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^{31}Si(2.6 h)(n,γ)^{32}Si cross section measured by accelerator mass spectrometry

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Radioactive ^{32}Si was produced by double neutron capture in natural silicon. From a measurement of the isotopic ratio ^{32}Si/^{30}Si by means of accelerator mass spectrometry the neutron-capture cross section for radioactive ^{31}Si ($T_{1/2}=2.622$ h) was deduced to be 73±6 mb.

Neutron activation cross sections are usually determined by measuring the activity of the product formed in a known amount of target after irradiation. However, the half-life of the radioactive product must also be known. Previous attempts to obtain a neutron-capture cross section of ^{31}Si(n,γ)^{32}Si have been of limited value because the half-life of ^{32}Si is disputed. The half-life is probably in the range of 100–200 yr.[1–4] Fortunately, the recent development of accelerator mass spectrometry (AMS) has changed the situation. This technique has made it possible to detect ultralow concentrations of long-lived nuclides including ^{32}Si (Ref. 5). Thus, neutron cross sections may now be determined simply from measuring the isotopic ratio of product to target nuclei without any knowledge of the half-life of the product. In the present paper we apply the method to the reaction ^{31}Si(n,γ)^{32}Si. The nucleus ^{32}Si has a special interest since it is cosmogenic: It is produced in the atmosphere as a result of cosmic-ray interactions with argon nuclei and transferred to the terrestrial environment by rain and snow. Cosmogenic ^{32}Si has found several applications as a tracer in studies of geochemical and cosmochemical processes (e.g., Refs. 6 and 7).

We have produced ^{32}Si by double neutron capture in ^{30}Si in a long bombardment where the intermediate product is saturated:

$$^{30}\mathrm{Si}(n,\gamma)^{31}\mathrm{Si}(T_{1/2}=2.622\ \mathrm{h})(n,\gamma)^{32}\mathrm{Si}\ .$$

Since the cross section of ^{30}Si and the half-life of ^{31}Si are well known, the cross section of ^{31}Si can be deduced from the ^{32}Si/^{30}Si ratio once the integrated neutron flux is known.

The targets, two disks (6 mm diam, 1 mm thick) of a silicon crystal with a natural isotopic abundance, were irradiated in the DR-3 reactor at Research Center Risø, Denmark, for 23 d in a neutron flux of 1.35×10^{14} cm^{-2}s^{-1} (thermal) and 0.12×10^{14} cm^{-2}s^{-1} (epithermal). As a control, cobalt flux monitors were irradiated simultaneously. The ^{60}Co activity was measured with an absolute-calibrated Ge(Li) detector. By using thermal and epithermal cross sections of 37 and 74 b, respectively, for the reaction ^{59}Co(n,γ)^{60}Co we obtained good agreement (within the total experimental error of 3%) with the neutron flux reported by the Risø Laboratory.

The AMS measurements were carried out at the EN-tandem accelerator facility at the University of Aarhus. Prior to the measurements, the neutron-irradiated silicon disks were mounted in sample holders for the negative-ion sputter source without any chemical pretreatment. ^{32}Si was injected as the molecular ion ^{32}SiH$_3{}^-$ since this avoids interference from the stable isobar ^{32}S (Ref. 5). At the high-energy end of the accelerator the charge state 7+ of the silicon ions was selected. The total efficiency for detecting ^{32}Si in a sample can be estimated as follows: About 1% of the silicon is sputtered as Si$^-$. The efficiency of producing ^{32}SiH$_3{}^-$ relative to ^{32}Si$^-$ is ~4%. The transmission through the tandem accelerator is about 50% and the efficiency for the charge state 7+ is 20% when foil stripping (3-μg/cm^2 carbon foil) is applied. This yields an overall efficiency for detecting ^{32}Si in a sample of 4×10^{-5}.

During a measuring cycle, ^{32}Si counts were accumulated in the ΔE-E detector for periods of 20 min. The total counting time on ^{32}Si during all runs on the irradiated samples was 16 h. After each accumulation period on ^{32}Si the currents of the stable isotopes ^{29}Si and ^{30}Si were measured for about 10 s in the Faraday cups of the AMS system. It was chosen to measure both stable isotopes in order to get an important check of the stability of the system and obtain independent information on the isotopic fractionation.

Figure 1 shows a two-dimensional mass-32 spectrum of the total energy (E_T) versus the energy loss (ΔE) of the ions injected as mass-35 accumulated during one run (about 5 h) on a neutron-irradiated silicon sample. The ^{32}Si events are well separated from the other peaks. Figure 2 shows a spectrum of a silicon blank sample recorded in 1 h. The area marked by dashed lines is the expected position of ^{32}Si. No counts are actually observed. This result corresponds to a detection limit of ^{32}Si/Si $<4\times10^{-15}$.

