Half-life of ^{32}Si

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Previous measurements of the half-life of ^{32}Si, which is produced by cosmic ray spallation of ^{40}Ar and of interest for geophysical dating, display large discrepancies. We present a new method for half-life measurements of this and other isotopes. Two samples containing 6.7×10^8 and 7.4×10^8 ^{32}Si nuclei, respectively, were prepared by implantation of separated projectile fragments. The half-life of ^{32}Si was calculated from the two independent measurements of the specific activity to be 132 ± 13 yr.

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I. INTRODUCTION

The isotope ^{32}Si is produced in the Earth's atmosphere by cosmic-ray spallation of ^{40}Ar and is removed from the atmosphere by aerosols. Although ^{32}Si has a low natural concentration, it has been applied as a radioactive tracer in the study of glacier and sediment formation [1–3], of the circulation of the ocean water [4, 5], of the stability of underground water on a time scale less than 1000 yr [6] (by measurements on siliceous sponges), and of the bio-geochemical cycles of Si in the oceans [7]. However, its usefulness in these applications is hampered by large uncertainties in its half-life. As shown in Fig. 1, there have been a number of measurements of the ^{32}Si half-life [1, 3, 5, 8–13], but with significant variation in the results which are not correlated with the technique used. There are two ways of measuring the half-life of a long-lived isotope.

First, the change in the decay rate of a sample can be followed over a period of time until the change is significant enough to allow an accurate determination of the half-life, the so-called "decay" method [10]. In principle, this method can be very accurate and the initial number of nuclei in the sample does not need to be known. It should be noted, however, that in the case of ^{32}Si several years of continuous counting are needed to make a precise measurement. For the result of the decay measurement shown in Fig. 1 [11], the activity of a ^{32}Si sample was measured over a period of 4 yr, during which the decay rate of the ^{32}Si decreased by 1.6%. Accurate determination of such a small change requires very stable and precise measurements [10].

The second approach is the "equilibrium" method, in which both the number of the nuclei in the sample and the absolute activity of the sample are determined. Most of the results labeled AMS (accelerator mass spectrometry) in Fig. 1 used this approach, with the exception of the measurement of Thomsen *et al.* [13]. In the AMS measurements, ^{32}Si nuclei were produced in various nuclear reactions and then all silicon nuclei were separated from other reaction products and target materials by chemical means. The ratio of the number of ^{32}Si atoms to all silicon atoms, ^{32}Si/Si, in the resulting samples was measured by accelerator mass spectrometry, and the number of ^{32}Si in the sample was determined. The activities of these samples were also measured to determine the half-life.

The results from three geophysical measurements are also shown in Fig. 1. Measurements of the depth distribution of cosmogenic ^{32}Si in an ice core by Clausen [1] and a similar measurement by DeMaster [3] in sediment cores relied on the decay method. Somayajulu *et al.* [5] determined the lower limit for the half-life with

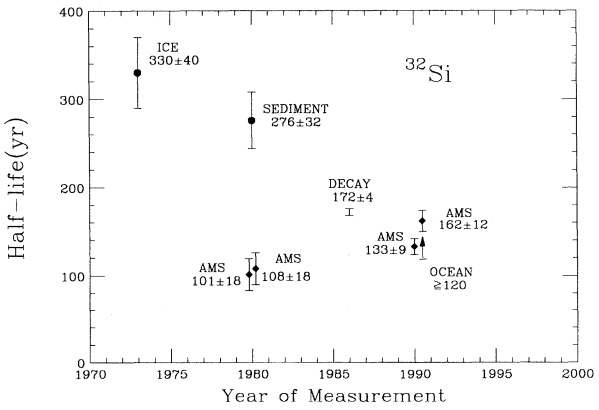

FIG. 1. Summary of previously reported half-life measurements of ^{32}Si. The results are labeled by the method or the natural reservoir and by the half-life value in years. Taken from the review by Kutschera *et al.* [10].

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the equilibrium method from a comparison of the global ^{32}Si inventory of the oceans with the average cosmic-ray production rate.

In the present study we have employed a completely different production and implantation method to accurately determine the number of ^{32}Si atoms deposited into an inert collector. The activity of the sample was then followed for a "short" period of time and the half-life determined by the equilibrium technique. Two independent samples were prepared and measured as described below.

