^{26}Al: Measurement and Applications [and Discussion]

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^{26}Al: measurement and applications

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^{26}Al ($\tau_{\frac{1}{2}} = 0.7$ Ma), like ^{10}Be ($\tau_{\frac{1}{2}} = 1.5$ Ma), is produced by cosmic rays in the atmosphere, but its production rate is low (*ca.* 4×10^{-3} that of ^{10}Be) and the crustal abundance of ^{27}Al is high, resulting in ^{26}Al : ^{27}Al ratios rarely exceeding 10^{-14}. Both isotopes are also produced in surficial rocks by cosmic rays; in quartz the production rates of ^{26}Al and ^{10}Be are *ca.* 70 atoms g^{-1} a^{-1} and *ca.* 10 atoms g^{-1} a^{-1}, respectively (at sea level). Because quartz frequently contains ^{27}Al at less than 100 p.p.m. (by mass), the ^{26}Al : ^{27}Al ratio resulting from this *in situ* production can be as high as 3×10^{-11}. In extraterrestrial matter, where the production rates are several hundred times greater, samples containing a few percent of ^{27}Al have ^{26}Al : ^{27}Al ratios of 10^{-11} or larger.

We describe an experimental arrangement that was developed at the University of Pennsylvania for measuring ^{26}Al : ^{27}Al ratios as low as 10^{-15}. The size requirements for samples are approximately a factor of 100000 smaller than those of decay counting, and measurement times are less than an hour, compared with weeks or months.

We discuss several applications of the technique to the measurement of terrestrial as well as extraterrestrial samples. Emphasis is placed on the combined measurement of ^{10}Be and ^{26}Al and the various uses to which the ratio of ^{26}Al : ^{10}Be in a sample can be put. In particular, we discuss the very high ^{26}Al : ^{10}Be ratios that are found in the outer few millimetres of extraterrestrial bodies, resulting from low-energy protons emitted by solar flares, and the diagnostic value of these ratios in determining the size of objects in space. The low value of the ^{26}Al : ^{10}Be ratio resulting from atmospheric production is used to prove the terrestrial origin of tektites. And finally, we discuss the manner in which the variable ^{26}Al : ^{10}Be ratio that results from *in situ* production in objects on the Earth's surface can be used to measure rates of weathering and exposure histories.

1. INTRODUCTION

^{26}Al ($\tau_{\frac{1}{2}} = 0.7$ Ma), like ^{10}Be ($\tau_{\frac{1}{2}} = 1.5$ Ma), is a radioisotope that is produced on Earth by the interaction of cosmic rays with the atmosphere and with surface rocks. Atmospheric production is low because its progenitor, ^{40}Ar, constitutes only about 1% of the atmosphere and its production in surficial rocks is also low because of the attenuation of the cosmic-ray flux by the atmosphere. It is noteworthy that the ^{26}Al : ^{10}Be production ratio is about 4×10^{-3} in the atmosphere and about 7 in high-silica rocks. This large difference can have important implications and is one of the many reasons why the study of both isotopes is so much more valuable than the study of either one alone.

The low production rate of ^{26}Al, coupled with the relatively high abundance of ^{27}Al, results in very small ^{26}Al : ^{27}Al ratios in most terrestrial samples, usually in the range 10^{-14}–10^{-15}. Not surprisingly, before the development of accelerator mass spectrometry (AMS), very few measurements were made on terrestrial samples and then only in rather special cases. For example, McCorkell *et al.* (1967) were able to measure the ^{26}Al content of Arctic ice after extracting aluminum from 1.2×10^6 kg of ice. The extraordinary sensitivity of AMS is well illustrated by the fact that we have recently made similar measurements on two samples of Antarctic ice weighing 6 and 14 kg.

Although AMS affords about a 10^5-fold improvement in sensitivity compared with decay counting, the majority of terrestrial samples, such as soils, sediments, manganese nodules, etc., still present an experimental challenge. The reason is that it is difficult to obtain much more than about 1 μA of ^{27}Al negative ions from a typical sample of aluminum oxide. For a sample with an ^{26}Al : ^{27}Al ratio of 10^{-14}, yielding 1 μA of negative ions and with an accelerator transmission of 15%, the ^{26}Al count rate is about 30 per hour.

Until recently, the measurement of terrestrial ^{26}Al was hampered by a fixation on the idea that the ^{26}Al of interest was that produced in the atmosphere. Partly as a result of an earlier collaboration with F. Yiou & G. M. Raisbeck (Yiou *et al.* 1984) and some recent work of Lal & Arnold (1985), we now perceive the measurement of terrestrial samples in a dramatically new light. What was not fully appreciated was that in certain crystals such as quartz, the ^{27}Al content can be as low as, or less than, 10 p.p.m. (by mass), so that although the *in situ* production of ^{26}Al in surficial rocks is small, even after relatively short exposure times, the ^{26}Al : ^{27}Al ratio can be substantially larger than 10^{-14}. For example, assuming an *in situ* production rate of 70 atoms g^{-1} a^{-1} at sea level and 10 p.p.m. (by mass) of ^{27}Al, the ^{26}Al : ^{27}Al ratio after 10^3 years of exposure is 3.3×10^{-13}, rising to 3.5×10^{-10} at saturation (ratios easily measured with AMS).

Production rates in extraterrestrial materials are about 10^3 times greater than in terrestrial rocks yielding typical concentrations at saturation in stony meteorites of *ca.* 3.4×10^{10} atoms g^{-1} and in the lunar surface (0–50 cm) of *ca.* 2.7×10^{10} atoms g^{-1}. Production is primarily caused by the interaction of galactic cosmic rays and their secondary particles with silicon and aluminum. Because ^{26}Al (unlike ^{10}Be), can be produced by relatively low-energy particles, the outermost few millimetres of extraterrestrial bodies have even higher concentrations of ^{26}Al as a result of solar flares. Such effects are usually not observed because in meteorites the outer skin is lost owing to ablation, and both erosion and 'gardening' serve to dilute the effect on the lunar surface.

The high ^{26}Al concentration in extraterrestrial matter can readily be measured either by decay counting or by AMS. The important difference between the two techniques is that decay counting requires at least a 5 g sample, 20 g being preferred, whereas AMS measurements can be made with less than 1 mg of sample. Thus AMS makes it possible to measure the ^{26}Al (and ^{10}Be) contents of small extraterrestrial objects such as individual lunar grains or, as in possibly one of the most outstanding achievements of AMS to date, in individual cosmic spherules having masses as small as 130 μg (Raisbeck *et al.* 1984).

2. EXPERIMENTAL TECHNIQUES

The experimental difficulties associated with measuring a particular cosmogenic nuclide vary widely and depend on several factors. Among these are the electron affinity of the negative ion, the possible existence of an isobar and whether or not it forms a negative ion, the atomic number Z of the isobar, and the mass of the cosmogenic nuclide. The last-mentioned is important because particle identification becomes more difficult with increasing mass (^{129}I is an exception because the interfering isobar, ^{129}Xe, does not form a negative ion).

According to these criteria, ^{26}Al is of intermediate difficulty. Although its electron affinity is not unusually small (0.44 eV), it is difficult to make the negative ion (a fact that is only in part attributable to the need to form it from its oxide rather than from the metal). Lack of negative-ion current is by far the largest experimental difficulty with samples having ^{26}Al concentrations of less than 10^{-13}. Fortunately, the only isobar that might interfere with the measurement of ^{26}Al is ^{26}Mg, and it does not form a stable negative ion.

Figure 1 shows the high-intensity caesium-sputter source that we developed a few years ago (Middleton 1983), and that we routinely use to generate beams of ^{27}Al$^-$ ions. The sample, which is contained in a disposable copper cathode, usually consists of 1–5 mg of compressed Al_2O_3. We normally do not mix powdered metal with the oxide unless the sample mass is less than 1 mg, and then only to increase the volume. A typical negative-ion spectrum obtained from reagent Al_2O_3 is shown in the upper half of figure 2. The 2.8 μA of ^{27}Al$^-$ ions is typical of reagent grade Al_2O_3 and is to be compared with the 1–2 μA usually obtained from samples. Also shown in figure 2 is a negative-ion spectrum obtained from MgO. Although not entirely evident from the figure, there are no peaks corresponding to Mg$^-$ ions, but there are relatively strong peaks corresponding to ^{24}MgO$^-$, ^{25}MgO$^-$, and ^{26}MgO$^-$. Unfortunately, the existence of the ^{26}MgO$^-$ prevents the use of the intense aluminum-oxide beam in ^{26}Al studies. The ionization efficiency of the source has been measured and is about 0.25% for ^{27}Al$^-$ ions.

