Measurement of ^{26}Al in Terrestrial Silicate Rock, Revisited

Seiichi Shibata,*,a Yasuhiro Okuda,a Koichi Takamiya,a Yuichi Oki,a Takayuki Sasaki,b Hiroyuki Matsuzaki,c and Koh Sakamotod

aResearch Reactor Institute, Kyoto University, Kumatori-cho, Sennan-gun, Osaka 590-0494, Japan
bDepartment of Nuclear Engineering, Kyoto University, Sakyo-ku, Kyoto 606-8501, Japan
cDepartment of Nuclear Engineering and Management, School of Engineering, The University of Tokyo, Bunkyo-ku,
 Tokyo 113-0032, Japan
dDepartment of Chemistry, Faculty of Science, Kanazawa University, Kakuma, Kanazawa 920-1192, Japan

Cosmic-ray-induced ^{26}Al in terrestrial silicate rock was remeasured by accelerator mass spectrometry (AMS) in order to confirm the value reported more than 30 years ago. The result of $(5.2 \pm 1.0) \times 10^6$ ^{26}Al atoms/g SiO$_2$ for the surface rock sample was consistent with the previous result determined by a low background $\gamma-\gamma$ coincidence spectrometry system within experimental uncertainties. The lower amount of ^{26}Al was found in the deeper part of the rock samples. From the result, the surface exposure age and the erosion rate at the sampling location were estimated to be 1.4×10^5 y and 3.7×10^{-4} cm/y, respectively.

1. Introduction

^{26}Al ($T_{1/2} = 7.2 \times 10^5$ y) observed in terrestrial and extraterrestrial matters provides one of the important clues to the deciphering fossil records stored in those materials and allows investigations of their irradiation history by cosmic radiation.

In their pioneering work in 1968, Tanaka et al.[1,2] searched for ^{26}Al produced in terrestrial silicate rock (chert) by the interaction of muons with ^{28}Si, which is the most abundant isotope in the earth's crust except for ^{16}O, in order to investigate the secular variation of cosmic radiation intensity. They chemically extracted and purified aluminum from several tens of kg of silicate rock collected from an area with a low erosion rate. The aluminum separated was ignited to Al$_2$O$_3$. Each 100 g of the Al$_2$O$_3$ was pressed into a disk of 7 cm in diameter and 2 cm thick. The annihilation γ rays from the decay of ^{26}Al in these samples were measured using a low-background $\gamma-\gamma$ coincidence spectrometer consisting of two 7.6 × 7.6 cm NaI(Tl) crystals installed in the underground counting facility.[3] Unfortunately, they were not able to obtain the positive results, and reported the ^{26}Al activities of 0.02 ± 0.12 dpm/10 kg SiO$_2$ for the surface rock and of 0.00 ± 0.08 dpm/10 kg SiO$_2$ for the sample collected at a depth of 24 meters of water equivalent (mwe).

In 1975, Hampel et al.[4] measured again the same sample prepared by Tanaka et al.[1] by means of a low-level $\gamma-\gamma$ coincidence spectrometry system consisting of two 15 × 10 cm NaI(Tl) crystals with improved statistics. For the measurement, four original disks of the sample 1A prepared from the surface rock by Tanaka et al.[1] was repressed into a larger disk (a 402 g disk of Al$_2$O$_3$) of 15 cm in diameter and 1.65 cm thick. The result was reported to be 0.0068 ± 0.0028 dpm ^{26}Al/kg SiO$_2$. Based on the result, they discussed the muon flux during the last several million years and the erosion rate at the collecting location of the measured sample. In the conclusion of their paper, a more quantitative measurement in the future was expected for wide applications of cosmic-ray-produced long-lived nuclides in geology, geophysics, and cosmic ray physics.

For trace analyses of long-lived nuclides, accelerators have now extensively been applied as high-energy and extremely high-sensitive mass spectrometers, accelerator mass spectrometry (AMS). By the advances in AMS, it became possible to apply cosmogenic radionuclides produced *in situ* in terrestrial samples for studies of erosion and sedimentation rates and surface exposure ages. Recently, quartz, which is geologically abundant mineral, is used as one of the ideal samples for these studies.