II. EXPERIMENTAL METHOD

A. ^{32}Si production

In the present measurement, ^{32}Si was produced by fragmenting an E/A=65 MeV ^{40}Ar beam in a 235 mg/cm^2 Be target. Peripheral interactions at these intermediate energies cause the projectile to be fragmented and excited in a "rapid" encounter with the target. The excited projectile fragments undergo sequential decay, leading to the final projectile residues. The projectile fragments and the final residues retain the beam (vector) velocity and thus emerge from the reaction at high velocities near zero degrees in the laboratory. Transfer, pickup and other related processes are rare due to the large mismatch between the target and projectile momenta. Central collisions lead to explosive processes.

The A1200 fragment mass analyzer as shown in Fig. 2 at the National Superconducting Cyclotron Laboratory was used in a momentum-loss achromatic mode to separate the ^{32}Si ions from the many other projectile residues [14]. Briefly, these reactions produce a large variety of products ranging in mass from that of the projectile to the lowest values, all focused at zero degrees with nearly the beam velocity. The A1200 uses a primary magnetic analysis (to select all products with an m/q value) followed by passage through a momentum-loss foil (introducing a Z-dependent shift breaking the ambiguity) and a second magnetic analysis to select individual products. This leads to contaminants that are close ($\pm$1 unit in Z or N) to the selected fragment (see below). The thickness of the target was chosen to maximize the yield of ^{32}Si.

The momentum-loss degrader was a 150-mg/cm^2-thick Al foil shaped to maintain the achromaticity of the A1200 device for ^{32}Si nuclei [15].

Because of the relatively long half-life of ^{32}Si, the number of ^{32}Si nuclei needed for an accurate measurement of the decay rate was determined to be in the range of 10^8–10^9. With a measured collection rate of (8–10)$\times 10^3$ ^{32}Si ions per second at a primary beam current of 40–50 electrical nA, 24 h of continuous implantation were needed. The total number of implanted ions created sufficient radiation damage in the stopper such that continuous monitoring of the total number of implanted ions was not possible. Instead, two different techniques were developed and used to monitor the implantation beam.

These techniques are described in the following two subsections.

B. Experiment 1

In the first experiment, the ^{32}Si nuclei were implanted into a 123 mg/cm^2 Al collection foil located at the focal plane of the A1200. The collection foil was preceded by an aluminum mesh and followed by a silicon surface barrier detector (SSD), as shown in Fig. 3. The mesh was uniformly perforated with 0.34-mm-diam holes with an interhole distance of 1 mm. Ions that passed through the holes would not stop in the collector foil but rather in the 100-μm-thick SSD. The thickness of the mesh was such that those ^{32}Si nuclei that passed through the bulk part of the mesh would be stopped at the middle of the collection foil. At the beginning of the experiment, the transmission efficiency of the mesh was calibrated by replacing the 123 mg/cm^2 collection foil with a SSD of equivalent thickness. Using a lower beam intensity, the transmission efficiency was determined to be 10 $\pm$ 1% in good agreement with the expected value. By detecting only a fraction of the ^{32}Si beam, the Si detector was not expected to fail during the implantation and could be used to continu-

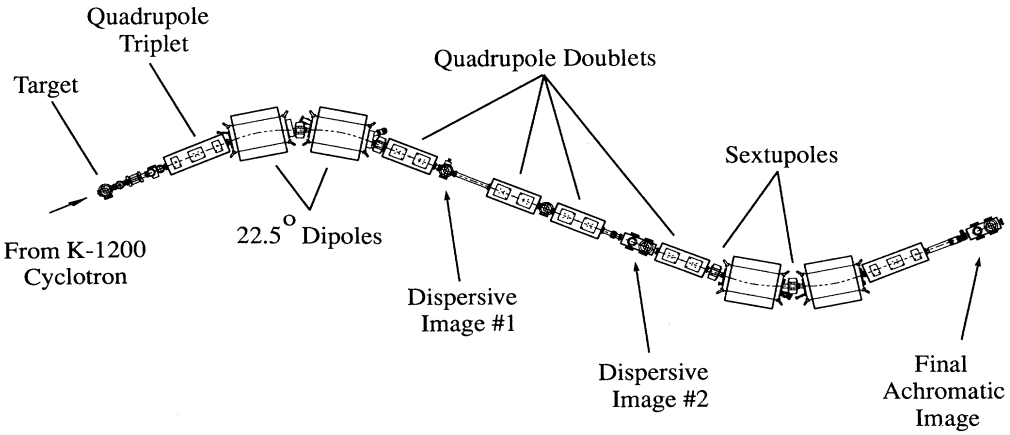

FIG. 2. Schematic drawing of the A1200 analyzer [14].

