New Nuclides Produced in Chlorine Spallation*

MANFRED LINDNER
California Research and Development Company, Livermore, California
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The radiation properties of the beta-emitting nuclides Si^{32} and Mg^{28} are described. Si^{32} was found to have a maximum probable half-life of 710 years, emitting beta-particles of $E_{max}\sim100$ kev. Absence of gamma radiation with this nuclide is indicated.

INTRODUCTION

THE formation of Mg^{28} in bombardments of sodium chloride with 340-Mev protons,[1] in bombardments of magnesium with 40-Mev helium ions, and of silicon with high-energy gamma rays[2,3] and with high-energy protons[4,5] has been reported. Sheline[3] has proposed a decay scheme for this nuclide based upon the gamma rays detected in a scintillation counter. Marquez[4] studied the beta spectra for the pair Mg^{28}-Al^{28}. Both were found to have "allowed" shapes. The present paper describes experiments with Mg^{28} and the characterization of the nuclide Si^{32}.

EXPERIMENTAL PROCEDURE

Samples of sodium chloride weighing about five grams were wrapped in aluminum foil and bombarded in the Berkeley 184-inch cyclotron with 340-Mev protons. The Mg^{28} was isolated in a carrier-free state for beta-spectrum determination, while the Si^{32} was isolated with about one milligram of added silicon carrier.

CHEMICAL SEPARATIONS

The irradiated sodium chloride was dissolved in water, about one milligram of iron carrier added to the solution, and the solution made alkaline with NaOH. The ferric hydroxide was centrifuged, washed and dissolved in 6*N* hydrochloric acid. One milligram of phosphoric acid and an amount of zirconium in slight excess of that required to cause complete precipitation of the phosphate was added. The supernatant from this precipitation was made 10*N* in HCl, then passed through a Dowex-A2 anion exchange resin column. This removed all the iron and excess zirconium. The HCl eluate, containing only the magnesium tracer, was was evaporated to dryness, taken up in water, and the solution passed through a Dowex-50 cation exchange resin column. The Mg^{28} remained on the column, was then removed by elution with 0.5*N* HCl. This eluate was evaporated to dryness leaving only a very slightly visible residue containing the Mg^{28}. This residue may have been due to the presence of a small amount of magnesium chloride present in the original sodium chloride target. Extraordinary precautions had been taken to avoid possible introduction of carrier magnesium through the use of reagents made only from conductivity water.

To the supernatant solution from the original carrying of Mg^{28} on ferric hydroxide was added about one milligram of silicon as $(NH_4)_2SiF_6$. About 10 ml of concentrated H_2SO_4 was added, the resultant solution brought to fuming, cooled, and diluted to about 10*N* H_2SO_4. A small gelatinous precipitate was centrifuged, washed, and dissolved in 6*N* NaOH. After several ferric hydroxide precipitations were made from this solution and discarded, the solution was made 6*N* in HCl, one milligram of phosphorus (as H_3PO_4) and excess zirconium added (as described above). After removal of the zirconium phosphate, the supernatant was heated to fuming with sulfuric acid and the resultant precipitate of SiO_2 separated, washed with acetone, mounted, and counted.

It was found that the zirconium phosphate precipitation step was essential, since P^{32} formed by the reaction $Cl^{37}(p,p\alpha n)P^{32}$ was found to be a major contaminant in silica precipitates, even when the SiO_2 precipitations were made in the presence of phosphoric acid.

EXPERIMENTAL RESULTS

Mg^{28}

About 10^8 disintegrations per minute of Mg^{28} were mounted for a beta-spectrum analysis. The gamma rays associated with the Mg^{28}-Al^{28} pair were determined with a thallium-activated NaI scintillation crystal coupled with a 50-channel pulse-height analyzer. The results of these investigations essentially reproduced those reported by Sheline[3] and by Marquez[4] and, therefore, need little further discussion. The 0.03-Mev gamma rays reported by Sheline were determined in this laboratory as 27 kilovolts on a proportional counter.

