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THE FORMATION OF P^{32} FROM ATMOSPHERIC ARGON BY COSMIC RAYS

by

L. MARQUEZ and NEYLÁ L. COSTA

CENTRO BRASILEIRO DE PESQUISAS FISICAS

Av. Wenceslau Braz, 71

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THE FORMATION OF P^{32} FROM ATMOSPHERIC ARGON BY COSMIC RAYS *§

L. Marquez and Neyla L. Costa

Centro Brasileiro de Pesquisas Físicas

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Summary. - It is shown by experiments that radioactive P^{32} is formed as a spallation product of argon by cosmic rays. This P^{32} is found in the rain water, and its average activity in fresh rain water of Rio de Janeiro is 0.20 dpm/liter.

1. INTRODUCTION

It is known that the interaction of the cosmic rays with the nuclei of nitrogen and oxygen of the air produces a substantial number of neutrons which are captured after being slowed down by the reaction $N^{14} (n, p) C^{14}$. This radioactive C^{14} was discovered by Libby and co-workers, and it has been and is being used for dating archeological samples. Details of the work are found in a book by Libby¹. It is also expected that as spallation products

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of nitrogen and oxygen, all the isotopes lighter than O^{18} will be formed, and of these tritium was found by Kaufman and Libby², and Be^7 was found by Arnold and Al-Salih³ in rain water.

There is also in the atmosphere a substantial amount of argon, namely 0.93 percent by volume, and therefore one would expect that as spallation products of argon all the nuclides lighter than A^{40} would be formed. Of these nuclides, P^{32} is probably the easiest one to detect. We looked for and found P^{32} as a spallation product of argon by cosmic rays.

From cosmic ray data⁴ it is estimated that the maximum rate of formation of P^{32} lies between 10 and 20 Km height, and this rate decreases for lower and higher altitudes, being much smaller at sea level. From thermodynamic considerations one would expect that if the water in the atmosphere condenses, the H_3PO_4 would concentrate in the water phase. We would expect then to find the P^{32} in the rain water or in the water condensed from the air by any process.

We decided to look for it in the rain water and for this purpose we collected the water from the roof of our laboratory in an area of about 50 m² and this water was lead through pipes into tanks of 200 liters capacity. There were two unexpected difficulties in the chemical procedure. The rain water carried always a large amount of silica from the tile roof, and this made necessary the separation of silica. The rain water also carried some 5 mg of P per tank from the decomposition of leaves and other organic materials that fell on the roof between rains. This made impossible to prepare thin samples of P.

2. EXPERIMENTAL METHOD

The chemical procedure adopted was as follows. To each tank with 200 liters of rain water was added 5 mg of P as phosphate, 15 ml of concentrated HNO_3 and one gm of Fe as ferric ion. The water was stirred and allowed to stand for one hour; after that some base was added to bring the solution to pH of 10 or greater to precipitate the ferric hydroxide which would carry the phosphate. The solution was allowed to stand until the precipitate settled in the bottom and the supernatant was clear. This took several hours and sometimes it was necessary to add a solution of K_2SO_4 to help the coagulation of the ferric hydroxide. The supernatant was siphoned and the residue was filtered.

The hydroxide was dissolved by boiling it with 500 ml of 4 N HCl for 15 minutes. At this point it was necessary to filter insoluble leftovers from the tiles and the leaves. The filtrate was oxidized with bromine water, evaporated to dryness and dehydrated for one hour at 110°C . The residue was taken up with some 12N HCl, diluted with water and the silica filtered. The filtrate was evaporated to near dryness, boiled with concentrated HNO_3 to expel HCl, diluted with a 10 percent solution of ammonium nitrate, and the phosphate precipitated with ammonium molybdate reagent.