There can be little doubt that higher ^{27}Al$^-$ currents would greatly benefit the measurement of samples with ^{26}Al concentrations of less than 10^{-13}. We are currently developing a new sputter source with a spherical ionizer that has produced currents of up to 5 μA, but it seems unlikely that significantly higher currents will be obtained. An alternative approach might be to

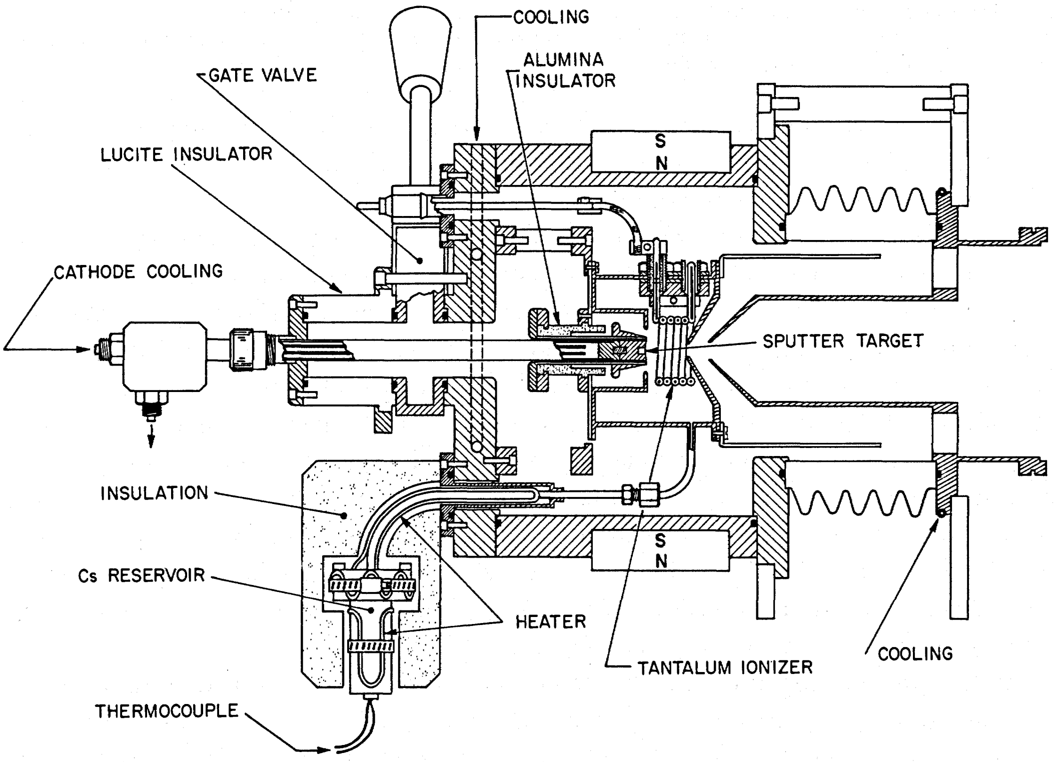

Figure 1. The high-intensity caesium sputter source used to generate ^{27}Al beams from small (1–5 mg) samples of Al_2O_3. Currents usually range from 1 to 3 μA, depending on the purity of the sample. The ionization efficiency is about 0.25%.

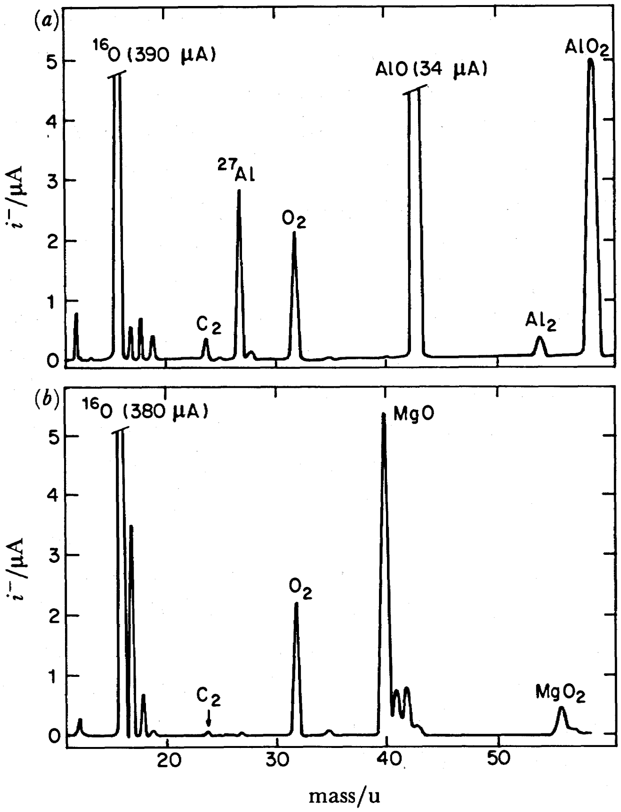

FIGURE 2. Negative-ion spectra obtained from sputter targets of reagent-grade Al_2O_3 (*a*), and MgO (*b*). The weak peak corresponding to mass = 24 in the lower spectrum is entirely due to C_2^-; magnesium does not form a negative ion. Note that the oxides of both aluminum and magnesium form quite intense negative-ion beams.

generate the negative ions by charge exchange rather than by sputtering. Heinemeier & Hvelplund (1978) have shown that up to 13% of $^{27}Al^+$ ions can be converted into negative ions when passed through a canal containing sodium vapour.

Figure 3 shows schematically the experimental arrangement that is currently used at the University of Pennsylvania to measure $^{26}Al : ^{27}Al$ ratios. Between the ion source and the detector there are four stages of analysis. Before the accelerator, there is a 90° deflecting magnet with a mass resolution, $\Delta m/m \approx \frac{1}{40}$, and after it a velocity selector of length 90 cm ($E \approx 30$ kV cm^{-1}) followed by 90° and 45° deflecting magnets. When the system is tuned for ^{26}Al, the main function of each of the various elements is as follows. (1) Low-energy magnet: selects $m = 26$ negative ions for injection into the accelerator. (2) Velocity selector: eliminates the portion of the continuous energy spectrum of ^{27}Al that has the same magnetic rigidity as the ^{26}Al ions. (3) 90° magnet: selects ^{26}Al ions with the desired energy and charge state. (4) 45° magnet: eliminates the ^{27}Al ions that have the same energy as the ^{26}Al ions (indistinguishable in the counter telescope) and that succeed in passing through the 90° magnet as a result of a charge exchange in the vacuum chamber of the magnet.

Because the transmission through our FN tandem accelerator is best at terminal voltages between 7 and 8 MV, most measurements have been made at 7.5 MV where the most probable charge state for aluminum is 7+ and the final energy is 60 MeV. At this energy and with a carbon-foil stripper, the transmission through the accelerator is about 15%.

Figure 4 shows a sectional drawing of our compact ΔE–E counter telescope. Other than the

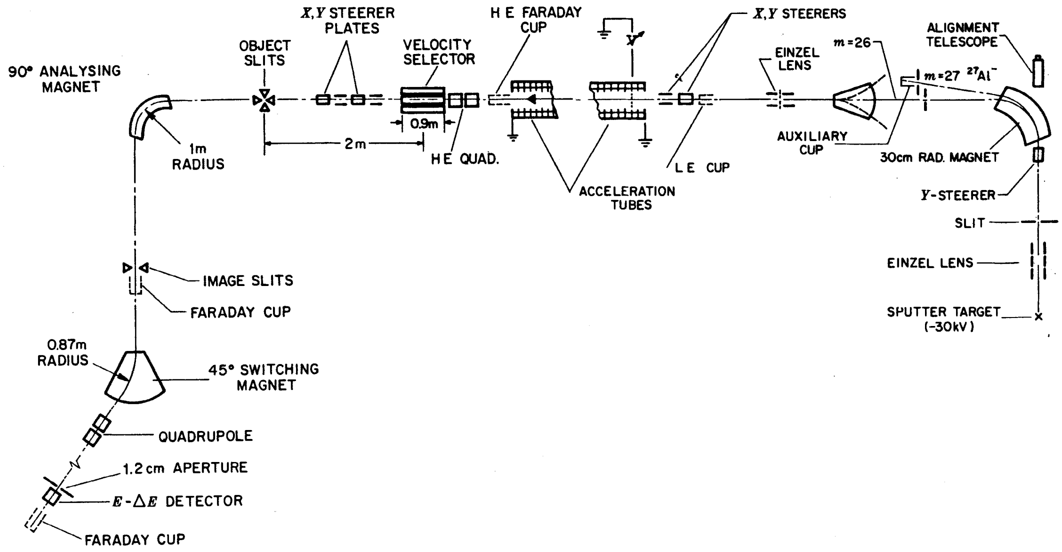

FIGURE 3. The experimental arrangement used at the University of Pennsylvania to measure ^{26}Al : ^{27}Al ratios. The tandem accelerator is usually operated at a terminal voltage of 7.5 MV and measurements are made on aluminum ions of 60 MeV, with a charge state of 7+.