We have applied the AMS method to measure the trace amount of cosmogenic radionuclides produced in terrestrial and extraterrestrial substances. In the course of this study, ^{26}Al induced by cosmic ray secondaries in terrestrial silicate rock was measured in order to confirm the result reported by Hampel et al.[4] While the large amount of the rock sample is needed for the radioactivity measurement of the cosmogenic radionuclides, only a few grams of the rock is sufficient to determine ^{26}Al by AMS.

In this paper, we report the result for the AMS measurement of ^{26}Al in the terrestrial silicate rock collected at the surface and deeper parts of rock in the same sampling place as reported in Tanaka et al.[1]

2. Experimental Procedure

A sample of silicate rock was collected at an altitude of about 500 m in Kutami, Yaotsu-cho, Kamo-gun, Gifu-prefecture, Japan. Details of the sampling place were already reported by Tanaka et al.[1,2] In the first sampling in December, 2000, samples were collected at the surface, 2.5 mwe and 12 mwe of rock, and in the second sampling in June, 2003, samples at the surface, 2.5 mwe, 5 mwe, and 12 mwe.[5]

The chemical separation method of aluminum from large quantities of the rock samples (> 10 kg) developed by Tanaka et al.[1] was improved for the AMS measurement in this work.[5] The rock sample was crushed and pulverized after the intruded clay was removed by brushing. 0.2 – 1.5 g of the powdered sample was dissolved in mixed acid solution of HNO$_3$, HF, and HClO$_4$ by heating under pressure. The solution was evaporated in order to volatilize silicon as SiF$_4$. The residue was dissolved in concentrated H$_2$SO$_4$ and evaporated to dryness again in order to decompose Al-Si complex and to expel the excess HF. Then the residue was leached with concentrated HCl, and the solution obtained was evaporated. Finally the residue was dissolved in H$_2$O.

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*Corresponding author. E-mail: shibata@rri.kyoto-u.ac.jp.
FAX: +81-72-451-2632.

In the obtained solution, the hydroxide was precipitated with NaOH solution at pH = 5 – 7 after adding NH_4Cl as a buffer solution. After centrifuging, the precipitate was suspended in NaOH solution. The supernatant solution was adjusted to pH = 5 – 7 by adding HCl. Then the white precipitate of aluminum hydroxide was obtained.

The aluminum hydroxide was dissolved in HCl. The aluminum was purified by cation exchange with 1.75 M HCl.[5] The aluminum hydroxide was precipitated again from the eluted aluminum fraction by adding NaOH solution. The precipitate of aluminum hydroxide was washed by pure water, and dissolved in HCl. The solution was evaporated, and heated to 900 °C in an electric furnace. The white residue of Al_2O_3 obtained was used for AMS.

The aluminum contents in the rock samples were determined by instrumental neutron activation analysis (INAA) using Kyoto University Reactor (KUR). About 2 mg of the rock samples were irradiated for 30 s at the neutron flux of 2.34×10^{13} n/cm^2 s together with the aluminum standard samples. After irradiation, 1779 keV γ-ray emitted from ^{28}Al ($T_{1/2}$ = 2.25 min) produced by ^{27}Al(n,γ) reaction was measured by γ-ray spectrometry. The aluminum contained in the rock sample was used as a carrier in the chemical separation mentioned above.

The isotopic ratio of ^{26}Al/^{27}Al was determined by the AMS system using the 5 MV tandem Van de Graaff accelerator at MALT (Micro Analysis Laboratory, Tandem accelerator), the University of Tokyo.[6] Negative ions of ^{26}Al$^-$ and ^{27}Al$^-$ produced in the ion source by sputtering Cs$^+$ ions were injected alternatively to the accelerator. At the terminal of the tandem accelerator, negative ions were converted into positive ions. The ^{27}Al^{3+} beam was measured by the off set Faraday cup outside the main course of the accelerator, and the ^{26}Al^{3+} ions were finally detected by a $\Delta E - E$ counter.[7] The measurements were carried out with a standard, whose ratio of ^{26}Al/^{27}Al is 5.1×10^{-11}, for the normalization of the data, chemically prepared samples from the rock and a blank sample. The detection limit of ^{26}Al/^{27}Al in this AMS system is estimated to be $\lesssim 5 \times 10^{-15}$ at present.