The ammonium phosphomolybdate was centrifuged, washed with dilute HNO_3 , and dissolved in NH_4OH containing some citric acid. The solution was made faintly acid, magnesium chloride was added and then made basic with NH_4OH to precipitate the magnesium ammonium phosphate. This precipitate was centrifuged, washed with

dilute NH_4OH , dissolved in HCl , evaporated to dryness, and dehydrated as before to eliminate the silica. The residue was taken with HCl , diluted and the silica filtered. The filtrate was diluted to 0.3 N in HCl and passed through a cation exchange column. For the preparation and use of the column we followed Samuelson⁵. The effluent and washings were evaporated to a small volume and the phosphate precipitated as the magnesium ammonium phosphate. It was filtered through a circular paper filter of 2.5 cm diameter, washed, dried, weighed, and mounted for counting.

The samples were counted at 3 mm from the end window of a Geiger counter. The mica window had a diameter of 2.5 cm and it was 2 mg/cm^2 thick. Initial activities ranged from 10 to 40 cpm. The background in a lead shield was 27 cpm. The counts were recorded in a scaler of conventional type.

Once we found that the P^{32} appeared consistently in the rain water, we made a test to find out whether part or all of this P^{32} could come from a nuclear explosion. A nuclear explosion produces neutrons in the Mev region. These neutrons can make P^{32} by three processes; $\text{P}^{31} (n, \gamma) \text{P}^{32}$, $\text{S}^{32} (n, p) \text{P}^{32}$ and $\text{Cl}^{35} (n, \alpha) \text{P}^{32}$. The first reaction is more rapid with slow neutrons, and the two others only go with neutrons in the Mev region. For that test we made a separation of Ba, with the idea of observing Ba^{140} , which has a large fission yield and its half-life is almost the same as the half-life of P^{32} . We added 5 mg of Ba per tank of 200 liters of rain water. The ferric hydroxide was precipitated with a mixture of sodium hydroxide and sodium carbonate to precipitate the

Ba also. The rest of the procedure was the same as before and the Ba was precipitated as BaSO_4 from the supernatant of the ammonium phosphomolibdate. We could not observe any Ba^{140} , but from the activity observed we could estimate that the activity of Ba^{140} is at least four times smaller than the activity of P^{32} in the same amount of rain water. This shows that the P^{32} that we found is not formed in a nuclear explosion, since it seems that much more Ba^{140} would be formed in a nuclear explosion than P^{32} .

3. RESULTS AND DISCUSSION

The results of six experiments are shown in Table I. The activities were followed until they got lost in the back-ground; this was from three to five half-lives and no other activity was seen throughout this period, which shows that the chemistry was satisfactory. The maximum energy of the beta particles was found by measuring the absorption coefficient at the initial part of the absorption curve with Al; and it was in agreement with the maximum energy of the beta particles from P^{32} . In order to convert initial activities into desintegration rates we had to apply correction factors for geometry, absorption, self-absorption back-scattering and chemical yield. For these corrections we estimate that the absolute activities might be in error as large as 30 percent.

There is a spread of a factor of three between the smallest and the largest activity of P^{32} found; this is similar to the spread of values found by Arnold and Al-Salih³ in the case of Be^7 . The average value of the activity of P^{32} in the rain water

of Rio de Janeiro is 0.20 dpm/liter. This is compatible with what one would estimate from the activity of Be^7 found by Arnold and Al-Salih, taking into account the amount of Li in the air, plausible yields for P^{32} from Li and of Be^7 from N and O , and the decay from the time of the formation to the time of the rain-fall. We do not attempt to calculate the formation rate of P^{32} , since this involves meteorological times which are uncertain.

We could not detect the activity of P^{33} , although it should be there. This was due to the fact that our samples had thickness of about 30 mg/cm^2 . Taking into account the absorption in the sample, window, etc., and estimating the yield of P^{33} from the empirical relation for spallation reactions used by Rudstam⁶, we calculate that the P^{33} could not appear neatly in our decay curves. It could shift the half-life of P^{32} towards a larger value, and there seems to be some indication of that, but we can not consider that as a definite proof that P^{33} is there.

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Table I. The activity of P^{32} in rain water of Rio de Janeiro and related data.

Date 1955	Amount of rain water liters	Observed half-life days	Initial activity dpm/liter
May 30	500	12	0.09
May 31	600	17	0.23
June 9	300	15	0.30
June 21	600	15	0.26
June 23	800	15	0.23
July 8	800	14	0.10