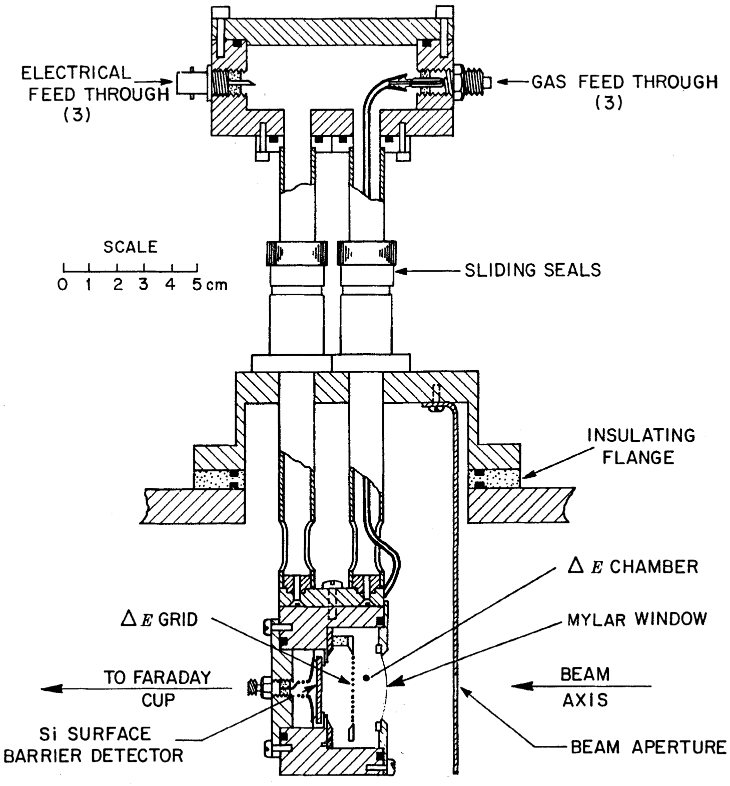

FIGURE 4. The compact ΔE–E telescope used to detect ^{26}Al and also, in slightly modified form, ^{10}Be ions. The latter requirement largely dictated the design and was responsible for the choice of a ΔE ionization chamber with an axial electric field. The acceptance aperture of the detector is 1.2 cm in diameter and the silicon surface-barrier detector has an area of 300 mm^2.

ΔE ionization chamber, which has an axial electric field, this is of fairly conventional design (for further details see Middleton *et al.* 1983). A fairly typical two-dimensional $E-\Delta E$ spectrum obtained with the system described in the foregoing is shown in figure 5. It will be noticed that the peak corresponding to ^{27}Al, having the same magnetic rigidity as the ^{26}Al, has been almost completely eliminated by the new enlarged velocity selector. The two counts below the ^{26}Al peak probably correspond to ^{26}Mg that is injected into the accelerator as ^{26}MgH$^-$.

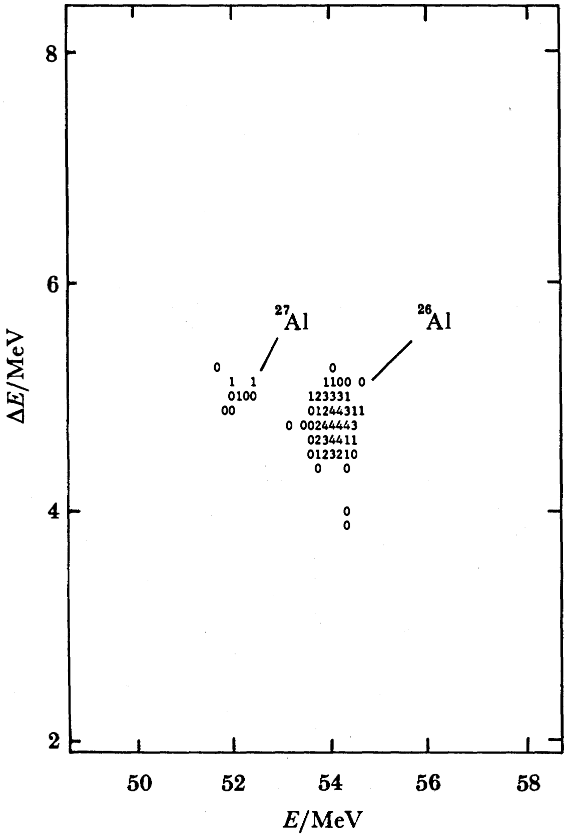

FIGURE 5. A typical particle-identification spectrum, for Libyan Desert glass, with energy loss (ΔE) plotted against total energy (E). The numbers in the spectrum are the logarithm (base 2) of the number of counts. The two counts below the ^{26}Al peak probably correspond to ^{26}Mg, injected into the tandem as ^{26}MgH$^-$. The ratio ^{26}Al : ^{27}Al = 1.5×10^{-13}, and ^{26}Al = 13.4×10^{6} atoms g^{-1}. For ^{26}Al, 312 counts were made, over a measurement time of 60 min, on a sample of mass 24.8 g.

Our procedure is to measure the transmission of ^{27}Al ions through the accelerator before and after a period of counting ^{26}Al ions. During the counting period, the ^{27}Al$^-$ current is integrated by the auxiliary Faraday cup shown in figure 3. From the known transmission and the integrated ^{27}Al$^-$ current, the ^{27}Al positive-ion current anticipated during the ^{26}Al counting period can be calculated and hence the ^{26}Al : ^{27}Al ratio determined. Depending upon the counting duration, this procedure may be repeated 2–4 times. From replicate measurements of standards, we believe that this procedure permits ^{26}Al : ^{27}Al ratios to be determined to within 7 %.

The overall efficiency of our system for detecting ^{26}Al is 3.8×10^{-4}. This implies that samples placed in the ion source must contain at least 3×10^6 atoms of ^{26}Al to obtain 1000 counts. It is difficult to quantify the minimum measurable ^{26}Al : ^{27}Al ratio because this depends on several factors, but, in most cases, we believe that a 20% measurement is possible at the 10^{-15} level.

3. Extraterrestrial samples

(a) *Introduction*

The measurement of ^{26}Al in extraterrestrial samples is comparatively straightforward because of its large concentration of about 3.4×10^{10} atoms g^{-1} at saturation (in H-type chondrites). Because this concentration results in a specific activity of about 60 disintegrations min^{-1} kg^{-1}, a fairly large number of samples have been measured by using the γ–γ coincidence method of decay counting with samples weighing 5–20 g.

The major advantages of AMS over decay counting are the ability to measure smaller samples (*ca.* 100 µg) and the shorter measurement times (*ca.* 20 min). The reduction in sample size enables measurements of ^{26}Al to be performed on the same samples as used for other types of measurements such as K–Ar ages and exposure ages determined by noble-gas concentrations and tracks. Furthermore, ^{26}Al can be measured in mineral separates of meteorites, especially in the iron phase. And, finally, the small-sample-size requirement allows measurements to be made in the top few millimetres of extraterrestrial bodies where one expects to see the contribution of solar flares.

The major disadvantage of AMS compared with decay counting is that whereas the latter is non-destructive, AMS measurements require the complete dissolution of the sample. Because ^{26}Al : ^{27}Al ratios are high (by AMS standards), *ca.* 10^{-9}, the procedure followed is similar to that used in ^{10}Be measurements. An ^{27}Al carrier of about 1 mg is added at the time the sample is dissolved, reducing the ^{26}Al : ^{27}Al ratio to *ca.* 10^{-12}. After dissolution, the Al is extracted by using ion-exchange resins, then converted to $Al(OH)_3$ with ammonia, and finally to Al_2O_3 by heating in air to about 800 °C.

(b) *Artificial meteorites*

Before one can reconstruct the exposure history of an extraterrestrial body from measurements of cosmogenic nuclides inside it, it is necessary to understand how the production of these nuclides is dependent on the meteorite's size and chemical composition. The effects of size are twofold. With increasing depth inside a body, the flux and energy of the incoming cosmic rays are attenuated. However, just beneath the surface, large numbers of secondary particles (largely neutrons) are produced that can also be effective in creating the nuclide of interest. Consequently, the rate of production for most nuclides does not decrease monotonically with distance from the surface. Quite often these secondary particles have cross sections for producing the nuclide of interest that vary greatly from one target element to another.

