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THE FORMATION OF ^{22}Na FROM ATMOSPHERIC ARGON BY COSMIC RAYS (*)(**)

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I- INTRODUCTION.

It has been established recently that several radio-isotopes are formed as spallation products of argon by cosmic rays. They have all been found in the rain water. The formation of ^{32}P was measured[1]. Winsberg[2] found the ^{39}Cl; Goel[3] found the ^{35}S; and Lal et al[4] verified the result of ^{32}P and established the formation of ^{33}P.

We have in this work isolated the 2.6 yr ^{22}Na. This isotope is the longest-lived found so far from the spallation of argon.

II- EXPERIMENTAL METHOD.

The rain water from a large roof was collected and

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stored in tanks of 1000 liters. The water was allowed to stand until it was clear and the dust settled. The water as passed through an ion-exchange column of 50 cm^2 area and 50 cm height, loaded with Amberlite IR-120 resin. The flow rate was about 30 liter/hr. After passing three to four thousand liters of rain water through the column, it was regenerated with 4N HCl, until it would not give more test for metals. About 20 liters of 4N HCl were required.

The 4N HCl containing the metals was evaporated to near dryness. The hydroxides ware precipitated with NH_4 OH and filtered. The filtrate was treated with $(NH_4)_2$ CO_3 until all the Ca, Mg, and the like were precipitated, allowed to stand and filtered. The filtrate was taken to dryness and then treated with a mixture of one part concentrated HCl and four parts concentrated HNO_3, boiled until all the ammonium salts were destroyed, and then taken to dryness again.

The dry residue contained only the alkaline metals. It was dissolved in water and passed through an ion-exchange column of 20 cm^2 area and 100 cm height and washed with a very dilute HCl solution to separate the anions. Then the column was eluted with 0.1N HCl and the Na was collected; the K came out afterwards and it was rejected, and then the column was washed with 4N HCl to determine the ^{137}Cs which comes with the rain water too.

The solution containing the Na was evaporated to a small volume and taken to dryness in a beaker having the same diame-

ter as the crystal detector. The amount of Na Cl from each sample varied from 15 to 60g and this is in agreement with the amount expected from the known amount of Na in rain water.

The solution containing the Cs was evaporated to a volume of 1 ml and transfered to a test tube.

The samples were counted with a scintillation spectrometer Model 516 built by Baird-Atomic, Inc., Cambridge, Mass., U.S.A.; we used a well crystal having 1 3/4" diameter and 2" height; the size of the well was 5/8" diameter and 1 1/2" height.

The ^{137}Cs was counted in the well, but the Na had to be placed outside because, in the volume of the well, one could only use a small fraction of the amount of Na Cl that we had at the end.

The ^{22}Na was measured by the annihilation radiation of the positrons that it emits. The ^{137}Cs was measured by the 662 Kev gamma ray of its daughter ^{137}Ba. The energy scale of the spectrometer was calibrated using positrons from an artificial source of ^{22}Na. The counting efficiency was determined by counting a sample of artificial ^{22}Na whose absolute activity had been determined previously by two methods already described (5).

The energy of the gamma rays from the natural ^{22}Na agreed with the energy of the annihilation radiation from the artificial ^{22}Na within 20 Kev. The ^{137}Cs had a greater activity and its gamma ray energy agreed with the tabulated energy within 5 Kev.

III- RESULTS.

The channel width of the spectrometer was adjusted to maximize the product of the figure of merit for detection S^2/B and the resolution. In this condition, the efficiency for the ^{22}Na placed outside the crystal was 3.5 % in the photoelectric peak. It would have been 7.3 % if we could have counted it inside the well. The resolution of the spectrometer was 8 %, measured as full width at half the maximum.

The background was counted with NaCl reagent, treated and mounted in the same way as the Na Cl from the rain water. Its counting rate in the photoelectric peak was 20 cpm.

The results of three experiments are shown in Table I. The spread in the values found for the activities is common in this kind of experiment. The average activity of ^{22}Na in the rain water of Rio de Janeiro is then 0.017 dpm/liter. This is compatible with other activities found already and with spallation data.

The average activity of the ^{137}Cs was 1.2 dpm/liter (6). It is 70 times greater than the average activity of the natural ^{22}Na. Since it is likely that in any nuclear weapon test the amount of ^{137}Cs formed would be hundreds of times greater than the amount of ^{22}Na formed, we conclude that the ^{22}Na observed has a natural origin and that it is formed as a spallation product of argon by cosmic rays.

We would not attempt to calculate the formation rate of ^{22}Na in this work, since this envolves the times of mixing of

the atmosphere. The use of these radio-isotopes to study atmospheric process is a new line of research and there has been made one work of this nature by Benioff[7] with the data on Be^7. Perhaps using the data of all the radio-isotopes produced in the atmosphere by cosmic rays, one could get a more complete picture.

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TABLE I. - The activity of ^{22}Na in rain water of Rio de Janeiro

Date 1957	Amount of water liters	Observed activity cpm	Absolute activity dpm/liter
January	2.500	0.0 ± 0.5	0.000
March	4.000	1.7 ± 0.4	0.012
June	3.000	4.0 ± 1.0	0.038