TABLE 1: Al Content Determined by Neutron Activation Analysis

Sample	Depth (mwe*)	Aluminum content** (%)
1-001201	surface	0.70 ± 0.01
2-001201	2.5	6.49 ± 0.07
3-001201	12	1.40 ± 0.02
1A-030620	surface	0.87 ± 0.01
1B-030620	surface	2.94 ± 0.02
2-030620	2.5	6.96 ± 0.04
3-030620	5	6.93 ± 0.05
1A-1968***	surface	0.74

*mwe: meter of water equivalent
**uncertainty: 1 σ
***1A-1968: Tanaka et al. (Ref.1)

TABLE 2: ^{26}Al Measured in Silicate Rock Samples

Sample	Depth (mwe)	^{26}Al atoms / g SiO$_2$ ($\times 10^6$)
1-001201	surface	4.6 ± 1.2
2-001201	2.5	1.7 ± 1.3
3-001201	12	0.8 ± 0.5
1A-030620	surface	6.4 ± 2.3
1B-030620	surface	9.4 ± 4.5
2-030620	2.5	< 2.2
3-030620	5	< 1.9

3. Results and Discussion

The aluminum contents determined by neutron activation analysis are summarized in Table 1. The experimental uncertainties attached to the aluminum contents refer only to those statistically estimated in γ-ray counting. The aluminum contents of the surface rock samples of 1-001201 and 1A-030620 are consistent with that determined by Tanaka et al.,[1] which is also included in Table 1, while the aluminum contents of rock samples collected at the 2.5 and 5 mwe of rock are higher than those at the surface and 12 mwe.

In the AMS measurement, more than three runs for each sample were carried out. Examples of the AMS spectra measured for the standard and the surface sample by using a $\Delta E - E$ counter are shown in Figures 1 and 2, respectively. There are almost no signals induced by impurity ions in the chemically separated sample as shown in Figure 2. For the surface samples, the ratios of ^{26}Al/^{27}Al were measured to be $(1 - 7) \times 10^{-14}$, while those for the deeper samples were less than 10^{-14}. The ratios for the blank were measured to be $(2.2 - 2.6) \times 10^{-15}$. In the case of Figure 2 for the surface sample (1-001201), the measured ratio of ^{26}Al/^{27}Al was $(2.7 \pm 0.5) \times 10^{-14}$.

From the both results of INAA and AMS, the numbers of ^{26}Al atoms produced in the rock samples were estimated and summarized in Table 2. The ^{26}Al atoms determined in three surface samples agreed with one another within experimental uncertainties. Thus we estimated to be $(5.2 \pm 1.0) \times 10^6$ ^{26}Al atoms/g SiO$_2$ as the weighted mean for the surface samples collected in 2000 and 2003. This value was consistent with 0.0068 ± 0.0028 dpm ^{26}Al/kg SiO$_2$ (($3.7 \pm 1.5) \times 10^6$ ^{26}Al atoms/ g SiO$_2$) reported more than 30 years ago.[4] For the deeper mwe
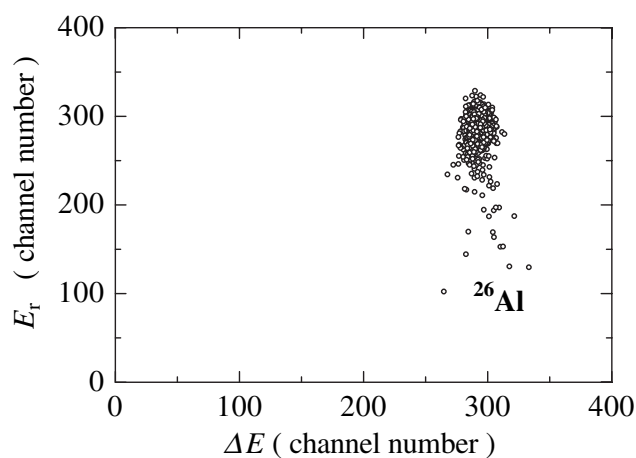