In an effort to quantify some of these effects, a series of modelling experiments was carried out in which granodiorite spheres of diameter 10 cm, 30 cm, and 50 cm were isotropically exposed to a proton beam of 600 MeV at CERN to simulate the 4π-irradiation of meteoroids. Short-lived and long-lived radionuclides and stable nuclides were measured in cosmochemically significant target elements placed at various locations within the spheres. (AMS was used only to measure ^{10}Be and ^{26}Al.) From these measurements, it has been possible to calculate absolute cross sections and depth profiles, and to determine the contributions of secondary and primary cosmic rays. Detailed descriptions of the experiment are contained in Englert *et al.* (1984) and Michel *et al.* (1986).

Among the significant results of this experiment were determinations of the relative cross sections for the production of ^{26}Al from Al, Si, and Mg. The ratio of the Al–Si cross section is approximately three (almost twice the literature value), and Mg seems not to contribute at all to the production of ^{26}Al, despite the suggestion that it might. The average ^{26}Al production rate in the 50 cm sphere was about 30% greater than in the 10 cm sphere, because of the contribution of secondary particles.

(*c*) *Lunar core*

To understand further the depth dependence of the cosmic-ray production of ^{26}Al, we measured the ^{26}Al in the long core from Apollo 15 (Nishiizumi *et al.* 1984*a*). This core extends from near the surface (about 1.5 cm were lost during sampling) to a depth of 2.18 m (about 390 g cm^{-2}). Although the ^{26}Al was measured in this core earlier with decay-counting techniques, the results obtained from AMS are higher in precision (and consistently lower by 10–17%). From this core, the half-attenuation length for ^{26}Al was determined to be 122 g cm^{-2} (in the 150–400 g cm^{-2} region). ^{10}Be and ^{53}Mn were also measured. The half-attenuation lengths determined for these nuclides are 120 g cm^{-2} and 123 g cm^{-2}, respectively (Nishiizumi *et al.* 1984*b*).

(*d*) *Lunar gardening*

The surface of the Moon is constantly bombarded by micrometeorites that are responsible for the production of the lunar regolith (the blanket of fragmented rock, sometimes called lunar soil, covering most of the lunar surface). These micrometeorites cause mixing of the upper few centimetres of the soil in a process that has been termed gardening. Because ^{53}Mn and ^{26}Al are both abundantly produced by solar protons in the top few millimetres, they can be used to determine the amount of mixing that has occurred at the surface. We measured the ^{26}Al, ^{53}Mn, and ^{10}Be (no appreciable surface enhancement) in eighteen rocklets from the top 12 cm of the Apollo 15011 core. Because all of the rocklets are at the near-surface saturation value for ^{10}Be, it is apparent that none have recently come from great depth. The ^{53}Mn and ^{26}Al values for the rocklets show a wide range of values, but generally are lower than the surrounding soil and higher than the values characteristic of what would be expected if the only source were galactic cosmic rays (GCRS). The ^{26}Al and ^{53}Mn are well correlated. From bulk soil measurements, interpreted earlier by Langevin *et al.* (1982) and Nishiizumi *et al.* (1985), it is concluded that the large solar cosmic ray (SCR) excesses result from downslope motion of individual grains produced by very small surface impacts.

(*e*) *Lunar and martian meteorites*

Great excitement heralded the announcement when Brian Mason of the Smithsonian Institution in Washington, U.S.A., recognized that a meteorite recovered from Antarctica (ALHA81005) was identical to breccias brought back from the Moon. Subsequently, several other 'lunar meteorites' have been identified. Another class of objects referred to as SNCS (an acronym for the three meteorites that define this class of objects, namely Shergotty, Nakhla and Cassigny), is thought by many to have originated on Mars.

The great burden of proof for these origins has fallen to detailed chemical and isotopic analyses and to a comparison with what is known about the compositions of the Moon and

Mars. In the case of the Moon, the breccias are distinctive, and the Apollo samples provide material with which to make comparisons. In the case of the 'martian meteorites,' adequate information about Mars is lacking to make the identification compelling.

Cosmogenic radioisotope data are useful in examining the dynamics of the processes whereby these objects were removed from the surface and transported to Earth. To this end, we made measurements of ^{26}Al and ^{10}Be in three shergottites, ALHA77005, EETA79001 and Shergotty. When these data are combined with measurements of ^{36}Cl, ^{53}Mn, ^{21}Ne, and track data, it is possible to construct approximate exposure histories for these meteorites that have important implications regarding their origin (Nishiizumi *et al.* 1986*b*). Particularly surprising is the evidence for SCR production of ^{26}Al and ^{53}Mn in ALHA77005. This is the first time that SCR production has ever been detected in a meteorite, because the outer few millimetres are almost always lost during ablation as the meteorite enters the atmosphere. This meteorite, with a radius of 5–6 cm, would appear to have lost less than 1 cm during ablation. The exposure age for ALHA77005 is similar to that of Shergotty (2.5 Ma and 2.2 Ma, respectively), but the meteorite EETA79001 has an exposure age of only 0.6 Ma. It seems highly unlikely that all three meteorites could have resulted from the same event, in light of these different exposure histories. None of the meteorites show evidence of any earlier exposure (based on the ^{21}Ne data; because ^{21}Ne is stable, it integrates the total exposure history), so that they must have been shielded by at least 3 m of material before beginning their exposure in space as bodies of approximately their present size. This implies that all three meteorites originally came from a much larger body. Whether that parent body was Mars or an asteroid, however, remains an open question.

We also measured the ^{26}Al and ^{10}Be in four lunar meteorites, and, again, there is evidence for more than one event responsible for the ejection and transport of these meteorites to Earth (Nishiizumi *et al.* 1986*c*). ALHA81005 appears to have come from a depth of about 70–80 cm beneath the lunar surface, where it had been exposed to cosmic rays for at least 15 Ma. Its journey to Earth was short (less than 0.1 Ma) and its residence time on Earth equal to 0.17 ± 0.05 Ma. Yamato 791197 came from closer to the Moon's surface, and it is still unclear whether it may have been the result of the same event as ALHA81005, as has been proposed. Finally, the two meteorites Yamato 82192 and Yamato 82193 appear from the radionuclide data to have arisen from the same event. Unlike the other two meteorites, they show extensive evidence of irradiation in space (from the ^{10}Be data), implying a long journey to Earth.

(*f*) *Cosmic spherules*

One of the most spectacular demonstrations of the sensitivity of AMS has been the measurement of ^{26}Al and ^{10}Be in individual cosmic spherules recovered from ocean sediment. These magnetic, usually silicate, spherules were discovered nearly a hundred years ago and suspected even then of being extraterrestrial. The question currently of interest is whether they are formed during the ablation of meteorites as they enter the atmosphere or whether they were small bodies in space, presumably of cometary origin. If the latter, they represent a unique source for a type of extraterrestrial material found nowhere else on Earth.

Once again use is made of the fact that ^{26}Al is abundantly produced by relatively low-energy protons emitted during solar flares, whereas ^{10}Be is not. As a result, the ratio of ^{26}Al : ^{10}Be in the outermost 1–2 mm of an extraterrestrial body can be nearly ten times higher than it is at depths of a centimetre or more, where, depending on the Al concentration, it is about 2. We have measured five particles, four of diameter approximately 450 μm (130–150 μg) and one of diameter 620 μm (316 μg) (Raisbeck *et al.* 1984). The ^{26}Al concentrations range from 25 to 61 × 10^{9} atoms g^{-1}, the ^{10}Be from less than 1 to 10 × 10^{9} atoms g^{-1}, and the ^{26}Al : ^{10}Be ratios from 5.7 to greater than 25. The spherules have higher ^{26}Al concentrations, lower ^{10}Be concentrations and considerably higher ^{26}Al : ^{10}Be ratios than saturated chondritic meteorites.

The high ^{26}Al concentrations and, in particular, the high ^{26}Al : ^{10}Be ratios suggest that the spherules are either ablation products of the outer 1–2 mm of meteorites or had pre-atmospheric sizes of less than 1 cm in diameter. But all five spherules had ^{10}Be concentrations substantially lower than is typical for chondritic meteorites, suggesting irradiation times in space of less than 1 Ma. This evidence supports the inference of a small pre-atmospheric size, but it is far from conclusive. ^{10}Be might have been lost preferrentially with respect to ^{26}Al during ablation, or by the action of seawater as the spherules lay on the bottom of the ocean. These alternative hypotheses can be tested. Ablation loss can be tested by comparing the ^{10}Be concentrations in the fusion crusts of meteorites, with its concentration just inside. As Raisbeck *et al.* (1987) have suggested, the loss of ^{10}Be by the chemical action of seawater can be tested by measuring the ^{10}Be in the spherules recently discovered in seasonal freshwater ponds in the ice of Greenland. But until these alternative explanations for the low ^{10}Be concentrations in cosmic spherules have been eliminated, the question of their origin will remain far from settled.

