSION

The procedure described previously was applied to many rains that fell from June 1957 to March 1958. All the Co_3O_4 was transferred to a small test tube. After correcting for chemical yield, it represented the amount of Co in 12,100 liters of rain water. The actual amount of water processed was 29,200 liters, so that the average chemical yield was 41.5 %.

The sample was counted with a well crystal scintillation spectrometer described previously by MARQUEZ, COSTA and ALMEIDA (1957). We determined the line shape and the efficiency of the gamma rays in the region of interest using the gamma ray of K^{40} that has an energy of 1.46 Mev, and a standard source of Co^{60} . The spectrum given by the K is shown in Figure 1. The spectrum of the standard Co^{60} from the rain water is shown in Figure 3. As can be seen from the mere inspection of the figures, the activity of the Co^{60} from the rain water corresponds unmistakably to Co^{60} . The small difference be-

tween the shape of the Spectra in Figure 2 and Figure 3 comes from counting the standard Co^{60} outside the crystal and the sample inside the well. The energy measurement of the two peaks at 1.17 Mev and at 1.33 Mev do fit well too.

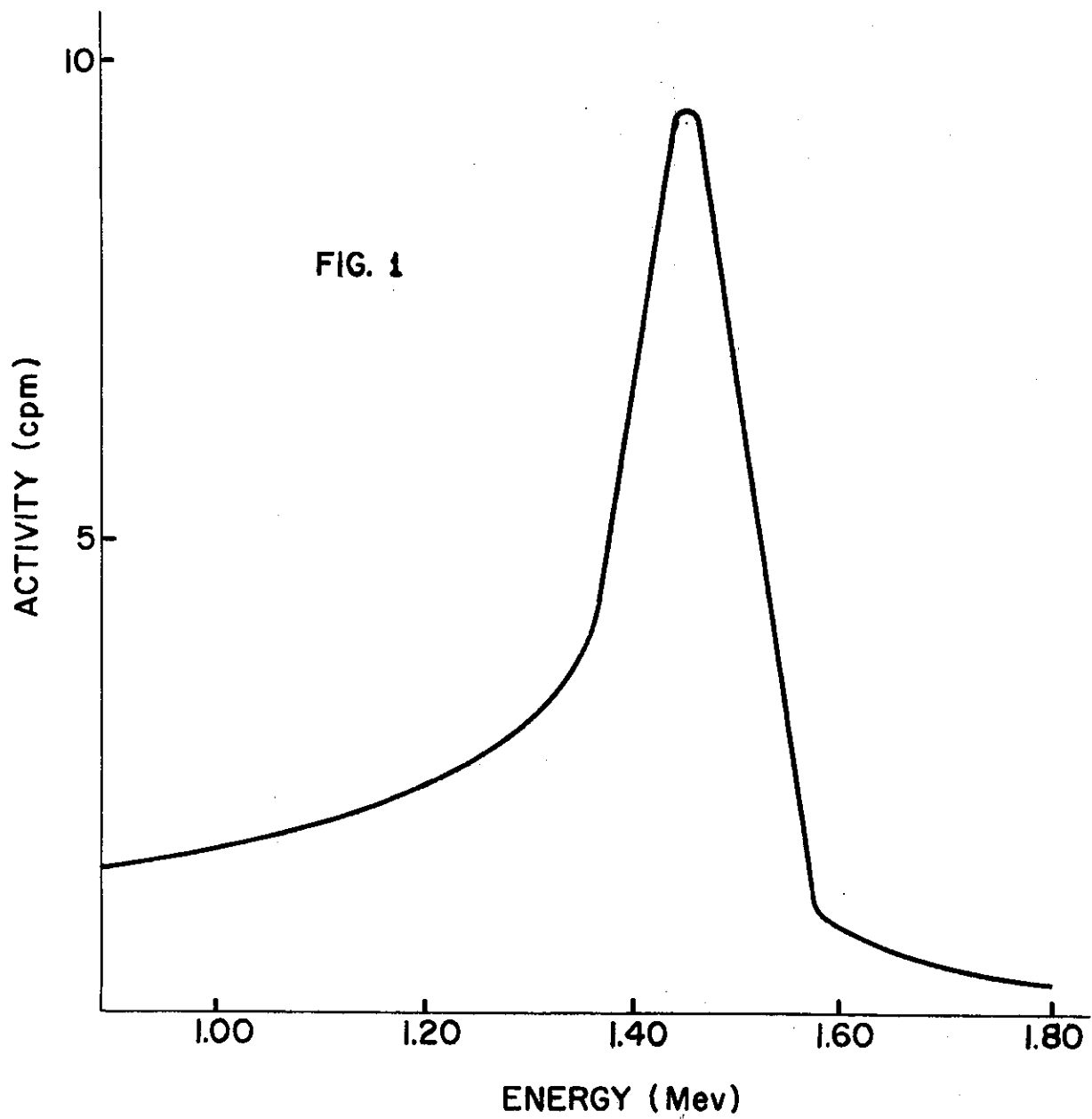
The activities at the two peaks were 2.4 cpm in the high energy one and 2.9 cpm in the low energy peak. The efficiency of the K gamma ray was determined at the peak as 2.07 %. From this we calculate for the Co^{60} peaks the efficiencies of 2.9 % and 4.0 % and get an absolute activity of 83 and 73 dpm for the two peaks. We estimated a weight of two for the first value and this gives an average activity of 80 dpm. The corresponding average activity in each liter of rain water is 0.0066 dpm/liter.

