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RADIOISOTOPES FROM FUSION IN RAIN WATER: Co^{57}, Mn^{54} AND Co^{60} *+

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INTRODUCTION

It has been suggested by Craig[1] and by Begemann and Libby[2] that a large fraction of the natural tritium found in the atmosphere and the surface of the earth comes from the sun. They have made this suggestion because they calculate with certain assumptions the rate of production of tritium from the tritium concentration in hydrogen, and this rate turns out to be greater than the rate calculated from cosmic ray fluxes and production cross sections.

These authors do not mention how this tritium is produced and how it comes from the sun to the earth. Evidently, these two questions are very difficult to answer from our lack of information

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about the detailed processes involved. We will mention, however, certain conclusions that can be drawn from the suggestion that tritium comes from the sun. Some of these conclusions are almost certain and some are only possible.

Since the half life of tritium is 12.26 yr, it is almost certain that the tritium is formed on the surface of the sun or near it.

Since the temperature in the surface of the sun is about one electron volt, it is almost certain that this tritium is not formed continuously, but it is formed in outbursts of activity such as solar flares. In those outbursts of activity, the local temperature should reach the Kev region to have an appreciable formation of tritium.

It is almost certain that tritium would be formed by the reaction $He^3(n,p)t$, by the reaction $d(d,p)t$, or both of them. The neutrons for the first reaction would come from reactions such as $C^{13}(\alpha,n)O^{16}$ or $Ne^{21}(\alpha,n)Mg^{24}$.

It is possible that in the outbursts that produce tritium the local temperature reaches the Mev region, and in this case the reactions (p,γ) and (p,n) would be prominent in the light and medium nuclei.

It is possible that the mechanism that brings the tritium from the sun to the earth, could bring also heavier nuclei.

We thought then that it would be worth while to see if one could find another radioisotope in the atmosphere which might come from the sun. Examining the situation, one can see that for that purpose Co^{57} has a privileged position. It can be formed by the reaction $Fe^{56}(p,\gamma)Co^{57}$. Since Fe^{56} is at least twenty times more abundant than any other stable isotope in the iron peak, we would expect the same ratio in the yield of Co^{57} and neighboring isotopes

formed by the same reaction. Co^{57} has a half life of 267 d; the mean life in the stratosphere for a radioisotope before being washed down by tropospheric rains is about ten years, so we expect that about 10 % of the Co^{57} will come down with the rain. Co^{57} has a mass number greater than 40 and can not be masked by the spallation products of atmospheric argon.

We looked for Co^{57} in the rain water and found it. Since there has been lately many nuclear tests in the atmosphere, we looked for other radioisotopes in the neighborhood of Co^{57} and found Mn^{54} and Co^{60} in yield comparable to the yield of Co^{57}. From this last fact we have been led to conclude that these radioisotopes come from nuclear tests in the earth. Mn^{54} has been identified recently as a fall out product in the Northern Hemisphere[3]. Although we started this work with a different purpose, the only positive conclusion is that there are many radioisotopes in the atmosphere produced in thermonuclear tests.

EXPERIMENTAL METHOD

The rain water that fell in an area of 200 m^2 was sent through channels and pipes into tanks. In the earlier experiments we used tanks of 1000 liters, now we are using a tank of 4500 liters. For every 1000 liters of water we put 20 mg of Co, Mn and Fe. The water was stirred and after settling it was passed through a large cation exchange column full with Amberlite-IR-120 in the acid form. The column was eluted with 4 N HCl and this evaporated to a small volume.

The detailed chemical procedure has been described elsewhere[4]. It will be outlined here. Silica was separated. Scavenging was done of zirconium phosphate; lanthanum oxalate; lead, strontium

and barium sulphate; bismuth, antimony, selenium, tellurium and copper sulphide in 0.3 N HCl solution; and iron and aluminum hydroxides. Undesirable anions were eliminated by retaining the cations in a smaller ion exchange column and washing it. The Mn was precipitated as MnO_2 from HNO_3 solution with $KClO_3$ and weighed to determine its chemical yield. The Co was precipitated with 1-Nitroso-2-Naftol, incinerated to Co_3O_4 and weighed to determine its chemical yield.

The final products were transferred to a small test tube and counted in a well crystal scinitllation spectrometer[4]. The energy scale of the spectrometer and its efficiency at each energy were calibrated using sources of Th^{234}, Na^{22}, Co^{60} and K^{40}.

The spectrum of Co^{57} showed a single line with an energy of 125 Kev. It was counted with an efficiency of 65 %. Some early data of Co^{57} were reported already[5]. All the results that we have until now are shown in TABLE I. The average activity from these results is 0.0145 dpm/liter.

The spectrum of Mn^{54} showed a line with its energy of 840 Kev. It was counted with an efficiency of 2.4 %. The results are shown in TABLE II. The average activity for Mn^{54} from these results is 0.0625 dpm/liter.

Finally all the samples of Co shown in TABLE I were placed in a single test tube and counted. It was not possible to detect Co^{56} or Co^{58}, but it was found that there was Co^{60}. It showed its spectrum with peaks at 1.17 Mev and 1.33 Mev. The activity of Co^{60} corresponds to an average activity of 0.0066 dpm/liter.

DISCUSSION

The average yield of Co^{57} is 0.23 times the average yield of

Mn^{54}. If they were of solar origin and formed by (p,γ) we would expect the ratio Co^{57}/Mn^{54} to be equal to the ratio of the abundances of Fe^{56}/Cr^{53} and this is 55 from the result of Goldberg[6]. Since it has been found that Mn^{54} is a fall out product[3], we are led to conclude that the bulk of Co^{57} comes from thermonuclear tests too. The ratio Co^{57}/Mn^{54} changes very little with time since they have almost the same half life.

The most likely reactions leading to the formation of Co^{57} in nuclear tests are $Fe^{56}(d,n)\ Co^{57}$ and $Fe^{57}(p,n)Co^{57}$.

The most likely reactions leading to the formation of Mn^{54} are $Fe^{54}(n,p)Mn^{54}$ and $Cr^{53}(d,n)Mn^{54}$.

The most likely reactions leading to the formation of Co^{60} in nuclear tests are $Co^{59}(n,\gamma)\ Co^{60}$, $Ni^{60}(n,p)Co^{60}$ and $Fe^{58}(t,n)Co^{60}$.

We can compare the results of this work with the activities found by us for Cs^{137} and Sr^{90}. They are 0.69 dpm/liter and 0.49 dpm/liter respectively[7]. We see that the activities reported in this work are from one to two orders of magnitude smaller than the activities of these fission products.

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TABLE I

Date of rain	Amount of rain water in liters	Absolute specific activity in dpm/liter
June 1957	8500	0.016
September 1957	3500	0.014
October 1957	3000	0.040
December 1957	7500	0.011
February 1958	3500	0.007
March 1958	3200	0.003

TABLE II

Date of rain	Amount of rain water in liters	Absolute specific activity in dpm/liter
December 1957	7500	0.075
February 1958	3500	0.048
March 1958	3200	0.050