

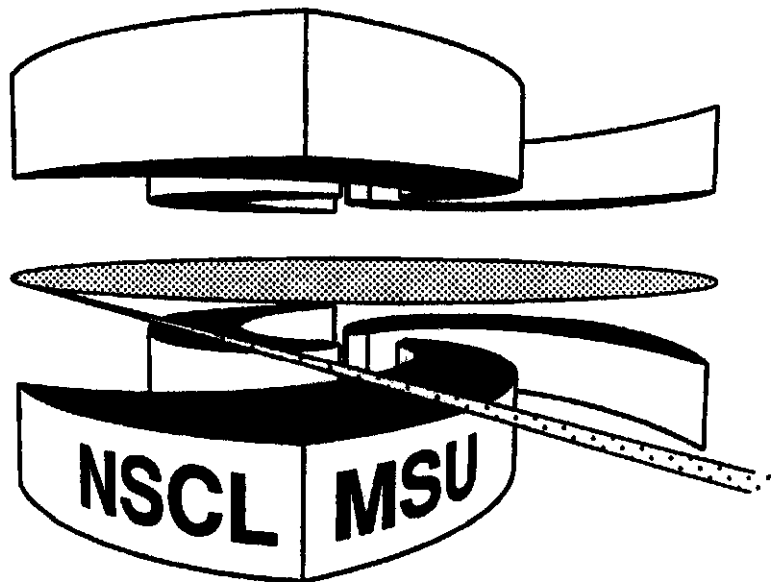


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**HALF LIFE OF  $^{32}\text{Si}$**

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Half-life of  $^{32}\text{Si}$ 

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Previous measurements of the half-life of  $^{32}\text{Si}$ , which is produced by cosmic ray spallation of  $^{40}\text{Ar}$  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  $6.7 \times 10^8$  and  $7.4 \times 10^8$   $^{32}\text{Si}$  nuclei, respectively, were prepared by implantation of separated projectile fragments. The half-life of  $^{32}\text{Si}$  was calculated from the two independent measurements of the specific activity to be  $132 \pm 13$  years.

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## I. INTRODUCTION

The isotope  $^{32}\text{Si}$  is produced in the earth's atmosphere by cosmic ray spallation of  $^{40}\text{Ar}$  and is removed from the atmosphere by aerosols. Although  $^{32}\text{Si}$  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 years[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 figure 1, there have been a number of measurements of the  $^{32}\text{Si}$  half-life[1, 3, 5, 8, 9, 11-13, 10], 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 the '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  $^{32}\text{Si}$  several years of continuous counting are needed to make an precise measurement. For the result of the Decay measurement shown in figure 1[11], the activity of a  $^{32}\text{Si}$  sample was measured over a period of four years, during which the decay rate of the  $^{32}\text{Si}$  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 figure 1 used this approach, with the exception of the measurement of Thomsen et al.[13]. In the

AMS measurements,  $^{32}\text{Si}$  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  $^{32}\text{Si}$  atoms to all silicon atoms,  $^{32}\text{Si}/\text{Si}$ , in the resulting samples was measured by Accelerator Mass Spectrometry, and the number of  $^{32}\text{Si}$  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 figure 1. Measurements of the depth distribution of cosmogenic  $^{32}\text{Si}$  in an ice core by Clausen et al.[1] and a similar measurement by DeMaster et al.[3] in sediment cores relied on the Decay method. Somayajulu et al[5]. determined the lower limit for the half-life with the Equilibrium method from a comparison of the global  $^{32}\text{Si}$  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  $^{32}\text{Si}$  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. $^{32}\text{Si}$ production