The yield of Co^{60} is comparable with the one found for Co^{57} and for Mn^{54} as reported by MARQUEZ, COSTA and ALMEIDA (1958). All the indications are that those radioisotopes in the neighborhood of iron are produced in nuclear tests of thermonuclear reactions. In fact, Mn^{54} was found in the fall out in the Northern Hemisphere by SHIPMAN, SIMONE and WEISS (1957), and they attributed it definitely to a thermonuclear test. However the two previous isotopes were formed accidentally in the test by reactions such as $\text{Fe}^{56}(\text{d},\text{n})\text{Co}^{57}$ and $\text{Fe}^{54}(\text{n},\text{p})\text{Mn}^{54}$. The Co^{60} could be formed accidentally only by nuclear reactions of neutrons on Ni such as $\text{Ni}^{60}(\text{n},\text{p})\text{Co}^{60}$, or else it is formed in the so called "cobalt bomb" by the reaction $\text{Co}^{59}(\text{n}, \text{gamma})\text{Co}^{60}$.

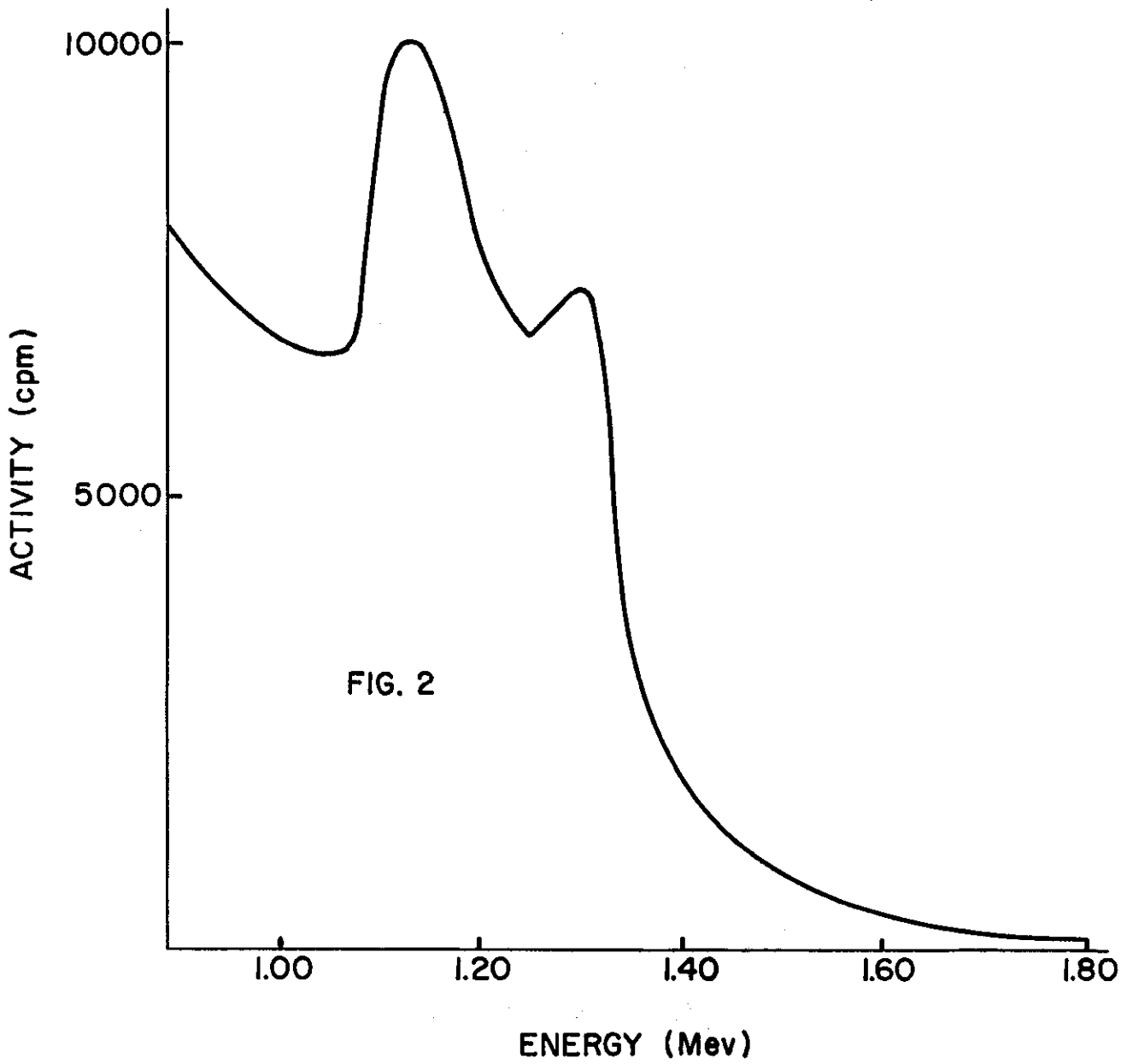
In conclusion, we have determined and measured the amount of Co^{60} in rain water and it is 0.0066 dpm/liter. We have very strong evidence that this Co^{60} comes from thermonuclear tests. There are only two possible origins for this Co^{60} , either the shell of the