The decay of Mg^{28} was followed for ten half-lives before evidence of contamination was observed. Together with experiments using magnesium carrier in which the decay could be followed for thirteen half-lives, it was possible to set a value of 21.2 ± 0.2 hours for the half-life of Mg^{28}. This is in agreement with Sheline's value of 21.3 ± 0.2 hours[3] but measurably different

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[1] M. Lindner, Phys. Rev. **89**, 1150 (1953).
[2] R. K. Sheline and N. R. Johnson, Phys. Rev. **89**, 520 (1953).
[3] R. K. Sheline, Phys. Rev. (to be published).
[4] L. Marquez, Phys. Rev. **90**, 330 (1953).
[5] J. W. Jones and T. P. Kohman, Phys. Rev. **90**, 495 (1953).

from the values of 22.1±0.3 hours reported by Jones and Kohman.[5]

The cross section for the reaction $Cl^{37}(p,2\alpha 2p)Mg^{28}$ with 340-Mev protons on NaCl was calculated to be approximately 3×10^{-27} cm^2. Contribution from the reaction $Cl^{35}(p,\alpha 4p)Mg^{28}$ was considered negligible. This conclusion is justified on the basis of spallation yields of other light elements,[6,7] in which it has been shown that the probability for neutron and for proton emission at very high energies become approximately equal for the light elements. Furthermore, Jones and Kohman[5] estimate a cross section of about 10^{-28} cm^2 for the reaction $Si^{30}(p,3p)Mg^{28}$, this being about three percent of the value found in chlorine spallation for the formation of Mg^{28}. Thus, with proton beams of approximately several tenths of a microampere available on the 184-inch Berkeley cyclotron, activities of the order of a millicurie of Mg^{28} could be produced with targets weighing several grams.

Si^{32}

The decay curve found for the silicon chemical fraction followed on a chlorine-filled Amperex argon counter is shown in Fig. 1. This shows the exponential decay of Si^{31} over twenty half-lives, followed by the growth of a fifteen-day daughter.

An absorption curve was obtained with aluminum absorbers in a nucleometer at the minimum of the decay curve in Fig. 1. Care was taken that there could be no contribution from the beta particles of Si^{31}. This curve is shown in Fig. 2. It is seen that beta particles

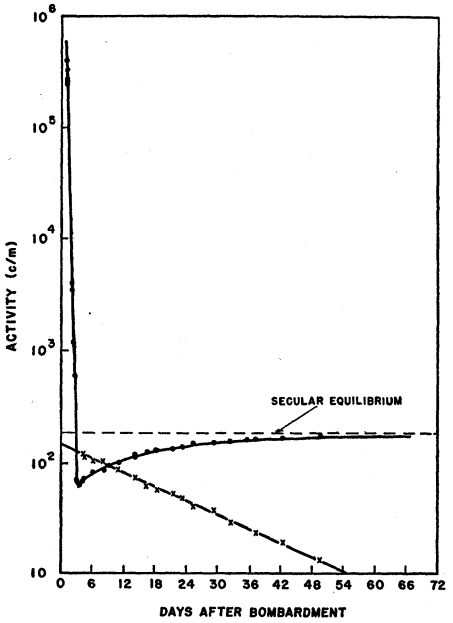

FIG. 1. Radioactive decay of silicon fraction. ●—Gross decay; ×—15-day line from resolution of curve at secular equilibrium.

[6] Batzel, Miller, and Seaborg, Phys. Rev. **84**, 671 (1951).
[7] Rudstam, Stevenson, and Folger, Phys. Rev. **87**, 358 (1952).

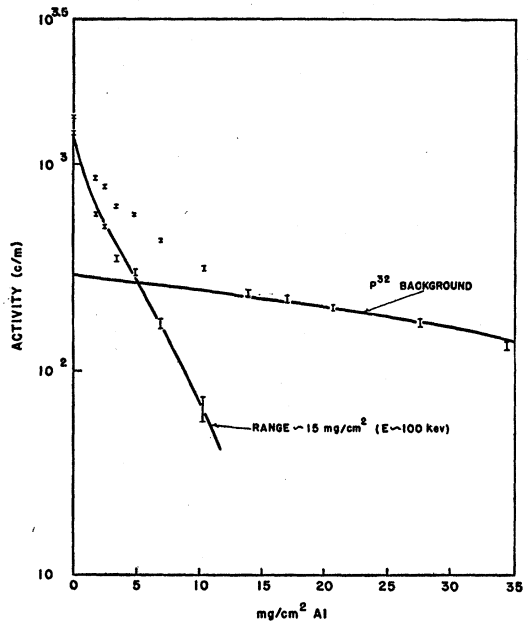

FIG. 2. Absorption curve of silicon fraction through aluminum showing some background of the P^{32} daughter. Curve obtained in a methane flow-type proportional counter of about fifty percent geometry.

of range approximately 15 mg cm^{-2} (corresponding to $E_{max}\sim$100 kev) and a lesser intensity of harder background radiation are present. Presumably the harder radiations are due to P^{32} daughter, which had grown into the sample during the several days, which had to be allowed for the Si^{31} decay. The ratio of the intensities of these two beta groups is approximately correct for the amount of P^{32} which is calculated to grow into a sample of Si^{32} during this period of time.