In the present measurement,  $^{32}\text{Si}$  was produced by fragmenting an  $E/A = 65$  MeV  $^{40}\text{Ar}$  beam in a  $235 \text{ mg/cm}^2$  Be target. 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  $^{32}\text{Si}$  ions from other reaction fragments[14]. The thickness of the target was chosen to maximize the yield of  $^{32}\text{Si}$ . The momentum-loss degrader was a  $150\text{ mg/cm}^2$  thick Al foil shaped to maintain the achromaticity of the A1200 device for  $^{32}\text{Si}$  nuclei[15]. Due to the relatively long half-life of  $^{32}\text{Si}$ , the number of  $^{32}\text{Si}$  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 8-10k  $^{32}\text{Si}$  ions per second at a primary beam current of 40–50 enA, 24 hours of continuous implantation were needed. The total number of implanted ions created sufficient radiation damage 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  $^{32}\text{Si}$  nuclei were implanted into a  $123\text{ mg/cm}^2$  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 figure 3. The mesh was uniformly perforated with 0.34 mm diameter holes with an inter-hole distance of 1 mm. Ions that passed through the holes would not stop in the collector foil but rather in the  $100\text{ }\mu\text{m}$  thick SSD. The thickness of the mesh was such that those  $^{32}\text{Si}$  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  $123\text{ mg/cm}^2$  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  $^{32}\text{Si}$  beam, the Si detector was not expected to fail during the implantation and could be used to continuously monitor the number of  $^{32}\text{Si}$  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  $6.7 \pm 0.7 \times 10^8$   $^{32}\text{Si}$  nuclei was implanted along with some contaminants ( $^{33}\text{P}$   $6.0 \pm 0.7 \times 10^7$ ,  $^{32}\text{P}$   $5.8 \pm 0.6 \times 10^5$ ,  $^{31}\text{Si}$   $4.2 \pm 0.4 \times 10^6$ , and  $^{28}\text{Mg}$   $3.6 \pm 0.4 \times 10^5$ ) during a 24 hour run. All of these isotopes have short half-lives compared to that of  $^{32}\text{Si}$ . One of the contaminant ions,  $^{31}\text{Si}$ , 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 silicon surface barrier detectors (SSDs) placed at opposite sides of the foil. The SSDs were 500  $\mu\text{m}$  fully depleted detectors which were calibrated using the 122 keV line of  $^{57}\text{Co}$  and the 80 and 356 keV lines of  $^{133}\text{Ba}$ . A pulser was used for calibration at higher energies. The calibration was checked periodically using both the sources and pulser. The SSDs 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  $\approx 1900$   $\text{cm}^3$ . Nitrogen gas was circulated inside the copper shielding to maintain a stable temperature. The typical background measured by the SSDs was approximately 2.0 counts/minute 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  $^{32}\text{Si}$  has a beta decay Q-value of only 0.225 MeV it is more convenient to measure the decay rate of the daughter activity.  $^{32}\text{P}$  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  $^{32}\text{S}$  by  $\beta^-$  emission[16]. In order to determine the decay rate of  $^{32}\text{Si}$  by observation of  $^{32}\text{P}$ , 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  $^{31}\text{Si}$  was used to determine the detection efficiencies for the two SSDs for the first implantation ( $^{31}\text{Si}$  has a half-life of 2.622 hours and its beta decay Q-value of 1.48 MeV is close to that of  $^{32}\text{P}$ . [16]). Approximately  $4.2 \times 10^6$   $^{31}\text{Si}$  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 hours of counting came from the decay of  $^{31}\text{Si}$ . The total efficiency obtained for the two detectors was  $0.48 \pm 0.05$ . The uncertainty in the efficiency arose from the 10 % uncertainty in the number of  $^{31}\text{Si}$  nuclei, an estimated 5 % error due to  $\beta$  spectral difference between  $^{31}\text{Si}$  and  $^{32}\text{P}$ , and the 1 % uncertainty from counting and background subtraction. The calculated rate of  $^{32}\text{Si}$  decay in this sample was found to be  $6.9 \pm 0.8$  decays per minute.

### C. Experiment 2

In the second experiment, an addition was made to the A1200 device to determine the number  $^{32}\text{Si}$  ions. A set of four PIN 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 PIN 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  $^{32}\text{Si}$  ions to the sum of the counts in the four PIN 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  $^{32}\text{Si}$  nuclei were stopped in the middle of a  $247 \text{ mg/cm}^2$  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 hours. A total of  $7.4 \pm 0.4 \times 10^8$   $^{32}\text{Si}$  nuclei was implanted during a period of approximately 36 hours. To verify the counting efficiency needed for activity measurements, two additional samples were similarly prepared with  $8.8 \pm 0.4 \times 10^6$  and  $9.0 \pm 0.4 \times 10^6$   $^{32}\text{P}$  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  $^{32}\text{Si}$  sample along with a background foil and the two  $^{32}\text{P}$  samples were counted in the off-line detector system described above. The  $^{32}\text{P}$  nuclei were implanted about  $30 \text{ mg/cm}^2$  from the middle of the  $247 \text{ mg/cm}^2$  Al foil resulting in an asymmetry in the number of counts observed in the two SSDs. The effect of this asymmetry was explored by wrapping a  $^{32}\text{P}$  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 the the source to within 6%. Thus, the counting efficiency of the two detectors was determined to be  $0.40 \pm 0.04$  for the second implantation. The uncertainty in the efficiency includes the uncertainty in the number of  $^{32}\text{P}$  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  $^{32}\text{P}$  from the measured decay curve 5 %, as well as the statistical uncertainties in the counting which associated with changing the threshold settings and background subtraction of less than 3%. The calculated rate of  $^{32}\text{Si}$  decay in the second sample was found to be  $7.2 \pm 0.8$  per minute.

### III. RESULTS

The half-life of  $^{32}\text{Si}$  is easily calculated from the number 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  $128 \pm 20$  years and the second gave  $134 \pm 16$  years. 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  $132 \pm 13$  years. 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 the smallest uncertainty[11].

#### IV. CONCLUSION

We have presented a new method for the production and measurement of the half-life of  $^{32}\text{Si}$  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 effects both the number of  $^{32}\text{Si}$  nuclei and the number of  $^{32}\text{P}$  ( $^{31}\text{Si}$  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  $^{32}\text{P}$  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 relative long lived isotopes such as  $^{44}\text{Ti}$ ,  $^{59}\text{Ni}$ , and  $^{60}\text{Fe}$ .

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## FIGURES

FIG. 1. Summary of previously reported half-life measurements of  $^{32}\text{Si}$ . 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[10].

FIG. 2. A schematic drawing of the A1200 analyser[14].

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