4. Terrestrial samples

(*a*) *Atmospheric production*

^{26}Al is produced in the atmosphere almost exclusively by the spallation of Ar by cosmic rays. Like ^{10}Be, it rapidly becomes attached to aerosols and is subsequently transported to the Earth's surface by rain. At the Earth's surface, it quickly becomes attached to soil particles and it is the fates of these particles that determine the subsequent history of the ^{26}Al.

We have determined the production rate of ^{26}Al by measuring its concentration in stratospheric-air-filter samples collected as part of the U.S. Department of Energy's project Airstream (Raisbeck *et al.* 1983). To avoid the question of collection efficiency, we measured the ^{10}Be concentration in the same samples to scale the production of ^{26}Al to that of ^{10}Be. The ^{26}Al : ^{10}Be production ratio we obtain is 3.8 × 10^{-3}. We have also measured the ^{26}Al concentration and ^{26}Al : ^{10}Be ratios in rain samples collected by one of our collaborators, K. Nishiizumi. Although rain is often contaminated with sufficient ^{27}Al from the Earth's surface to make these measurements impossible, occasionally rain is found with an ^{27}Al concentration low enough so that the ^{26}Al : ^{27}Al ratio is about 5 × 10^{-14}. From these measurements we obtain an ^{26}Al : ^{10}Be ratio of about 3 × 10^{-3}. The concentration of ^{26}Al in rain is about 70 atoms g^{-1}, and between 20 and 40 kg of rain were used in each measurement.

We have also obtained ice samples from Antarctica. These samples are ideal for measuring ^{26}Al, as the ^{27}Al concentration is typically 10–30 p.p.b.† (by mass). One sample of ice was collected from the Japanese station near Yamato mountain. The second sample was collected during an American expedition to the Theil mountains. The concentrations measured in these samples were 31.6 × 10^{3} ± 7 % and 184 × 10^{3} ± 7 % atoms g^{-1} for ^{10}Be, and 67.8 ± 22 % and 413 ± 10 % atoms g^{-1} for ^{26}Al, respectively. Two observations are in order. First, the concentrations of ^{10}Be and ^{26}Al are nearly six times higher in one sample than in the other.

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† Parts per billion; in this paper, 1 billion is used to represent 10^{9}.

Little is known about the depositional history of these samples, but the higher concentrations in the latter are suggestive of its having been deposited during Pleistocene time. Several workers have observed larger concentrations of ^{10}Be in Pleistocene ice compared with modern (Holocene) ice (H. Oeschger, this symposium; Beer *et al.* 1983; Raisbeck *et al.* 1981). Although the reason for these higher values is not well understood, it is believed to be related to climatic factors such as rainfall. The second observation is that despite the differences in absolute concentrations, the ^{26}Al : ^{10}Be ratio is the same in both samples, $2.15 \pm 0.50 \times 10^{-3}$ and $2.24 \pm 0.27 \times 10^{-3}$, yielding an average of $2.20 \pm 0.36 \times 10^{-3}$.

(i) *Geochemical comments*

A widespread assertion in the literature, no doubt occasioned by the frustration of trying to make a geological clock with atmospherically produced ^{10}Be, is that the geochemistry of Al and Be are similar enough that the ^{26}Al : ^{10}Be ratio can be used as a chronometer that would automatically compensate for variations in production rates and vagaries of geochemical pathway. Although our evidence is meagre, it already conclusively dismantles this idea.

The problem is the following. In analogy with ^{14}C, it has long been hoped that a clock operating on a time scale of about 3 Ma could be made by using ^{10}Be. But there are two significant ways in which the analogy between ^{10}Be and ^{14}C breaks down, despite the similarities in their atmospheric production. ^{10}Be, unlike ^{14}C, is not well mixed with its stable isotope at the time of its production, and this means that in many systems, the ^{10}Be : ^{9}Be ratio is of little significance. Second, carbon is a major constituent of the systems that ^{14}C is used to date, and the pathways by which the carbon (and hence the ^{14}C, because they are well mixed) is incorporated are well understood. Beryllium is a trace element (a few parts per million by mass in just about everything), and its incorporation normally is either the result of sorption or diffusion and therefore subject to influence by a myriad of external factors including pH and flow rate. This makes the interpretation of ^{10}Be per gram of material other than straightforward. Consequently, one is left in a quandary: neither the ^{10}Be : ^{9}Be ratio nor the ^{10}Be concentration (atoms g^{-1}) are immediately interpretable, leading many (L. Brown, this symposium) to despair of ^{10}Be ever being useful as a clock.

The one hope has been that the ^{26}Al : ^{10}Be ratio might somehow overcome these problems, if only the fates of the ^{26}Al and ^{10}Be produced in the atmosphere were the same. To examine this possibility, we measured the ^{26}Al : ^{10}Be ratios in manganese nodules, marine sediments and marine phillipsites. The marine setting was chosen as the ^{26}Al : ^{10}Be is generally higher than it is on land. Table 1 shows the results. The significance of these results is in the variability of the ^{26}Al : ^{10}Be ratio. Note, for example, that in coral and manganese nodules, the ^{26}Al : ^{10}Be ratios are about ten times lower than the atmospheric-production value. In phillipsites, it is several times greater. The ^{26}Al : ^{27}Al is roughly constant in authigenic minerals (with the exception of one value for coral J56.7), even though both the ^{26}Al and the ^{27}Al vary by almost five orders of magnitude. Unfortunately, ^{9}Be concentrations were not measured, so it is not possible to see if the ^{10}Be : ^{9}Be ratio is also constant. If so, then the ^{26}Al : ^{10}Be ratio could be corrected for chemical effects by the ^{27}Al : ^{9}Be ratio. Such an approach would fail for the sediments, however. In the sediments, the ^{26}Al : ^{27}Al ratio is lower than in the authigenic minerals, implying a second (undissolved) source of ^{27}Al. In addition, the ^{26}Al : ^{27}Al ratio is variable, probably because the dissolved and undissolved sources are poorly mixed. There seems little hope of using the ^{26}Al : ^{10}Be ratio in sediments to any value. In fact, the use of the ^{26}Al : ^{10}Be

TABLE 1. ^{26}Al AND ^{10}Be IN MARINE SYSTEMS

sample	^{27}Al (%)	10^{-14} ^{26}Al : ^{27}Al	^{26}Al 10^4 atoms g^{-1}	^{10}Be 10^7 atoms g^{-1}	10^{-3} ^{26}Al : ^{10}Be
authigenic					
coral†					
J56.F	$1.7 \pm 0.3 \times 10^{-4}$	37	1.40 ± 0.50	1.04 ± 0.10	1.35 ± 0.49
C3	$1.5 \pm 0.3 \times 10^{-4}$	7.6	0.26 ± 0.14	0.97 ± 0.10	0.27 ± 0.14
Mn-nodules†					
Aries 15 D (0–3.8 mm)	0.255	6.3	360 ± 110	2350 ± 190	0.21 ± 0.06‡
Aries 12 D (0–4.7 mm)	0.371	6.7	560 ± 220	1850 ± 210	0.40 ± 0.17‡
phillipsites					
core AMPH-48 (5–7 cm)	5.96	6.1	8100 ± 1200	330 ± 33	31.8 ± 5.6§
core AMPH-48 (16–17 cm)	2.90	6.2	4000 ± 460	317 ± 32	25.8 ± 3.9§
sediments					
calcareous					
MBC-3 site C (0–1.5 cm)	0.192	2.4	1200 ± 27	225 ± 16	0.53 ± 0.13
MBC-3 site C (1.5–3.5 cm)	0.77	8.8	4030 ± 65	384 ± 27	1.06 ± 0.19
siliceous					
MBC-3 site S (0–1.0 cm)	3.20	2.6	2100 ± 320	583 ± 41	0.36 ± 0.07
MBC-3 site S (2.8–4.6 cm)	2.38	1.8	1070 ± 320	599 ± 42	0.18 ± 0.06

† From Bourles *et al.* (1984).
‡ Extrapolated to surface.
§ Extrapolated to surface from accumulation rate of 0.21 mm ka^{-1} determined from ^{10}Be data.

ratio seems to be confined to systems in which the ^{26}Al and ^{10}Be are well mixed with their stable isotopes so that their stable isotopes can be used to correct for chemical effects.