bomb contained large amounts of Ni, or the Co^{60} comes from the test of one or more "cobalt bombs".

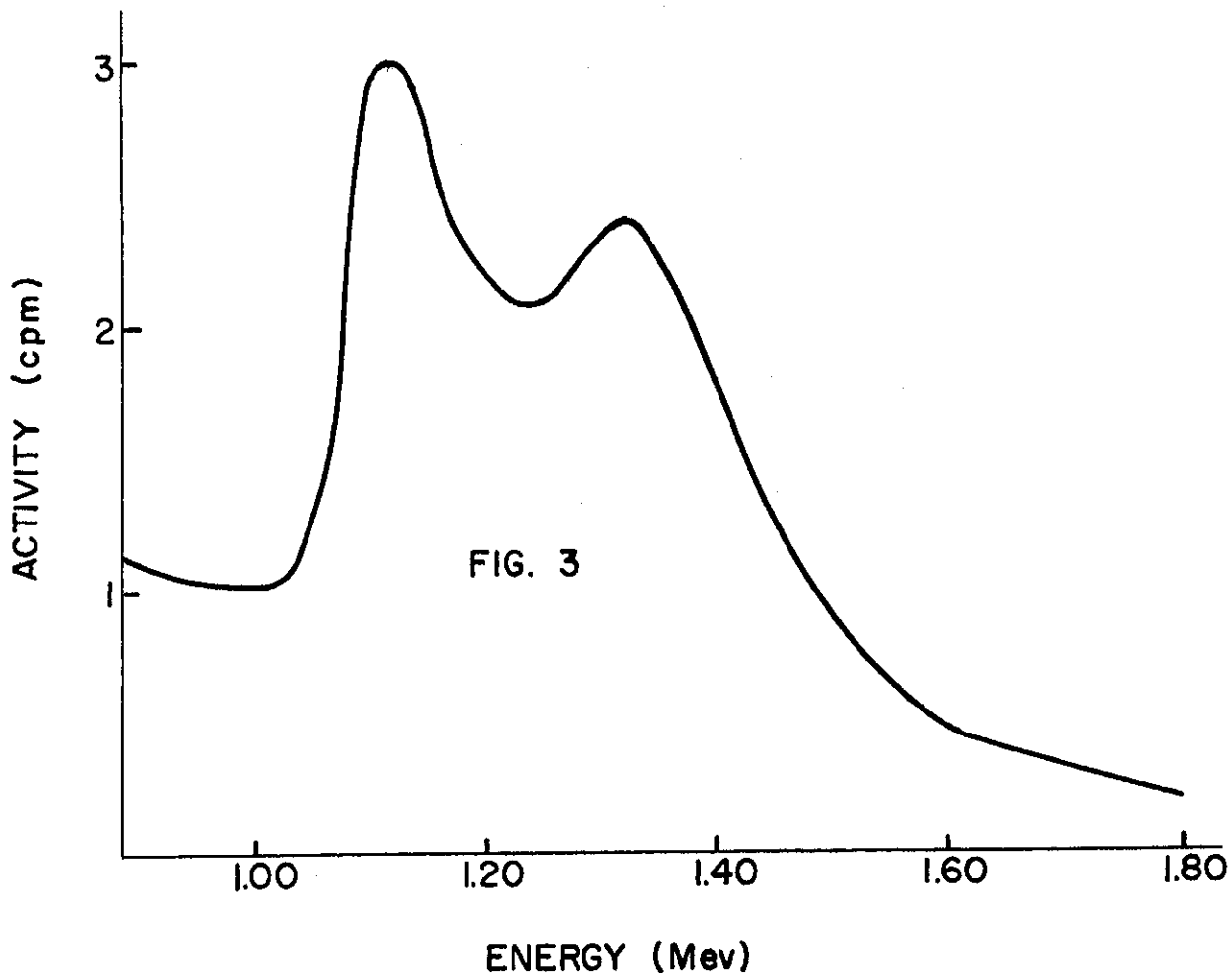
1. L. Marquez, N. L. Costa, and I. G. Almeida, *An. Acad. Brasil. Ci.* 29, 3, -361, (1957).
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The gamma ray spectrum of K^{40}



The gamma ray spectrum of Co^{60} from a standard source.



The gamma ray spectrum of Co^{60} from rain water