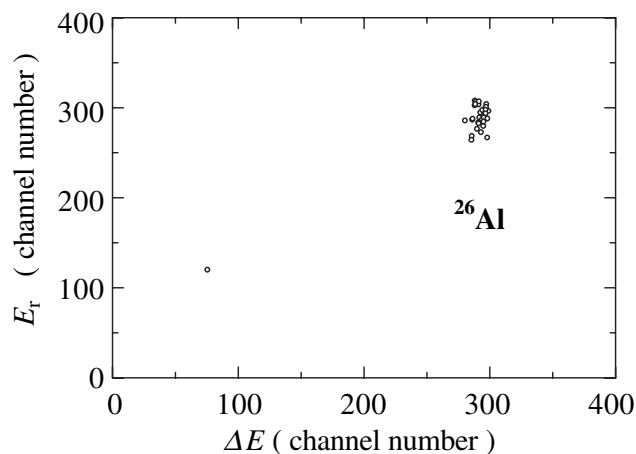**Figure 1.** Two-dimensional plot of ^{26}Al measurement for the standard. (Measurement time: 200 s)

Figure 2. Two-dimensional plot of ^{26}Al measurement for the surface rock sample (1-001201). (Measurement time: 3100 s) For this run, the measured ratio of ^{26}Al/^{27}Al is $(2.7 \pm 0.5) \times 10^{-14}$.

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samples, the measured ratios of ^{26}Al/^{27}Al were around the detection limit of this AMS system. Therefore large uncertainties were estimated for those results, and only upper limits of the ^{26}Al were obtained in the samples of 2-030620 (2.5 mwe) and 3-030620 (5 mwe).

The production rate of ^{26}Al at the sampling location can be calculated to be 38 ± 3 ^{26}Al atoms/g SiO$_2$ y using the altitude-dependent scaling factors given by Stone,[8] which is consistent with those calculated by Heisinger et al.[9] and Heisinger and Nolte.[10] From the production rate obtained and the value of ^{26}Al atoms estimated for the surface sample, the surface exposure age was estimated to be $(1.4 ± 0.3) × 10^5$ y at the sampling location. Based on the result, the erosion rate was calculated to be $(3.7 ± 0.9) × 10^{-4}$ cm/y using the following equation by Lal,[11]

$$\varepsilon = 1/\mu\,[1/T_{ex} - \lambda], \qquad (1)$$

where ε is the erosion rate, $1/\mu$ the mean cosmic-ray absorption length, T_{ex} the effective surface exposure age and λ the decay constant of ^{26}Al. In the calculation, it is assumed that the rock surface undergoes steady state erosion and the radionuclide concentration was zero when the rock was deep seated and shielded from cosmic radiation. The erosion rate calculated is consistent with the geological estimate for the erosion rate of < 10 m/10^6 y reported in Tanaka et al.[1,2]

4. Conclusion

Cosmogenic ^{26}Al in terrestrial silicate rock collected in the same sampling place where Tanaka et al.[1,2] collected the rock samples was remeasured by AMS in order to confirm the previous result measured by a low background γ–γ coincidence spectrometry more than 30 years ago.[4] The chemical separation method developed by Tanaka et al.[1,2] was improved for this work. The AMS result for the surface sample was obtained to be $(5.2 ± 1.0) × 10^6$ ^{26}Al atoms/g SiO$_2$, which was consistent with the previous result by the radioactivity measurement of $(3.7 ± 1.5) × 10^6$ ^{26}Al atoms/g SiO$_2$.[4] From the result obtained, the surface exposure age and the erosion rate at the sampling location were estimated. The erosion rate of 3.7 ± 0.9 m/10^6 y was consistent with the geological estimate of < 10 m/10^6 y.[1,2]

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