(ii) *Tektites*

Tektites are silica-rich glassy objects found in several limited areas around the world. They range in size from about 100 μm for microtektites found widely distributed in selected sedimentary layers over about 30% of the ocean floor, to 10 cm objects of the Muong-Nong type found in Indochina. By far, the majority of tektites found on land are a few centimetres in size, and show evidence of at least two stages of melting. Their origin is obscure. Chemically they are similar to terrestrial sediments and unlike volcanic glasses. They are extremely depleted in volatiles, especially water, and homogeneous (lacking any zoning or microcrystalline structures), characteristics that are indicative of a long thermal history at elevated temperature. But their distribution and aerodynamic sculpturing would imply creation by impact, hardly allowing time for the diffusion necessary to account for these physical characteristics. Theories of extraterrestrial origin fail to explain their limited geographical distribution, their differences in elemental composition from any known lunar or meteoritic material, or the occurrence of sealed vesicles containing gases of a composition similar to the terrestrial atmosphere. (For a more thorough discussion see O'Keefe (1963, 1976); Barnes & Barnes (1973).)

Brown (this symposium) summarizes measurements of ^{10}Be concentrations of fifty tektites from the Australasian strewn-field (age *ca.* 0.7 Ma). Although there are several reasons to believe that this ^{10}Be is atmospheric in origin and inherited from the sediments from which the tektites were arguably formed, it is impossible from the ^{10}Be measurements alone to rule out the possibility that the ^{10}Be is extraterrestrial in origin. It could, for instance, have been transferred from the surface of an extraterrestrial body as it impacted the surface of the Earth. Or if tektites were formed in space, the ^{10}Be could have been produced on the surface of some extraterrestrial body, such as the Moon, before (or even during) the tektites' journey to the Earth.

The key to ruling out these other possibilities is the ^{26}Al : ^{10}Be ratio. The ^{26}Al : ^{10}Be ratio that results from atmospheric production is about 1000 times smaller than that resulting from production in solid materials and hence in space. But a straightforward comparison is not sufficient. If production is suspended for some reason, the ^{26}Al : ^{10}Be ratio changes, decreasing by a factor of two for every 1.31 Ma. Because the concentrations of ^{26}Al and ^{10}Be in extraterrestrial objects are much greater than they are in terrestrial materials, a long period of decay can reduce both the ^{10}Be concentration and the ^{26}Al : ^{10}Be ratio to near their terrestrial values. An additional complication results from the production of ^{26}Al and ^{10}Be in solid objects as they lie on the surface of the Earth. This phenomenon will be discussed at length in the next section. It is sufficient to point out here that, for the tektites, this production of ^{26}Al and ^{10}Be *in situ* barely affects the concentration of ^{10}Be (the ^{10}Be concentration is already more than 10 times greater than the saturation value produced by *in situ* production), but greatly increases that of ^{26}Al, thereby increasing the ^{26}Al : ^{10}Be ratio. These considerations increase the range of the ^{26}Al : ^{10}Be ratios measured in terrestrial and extraterrestrial materials, and reduce the difference between them.

We measured the ^{26}Al concentrations in five tektites, but in all but one were only able to set upper limits on their ^{26}Al concentrations. At the 95%-confidence level, they give a limit for the ^{26}Al : ^{10}Be ratio of less than 0.11. In one tektite, Macumba, we measured an ^{26}Al concentration of $8.8 \pm 2.2 \times 10^6$ atoms g^{-1} and because its ^{10}Be concentration is $121 \pm 8 \times 10^6$ atoms g^{-1}, the resultant ^{26}Al : ^{10}Be ratio is $7.3 \pm 1.9 \times 10^{-2}$. These results are plotted in figure 6.

Figure 6 demonstrates why we feel that this is conclusive evidence for a terrestrial origin of tektites. If the ^{10}Be measured in tektites were produced in space, the ^{26}Al : ^{10}Be ratio, initially, would have to have been between about 2.7 and 5.4; the exact value determined by the duration of exposure. In principle, there are four ways in which the extraterrestrial ^{10}Be concentration could be made to match the value measured in tektites, but all can be eliminated because of the measured ^{26}Al concentration. First, it could result from a short exposure (*ca.* 26 ka), but then the ^{26}Al : ^{10}Be ratio would be equal to 5.4. Second, the exposure could have occurred at a depth (*ca.* 260 cm) on some extraterrestrial body where the saturation value was equal to the value measured in tektites. This would give an ^{26}Al : ^{10}Be ratio of about 2.7. Third, surficial extraterrestrial material containing ^{10}Be might be mixed with other material (either extraterrestrial or terrestrial) lacking ^{10}Be to make a mixture equal to that observed in tektites; but then the ^{26}Al : ^{10}Be ratio would be unchanged and still be in the range 2.7–5.4. Finally, the ^{10}Be might result from an exposure in space, followed by an episode on or beneath the Earth's surface. This scenario could actually produce the observed ^{10}Be concentration and ^{26}Al : ^{10}Be ratio. However, as shown in figure 6, it would require a short exposure on the Moon (*ca.* 1.2 Ma) followed by a long period on Earth (*ca.* 7.8 Ma), at a depth great enough (greater than 1 m) that the *in situ* production of ^{26}Al would be insignificant. The age of 0.7 Ma of tektites measured by K–Ar rules out this scenario, which requires at least 9 Ma.

Therefore, excluding all extraterrestrial sources, it remains to be demonstrated that the measured ^{26}Al : ^{10}Be ratios are consistent with an origin on the Earth. As already mentioned, the ^{10}Be concentration in tektites is unaffected by any *in situ* production that occurs after their formation. Consequently, their measured ^{10}Be is 72% (because of decay) of their ^{10}Be

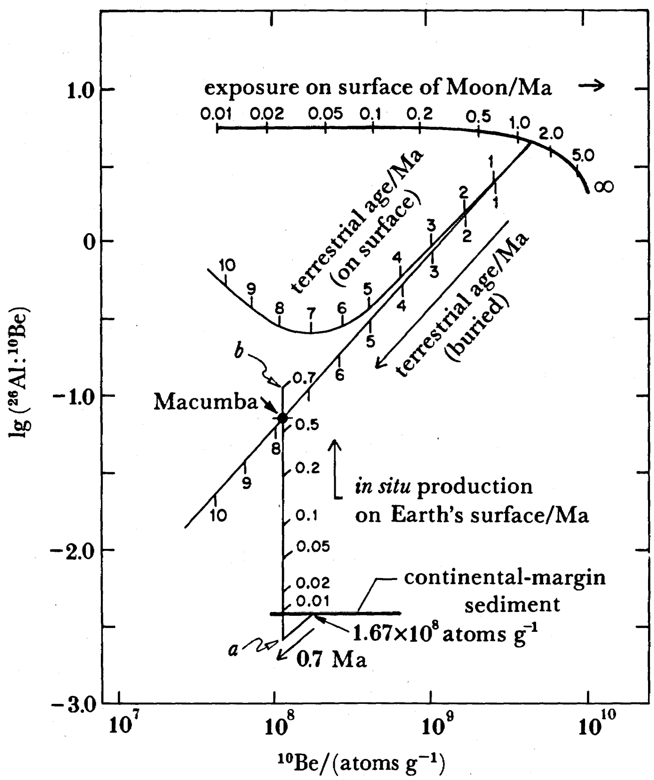

FIGURE 6. The result is shown for the australite, Macumba. Notice that its ^{10}Be concentration is consistent with a continental-margin sediment origin, and its ^{26}Al : ^{10}Be ratio with *in situ* production at or near the Earth's surface for *ca.* 0.7 Ma. *In situ* production rates are from Klein *et al.* (1986), with latitude correction factors from Yokoyama *et al.* (1977). The line representing continental-margin sediments shows the range of measured ^{10}Be values, and an assumed ^{26}Al : ^{10}Be ratio equal to that resulting from atmospheric production. The short-line segment from the continental-margin line to *a* shows the reduction in the ^{10}Be concentration and the ^{26}Al : ^{10}Be ratio that results from decay during 0.7 Ma, assuming no *in situ* production. The vertical line from *a* to *b* shows the increase in the ^{26}Al : ^{10}Be ratio that occurs with essentially no change in the ^{10}Be concentration during *in situ* production. The time scale of this line can either be interpreted as the amount of time the tektite has lain at the surface (the remainder of the time it was buried at a depth of greater than 2 m), or an estimate of the average burial depth; for Macumba this is *ca.* 20 cm. For a further explanation of the figure, see text.

concentration 0.7 Ma ago. On the other hand, their ^{26}Al concentration (and hence their ^{26}Al : ^{10}Be ratio) is almost solely the result of *in situ* production. The vertical line in figure 6, labelled *in situ* production on Earth,.shows that Macumba has lain at the surface of the Earth for about 0.5 Ma out of the past 0.7 Ma, or, on average, has been within the top 23 cm of the surface. The bonus from this study of ^{26}Al in tektites is that it corroborates their age (0.7 Ma) age as well as manifestly demonstrating their terrestrial origin.