The entire growth curve of Fig. 2 also indicates that the counting efficiency of the parent activity on the argon-chlorine counter, as a result of the low energy, is only about half that of the fifteen-day daughter. Finally, corroboration of this observation is found in Fig. 3, an aluminum absorption curve of the silicon sample when daughter growth was essentially complete. It is seen that, although the activity of the sample was very low, beta particles of range about 800 mg cm^{-2} are present, in addition to beta particles of much shorter range. The range of the harder radiation corresponds to 1.7 Mev, this being the E_{max} value for the beta particles of P^{32}.

Finally, a tentative observation from Fig. 3 is the probable lack of gamma radiation accompanying the beta decay. The counting rate at 800 mg/cm^2 of absorber was actually determined under conditions of geometry threefold greater than the rest of the curve, the value being adjusted accordingly.

These data, therefore, establish the existence of the long-lived Si^{32}, which emits beta particles of about 100 kev and apparently no gamma radiation.

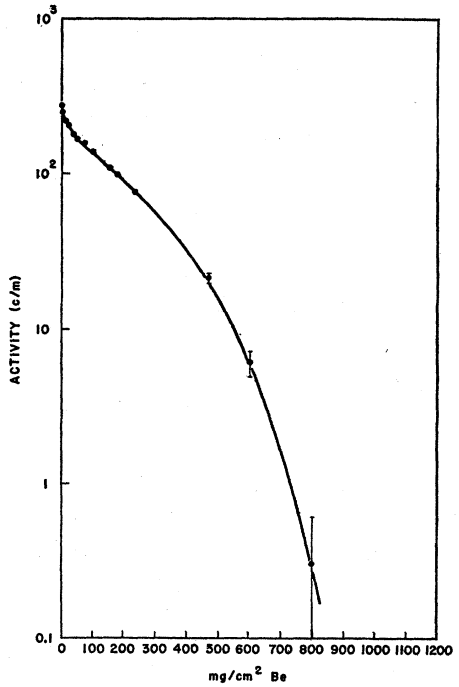

FIG. 3. Beryllium absorption curve of silicon fraction at secular equilibrium. Curve taken on an end-window type argon-filled counter under conditions of about five percent geometry.

The half-life of this isotope could be reasonably well estimated by comparing the activity of Si^{31} obtained from the reaction $Cl^{37}(p,\alpha 2pn)Si^{31}$ with the Si^{32} activity obtained in the same sample by the reaction $Cl^{37}(p,\alpha 2p)Si^{32}$. The half-life may then be calculated from

$$T_{32}=0.693\frac{A_{31}}{A_{32}}\cdot\frac{\sigma_{32}}{\sigma_{31}}\cdot\frac{t}{1-e^{-\lambda_{31}t}}. \quad (1)$$

In this equation, A refers to the activities of the two isotopes, σ to the formation cross sections, λ is the decay constant, and t the duration of the proton bombardment.

The measurement of the relative activities of Si^{31} and Si^{32} was comparatively simple, since this could be done in one sample of silicon if obtained in high chemical purity. Furthermore, although the radiation associated with Si^{32} was inconveniently soft, the equilibrium level of P^{32} daughter was easily determined, as shown in Fig. 1, and could be substituted for the parent Si^{32}. Since Si^{31} and P^{32} have almost identical radiation characteristics, the ratio A_{31}/A_{32}, if determined in the same sample, was independent of chemical yield, self-absorption, backscattering, etc. and could be obtained from Fig. 1.

The quantity σ_{32}/σ_{31}, in Eq. (1), could only be estimated. Information is meager concerning the relative yields of the $(p,\alpha 2p)$ and $(p,\alpha 2pn)$ reactions in light elements. Rudstam *et al.*[7] have established cross-section contours for spallation products of iron with 340-Mev protons which would, on the basis of approximately equal probability of neutron and proton emission from a highly excited light nucleus, permit one to estimate that the ratio for the $(p,\alpha 2p)(p,\alpha 2pn)$ reactions in iron would be about 0.2. However, because of the uncertainties in this estimation and in the extrapolation of this effect to Cl^{37} as a target nucleus, the ratio σ_{32}/σ_{31} has, for purposes of this calculation, been taken as unity. On the basis of this assumption and from the ratio of the activities shown in Fig. 1, a half-life of 710 years was calculated for Si^{32}. It is unlikely that the half-life is as low as 100 years.

Using the rapid method described by Moszkowski,[8] the (ft) value for this nuclide becomes about 5×10^6. This probably represents a first forbidden transition. A similar determination for P^{32} yields an ft value of about 15×10^6, thus indicating that both Si^{32} and P^{32} represent first forbidden transitions.

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[8] S. A. Moszkowski, Phys. Rev. **82**, 35 (1951).