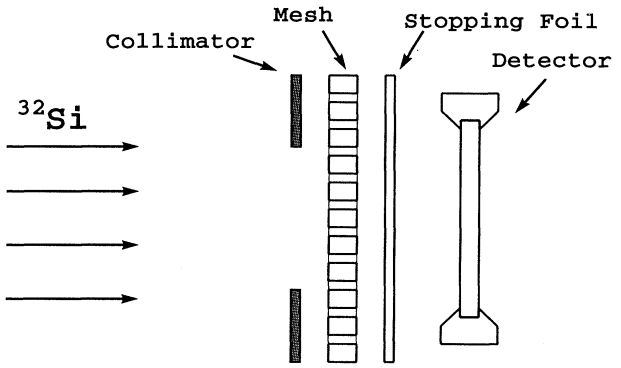

FIG. 3. Schematic arrangement of the mesh, stopper foil, and Si detector used in the first implantation.

ously monitor the number of ^{32}Si nuclei being implanted. This arrangement had two advantages: First, it allowed the implantation rate to be continuously sampled during the course of the entire experiment. Second, the particle identification and arrival time of the particles could be used in subsequent data analysis to correct for the decay of the short-lived nuclei during implantation. A total of $(6.7\pm0.7)\times10^8$ ^{32}Si nuclei was implanted along with some contaminants [^{33}P, $(6.0\pm0.7)\times10^7$; ^{32}P, $(5.8\pm0.6)\times10^5$; ^{31}Si, $(4.2\pm0.4)\times10^6$; and ^{28}Mg, $(3.6\pm0.4)\times10^5$] during a 24 h run. All of these isotopes have short half-lives compared to that of ^{32}Si. One of the contaminant ions, ^{31}Si, was used to calibrate the detector efficiency.

Following the implantation, the sample was counted in a rotating counting cycle (with the background foil) using two SSD's placed at opposite sides of the foil. The SSD's were 500 μm fully depleted detectors which were calibrated using the 122 keV line of ^{57}Co and the 80 and 356 keV lines of ^{133}Ba. A pulser was used for calibration at higher energies. The calibration was checked periodically using both the sources and pulser. The SSD's and the sample were contained in a shield with $\approx$5 cm of copper and $\approx$15 cm of lead and an internal volume of $\approx$1900 cm^3. Nitrogen gas was circulated inside the copper shielding to maintain a stable temperature. The typical background measured by the SSD's was approximately 2.0 counts/min for pulses with energies from 100 to 1710 keV, and the gains were found to be stable to within 3% over a period of several months.

Since ^{32}Si has a beta decay Q value of only 0.225 MeV it is more convenient to measure the decay rate of the daughter activity. ^{32}P has a well-known half-life of 14.282 days and a beta decay Q value of 1.71 MeV, and it decays to stable ^{32}S by β^- emission [16]. In order to determine the decay rate of ^{32}Si by observation of ^{32}P, a waiting period of about 80 days was required for the decay chain $^{32}\text{Si} \rightarrow {}^{32}\text{P} \rightarrow {}^{32}\text{S}$ to reach 98% of its equilibrium. In addition, other contaminants decayed to insignificant levels during this period. The sample and background (an aluminum foil with the same size as the sample that had been placed near the sample foil during implantation) were measured in rotation. All the data were collected by a dedicated acquisition system and recorded on disk. The measured sample activities together with the background measurements and the detector efficiency, discussed below, were used to determine the decay rate of the sample.

The decay of ^{31}Si was used to determine the detection efficiencies for the two SSD's for the first implantation (^{31}Si has a half-life of 2.622 h, and its beta decay Q value of 1.48 MeV is close to that of ^{32}P [16]). Approximately 4.2×10^6 ^{31}Si nuclei were present in the sample at the end of the experiment. As a result, more than 90% of the activity in the sample during the first 3 h of counting came from the decay of ^{31}Si. The total efficiency obtained for the two detectors was 0.48 ± 0.05. The uncertainty in the efficiency arose from the 10% uncertainty in the number of ^{31}Si nuclei, an estimated 5% error due to β spectral difference between ^{31}Si and ^{32}P, and the 1% uncertainty from counting and background subtraction.

The calculated rate of ^{32}Si decay in this sample was found to be 6.9 ± 0.8 decays per minute.