(*b*) *In situ production*

Despite the 1 kg cm^{-2} of shielding provided by the atmosphere, ^{26}Al and ^{10}Be are produced in rocks at the Earth's surface by secondary cosmic rays. The principal reactions producing ^{26}Al are neutron-induced reactions with Al and Si (Jha & Lal 1981). Muon capture by ^{28}Si constitutes about 10% of the total ^{26}Al production at the Earth's surface, but becomes increasingly important at depth because of the more-penetrating nature of muons compared with neutrons. ^{10}Be production results primarily from spallation of oxygen. In silica, there are no reactions involving muons that produce ^{10}Be. We estimate (Klein *et al.* 1986), although others disagree (Yokoyama *et al.* 1977; Lal & Arnold 1985), that the production rates of ^{26}Al and ^{10}Be in quartz at sea level and in the latitude band 50–90° are 70 atoms g^{-1} a^{-1} and 10 atoms g^{-1} a^{-1}, respectively. These production rates are dependent on latitude (geomagnetic shielding), elevation (thickness of overlying atmosphere), and depth beneath the surface (shielding from overlying material). The maximum effect of geomagnetic shielding reduces production at the magnetic Equator to about one quarter of its value at the Poles. Production increases with altitude: at 10^3 m it is about twice that at sea level and at 5×10^3 m it is about 30 times greater. The rate of production, due to neutrons, decreases by a factor of two for approximately every 40 cm of depth beneath the Earth's surface (half-attenuation length *ca.* 120 g cm^{-2}).

Because the atmospheric production of ^{26}Al is low (*ca.* 4×10^3 atoms cm^{-2} a^{-1}), and the terrestrial abundance of ^{27}Al is high, the ^{26}Al : ^{27}Al ratio on land is rarely greater than 10^{-15} and in marine sediments it rarely exceeds 10^{-14}. However, *in situ* production in materials having low ^{27}Al concentrations can result in very significantly higher ^{26}Al : ^{27}Al ratios. For example, in quartz with an Al concentration of 10 p.p.m. (by mass), the ^{26}Al : ^{27}Al ratio reaches 10^{-14} in 32 years! At saturation, the rate of production equals the rate of decay; the ratio is 3.2×10^{-10} (7.1×10^7 atoms g^{-1}). In contrast with the atmospheric ^{26}Al : ^{10}Be production ratio of 3.8×10^{-4}, at the inception of *in situ* production this ratio is 7 and at saturation it is equal to 3.3 (^{26}Al, because of its shorter half-life, obtains saturation before ^{10}Be). These ratios should also be compared with the chondritic value of 1.8 and the lunar value of 2.7.

(i) *Libyan Desert glass*

Our first application of *in situ* production was an attempt to unravel the exposure–burial history of Libyan Desert glass (LDG) in an effort to understand its transport and distribution during the past 3 Ma. LDG is a tektite-like material of unknown origin, high in silica (*ca.* 98%), found in the Western Desert of Egypt and estimated to be about 28 Ma old (fission track and K–Ar). (For more information see Giegengack & Issawi 1975; Weeks *et al.* 1984.)

Twelve samples were measured. Six were selected from a group collected from a small geographic region under the supposition that they might have experienced similar recent exposure histories. Five samples were chosen to be representative of as large a geographic area as possible, and one large piece was sampled twice. A thorough discussion and table of the results are given in Klein *et al.* (1986).

For the purposes of summary, the results are plotted in figure 7. This figure is a useful aid in understanding the effects of complicated exposure–burial scenarios. The upper line in the figure shows the ^{26}Al : ^{10}Be ratio as a function of ^{10}Be concentration during exposure at the surface for a sample with initial concentrations of ^{26}Al and ^{10}Be equal to zero. It is the maximum possible ^{26}Al : ^{10}Be ratio for a particular ^{10}Be concentration given any combination of exposures and burials. It is calculated from the production rates of ^{26}Al and ^{10}Be; for the purpose of constructing this figure, the estimates of Klein *et al.* (1986) were used. The bottom curve shows the decrease in the ^{26}Al : ^{10}Be ratio during decay as a function of the decrease in ^{10}Be concentration for a sample previously exposed long enough to be at saturation. This decay would result from the sample being shielded from cosmic rays, such as occurs during burial. It is the minimum ^{26}Al : ^{10}Be ratio for a given ^{10}Be concentration. All combinations of exposure–burial–exposure, etc., result in ^{26}Al : ^{10}Be ratios and ^{10}Be concentrations that fall between these two curves (assuming that the production rates used to construct the upper curve are consistent with those experienced by the sample).

It is immediately apparent from this figure that the various pieces of LDG have had very

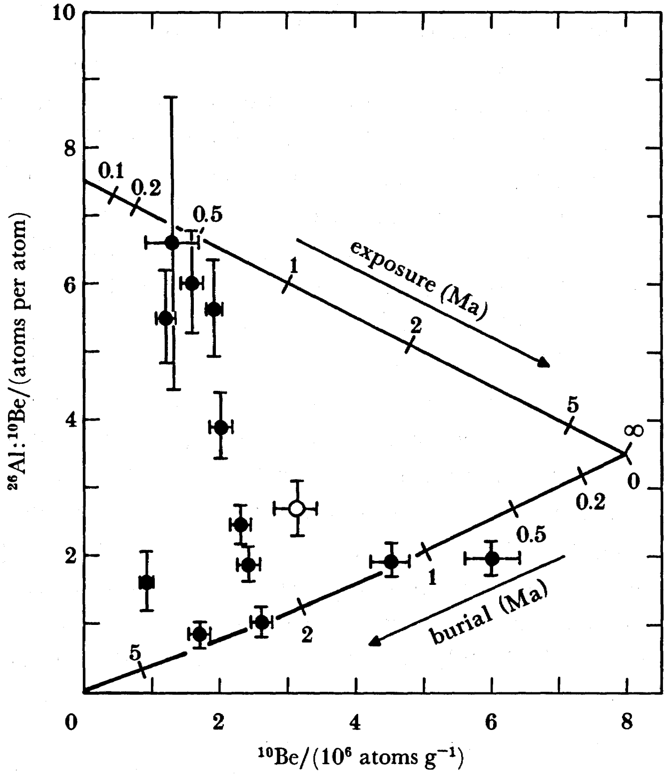

FIGURE 7. Measurements of Libyan Desert glass plotted against the 'exposure–burial' triangle based on the production rates of Klein *et al.* (1986). All possible exposure and burial sequences result in ^{26}Al : ^{10}Be ratios and ^{10}Be concentrations that plot within the 'triangle.' The great spread of values are indicative of the varied histories experienced by individual pieces of glass. The open circles are values from Yiou *et al.* (1984).

different exposure histories. There appears to be no correlation with location, leading to the conclusion that the LDG underwent significant redistribution within the past 100 ka or so. All ^{26}Al : ^{10}Be ratios are greater than one, proving that the ^{26}Al and ^{10}Be were produced *in situ*, and do not result from the scavenging of atmospherically produced isotopes. The 28 Ma age for the glass rules out the possibility that the ^{26}Al and ^{10}Be were either inherited from the precursor material from which the glass was formed, or from an impacting body (meteorite) that may have been responsible for its formation.

From these modelling considerations, it is possible to calculate cumulative exposure times, summed over the past 10 Ma, for each sample. It is also possible to calculate a lower limit for the amount of time each sample has remained buried, subsequent to its initial exposure. These results are tabulated in Klein *et al.* (1986); suffice it to say that there is no obvious systematic pattern in the exposure and burial times so calculated. Presumably the fate of a small glassy object adrift in a sea of sand is subject to many idiosyncrasies.

(ii) *Quartz and erosion*

An application of the cosmic-ray production of radionuclides in rocks that promises to be of extraordinary usefulness in answering important geologic questions is the production of ^{26}Al and ^{10}Be in quartz to determine rates of continental weathering and erosion. Quartz is an attractive mineral for this purpose because of its low Al concentration, the slow rate with which it undergoes alteration and hence its limited ability to pick up atmospherically produced ^{10}Be, and its occurrence in a great number of geologic settings. Although the idea of using cosmic-ray production of radionuclides in rocks is not a new one (Davis & Schaeffer 1955), once again, it has only been with the advent of AMS that measurements at the required level of sensitivity have been possible.