The isotopic ratios were measured in three independent runs. The average ^{32}Si/^{30}Si ratio was found to be $(4.23\pm0.22)\times10^{-12}$, where the quoted error is the statistical error due to counting statistics for ^{32}Si. This ratio has to be corrected for isotopic fractionation of the ions in the stripping process due to the velocity difference of the isotopes at the stripping foil. The correction factor which should be multiplied on the measured ^{32}Si/^{30}Si ratio to give the proper ratio in the sample is found to be 1.08±0.02. The correction was determined by a measurement of the isotopic abundance of the three stable

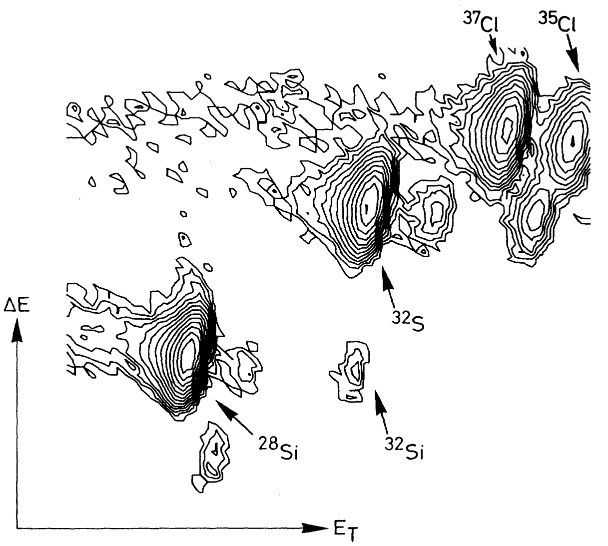

FIG. 1. Two-dimensional mass-32 contour plot of the total energy (E_T) versus the energy loss (ΔE) of the ions injected as mass 35 in a run on a neutron-irradiated silicon sample. An identification of the main peaks in the spectra is also shown.

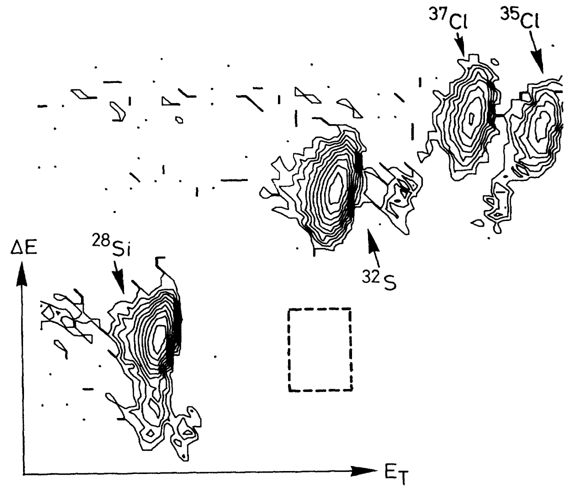

FIG. 2. Two-dimensional mass-32 contour plot of E_T vs ΔE of the ions injected as mass 35 during a 1-h run with a blank silicon sample in the ion source. The area marked by dashed lines is the expected position of ^{32}Si. No counts are observed.

isotopes ^{28}Si, ^{29}Si, and ^{30}Si. There is good agreement with the value of 1.09 expected from charge state distributions measured by Hofmann *et al.*[8] The corrected ratio in the sample is

$$^{32}\mathrm{Si}/^{30}\mathrm{Si}=(4.57\pm0.27)\times10^{-12} .$$

The ratio of ^{32}Si to stable silicon is $^{32}\mathrm{Si}/\mathrm{Si}=(1.42\pm0.09)\times10^{-13}$. This value is well above the upper limit of the background measured with the blank sample.

By using the well established half-life of ^{31}Si of 2.622 h and a thermal neutron-capture cross section in ^{30}Si of 0.107(2) b one calculates the cross section for the reaction $^{31}\mathrm{Si}(n,\gamma)^{32}\mathrm{Si}$ to be 73±6 mb.

No previous experimental determination of this cross section has been found in literature. Nevertheless, two values are quoted in tables. In Ref. 9, a value of 0.47 b is reported as personal communication, whereas Ref. 10 gives a value of 0.18 b by using the ^{32}Si half-life of 105 yr. These determinations were based on activity measurements, in which the measured quantity is the ratio $T_{1/2}(^{32}\mathrm{Si})/\sigma(^{31}\mathrm{Si})$. In both cases the value of $\sigma(^{31}\mathrm{Si})$ was obtained from a measured ratio of $T_{1/2}(^{32}\mathrm{Si})/\sigma(^{31}\mathrm{Si})=600$ yr/b by applying the most recent value of $T_{1/2}(^{32}\mathrm{Si})$ at the time of publication. Turkevich and Samuels[11] determined this ratio from a measurement of the ^{32}P daughter activity in neutron-irradiated quartz after a cooling period of 2 yr. However, they used a thermal neutron-capture cross section in ^{30}Si of 0.2 b, which is a factor of 2 larger than the value applied today.[10] In a similar experiment Roy and Yaffe[12] found an amount of ^{32}P so small that only a maximum value could be attributed to this nuclide; they quoted 600 yr/b as a minimum value.

The new value of the $^{31}\mathrm{Si}(n,\gamma)^{32}\mathrm{Si}$ cross section measured in the present work makes it possible to determine the half-life of ^{32}Si provided that either the absolute ^{32}Si activity or the activity ratio of ^{32}Si to ^{31}Si could be measured in a silicon target after a long neutron bombardment. Such an experiment is called for since none of the neutron-irradiation experiments done so far are considered precise enough to deduce a meaningful half-life for ^{32}Si.

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