C. Experiment 2

In the second experiment, an addition was made to the A1200 device to determine the number ^{32}Si ions. A set of four *p-i-n* diode Si detectors were installed around the production target in a configuration such that the total number of reaction products reaching the detectors would be approximately independent of minor changes in the beam position on the target. The sum of the number of events in the four *p-i-n* detectors was thus proportional to the beam current. A new SSD was placed in a mount connected to a piston at the focal plane of the A1200 with which the ratio of the number of separated ^{32}Si ions to the sum of the counts in the four *p-i-n* detectors could be determined. The ratio was measured at several primary beam intensities and was constant to within 5%. The holder was then moved by the piston so that the SSD was replaced with a stack of Al foils. The ^{32}Si nuclei were stopped in the middle of a 247 mg/cm^2 aluminum foil at the end of the stack. The intensity of the primary beam from the cyclotron was reduced, and the purity of the secondary beam was monitored every 1.5 h. A total of $(7.4\pm0.4)\times10^8$ ^{32}Si nuclei was implanted during a period of approximately 36 h. To verify the counting efficiency needed for activity measurements, two additional samples were similarly prepared with $(8.8\pm0.4)\times10^6$ and $(9.0\pm0.4)\times10^6$ ^{32}P nuclei. The tuning and isotopic resolution of the A1200 device was significantly better during this run than in first experiment, and consequently the number of contaminant ions was insignificant.

The ^{32}Si sample along with a background foil and the two ^{32}P samples were counted in the off-line detector system described above. The ^{32}P nuclei were implanted about 30 mg/cm^2 from the middle of the 247 mg/cm^2 Al foil, resulting in an asymmetry in the number of counts observed in the two SSD's. The effect of this asymmetry was explored by wrapping a ^{32}P source with aluminum foils of different thickness and determining the change in number of counts in the detectors. The results showed that, for a fixed total thickness of Al foils on both sides of the source, the sum of the counts in the two detectors is independent of the asymmetry in the thickness of Al foils on either side of the source to within 6%. Thus the

counting efficiency of the two detectors was determined to be 0.40 ± 0.04 for the second implantation. The uncertainty in the efficiency includes the uncertainty in the number of ^{32}P nuclei in the samples of 5%, the correction for the asymmetry in the implantation of approximately 6%, and the uncertainty in determining the half-life of ^{32}P from the measured decay curve of 5%, as well as the statistical uncertainties in the counting, associated with changing the threshold settings and background subtraction, of less than 3%. The calculated rate of ^{32}Si decay in the second sample was found to be 7.2 ± 0.8 per minute.

III. RESULTS

The half-life of ^{32}Si is easily calculated from the number of nuclei in the sample and their decay rate because

$$T_{1/2} = \ln 2 \left(\frac{N}{A} \right), \quad (1)$$

where N is the measured number of atoms and A is the activity of the sample. The first implantation gave a calculated half-life of 128 ± 20 yr and the second gave 134 ± 16 yr. Since the samples and calibrations of the two experiments are largely independent of each other, the average result of this work should be taken as 132 ± 13 yr. This value is in excellent agreement with all of the previous AMS measurements shown in Fig. 1, particularly with that of Hofmann *et al.* [12], but surprisingly not with the most recent AMS work [13]. The present result is also consistent with the limit set from the oceanic cycles [7], but not with the decay experiment which has the smallest uncertainty [11].

IV. CONCLUSION

We have presented a new method for the production and measurement of the half-life of ^{32}Si and the results from two such measurements. The result agrees with most of the AMS experiments, but disagrees with two older geophysical measurements and a direct decay measurement. The main sources of uncertainties in the present method are the uncertainty in the number of implanted ions and the efficiency determination of the counting system. It should be noted that any uncertainty in the amount of implantation enters twice, as it affects both the number of ^{32}Si nuclei and the number of ^{32}P (^{31}Si in the first experiment) nuclei needed for the efficiency determination. Clearly, direct identification of all of the implanted nuclei would improve the measurement. In addition, the large uncertainty in the efficiency included a correction for the asymmetry in the implantation position of ^{32}P and its decay. More precise control of the implantation and a more accurate determination of the efficiency could significantly reduce the uncertainty in these measurements. Future application of projectile fragment implantation to half-life determinations may be useful for other relatively long-lived isotopes such as ^{44}Ti, ^{59}Ni, and ^{60}Fe.

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