Radionuclides are produced in rocks within the top metre or so of the surface. The concentration continues to increase with time until the rate of production is equalled by the rate of decay. This process is about 90% complete within three half-lives. The concentration can be expressed as

$$C = [P(l,a)/\lambda]\,(1-\mathrm{e}^{-\lambda t})\,\mathrm{e}^{-\rho d/\Lambda},$$

where C is the concentration of the radionuclide; P, its production rate, a function of latitude (l) and altitude (a); λ, its decay constant; Λ, its mass attenuation length; d, its depth below the surface; and ρ, the density of the overlying material.

If erosion occurs, then the 'depth' or shielding of the sample is a function of time. If this erosion occurs at a constant rate, the formula is the same but with λ replaced by $\lambda' = \lambda(1+\rho\epsilon/\Lambda\lambda)$, where ϵ is the rate of erosion. Hence the concentration of a single isotope can, in principle, be used to determine the rate of erosion. If a sample is in secular equilibrium, then for a 'surface' sample, $C = P/\lambda'$ or

$$\epsilon = \frac{\Lambda\lambda}{\rho}\left(\frac{P}{\lambda c}-1\right) = \frac{\Lambda\lambda}{\rho}\left(\frac{C_0}{C_\mathrm{m}}-1\right),$$

where C_0 is the equilibrium concentration with no erosion, and C_m is the measured value. For the ^{26}Al–^{10}Be system, this gives a practical measurement range

$$2\times10^{-5} < \epsilon < 10^{-2} \quad (\mathrm{cm\ a^{-1}}).$$

It can be shown (Klein & Lal 1987) that the measured rate of erosion is within a factor of two of the average rate, if instead of occurring by a continuous process, erosion occurs stochastically with pieces of 30 cm or less spalling off at random intervals. The averaging period depends on the rate ($T = C_\mathrm{m}/P$). For very fast erosion, the averaging period is short (if $\epsilon = 10^{-2}$ then $T \approx 5.9$ ka).

If the process of erosion is even less constant than this, and periods of relatively high erosion are punctuated by periods of low erosion (interglacial–glacial, for example), then a second isotope is essential in understanding what is occurring, and the same arguments that were used to explain the LDG can be employed. The result is that not only can the rate of erosion be determined, but the duration of the periods during which erosion was absent can as well.

Figure 8 summarizes the results of measurements on sixteen samples, showing both maximum erosion rates and minimum burial times. A detailed discussion of these data are impossible here, but the interested reader is referred to Nishiizumi *et al.* (1986*a*) and Klein & Middleton (1987).

We believe that this method could be used to determine the extent and history of ice coverage during glacial times. This is because the ice itself, and the rock it removes during its retreat (in temperate zones), are effective shields preventing cosmic-ray-induced radionuclide production. By the methods already outlined, this period of 'burial' can be determined from measured concentrations of ^{26}Al and ^{10}Be.

A major problem, recognized from the outset, but not fully appreciated for its intractability, is the need for a closed system in which radionuclides, especially ^{10}Be, produced in the

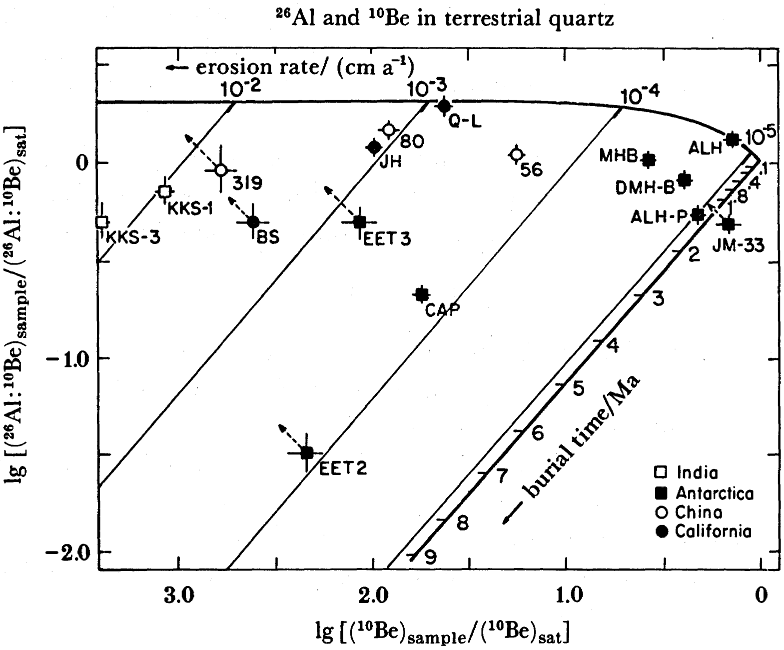

FIGURE 8. Summary of the measurements of ^{26}Al and ^{10}Be made to date in terrestrial quartz. The measured ^{10}Be concentrations and the ^{26}Al : ^{10}Be ratios have been normalized to their saturation values, based on rates of production calculated from Klein *et al.* (1986) and on elevation and latitude factors from Yokoyama *et al.* (1977). This normalization enables data from all over the world and from all altitudes to be plotted on a single set of coordinates. For five samples, the ^{10}Be represents an upper limit, either because of boron-induced backgrounds, or because it is suspected that not all of the atmospherically produced ^{10}Be was removed before measurement (JM-33). The broken-line vectors show how their positions on the plot would change if their ^{10}Be concentrations were reduced. Erosion rates are maximal values. Note that most samples show evidence for variable erosion rates (i.e. periods of suspended erosion or 'burial'). Another possible explanation for their lower-than-expected ^{26}Al : ^{10}Be ratios is contamination with atmospherically produced ^{10}Be.

atmosphere are completely excluded from the measurement. This can be extremely difficult in situations where the sample of interest is found in a matrix with a ^{10}Be concentration four orders of magnitude larger. (The highest concentration we have ever measured in a terrestrial sample was seen in a granular rock matrix from Antarctica, where we measured a ^{10}Be concentration of 10^{10} atoms g^{-1}.) Several of the samples in figure 8 are suspected of being contaminated with atmospherically produced ^{10}Be. Thus the plotted values represent upper limits for their ^{10}Be concentrations and lower limits for their ^{26}Al : ^{10}Be ratios. Other samples may be contaminated as well. The best indication we have of contamination is a low ^{26}Al : ^{10}Be ratio. We are currently developing improved chemical and physical separation procedures to limit this contamination.

Finally, we have measured one sample in which we are fairly confident that the ^{26}Al results exclusively from muon capture by ^{28}Si. This sample was recovered from a depth of 10 m at a road cut. The expected ^{26}Al concentration from neutron production at this depth is *ca.* 3 atoms g^{-1}. The measured value is 8×10^4 atoms g^{-1}, which is consistent with about a 10% muon contribution at the Earth's surface and an attenuation length of muon production of *ca.* 1.5×10^3 g cm^{-2}. Unfortunately, we were only able to obtain an upper limit for the ^{10}Be concentration (less than 5×10^4 atoms g^{-1}) that is many orders of magnitude greater than the concentration we would have expected (*ca.* 1 atom g^{-1}).

PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

(*c*) ^{10}Be *and* ^{26}Al *in uranium and thorium ores*

Recently, ^{14}C has been observed in the exotic α-like decay of several radium isotopes. Whereas theoretical estimates all but rule out the emission of ^{10}Be or ^{26}Al by a similar mechanism, either isotope might be produced at detectable levels by highly asymmetric fission or by ternary fission. Consequently, we measured the ^{10}Be and ^{26}Al concentrations in a number of uranium and thorium ores and in samples of ^{235}U and ^{238}U that had been irradiated in a reactor.

The ^{10}Be and ^{26}Al concentrations were measured in four uranium ores ranging in concentration from 1% to 58% and in a 14% thorium ore. In all cases, the observed concentrations were surprisingly high, with ^{10}Be ranging from 3×10^6 to 2.5×10^7 atoms g^{-1} and ^{26}Al from 1×10^7 to 1.4×10^{10} atoms g^{-1}. The large ^{10}Be and ^{26}Al to fission ratios in the uranium ores (10^{-4}–6×10^{-4} and 3×10^{-3}–10^{-1}, respectively) and the very presence of ^{10}Be and ^{26}Al in the thorium, which does not decay by spontaneous fission, make it highly unlikely that fission is the major source of the ^{10}Be and ^{26}Al. It is noteworthy that not only were the ore samples chosen to have minimum atmospheric contamination, but the large ^{26}Al : ^{10}Be ratios all but eliminate atmospherically produced ^{10}Be and ^{26}Al as the source.

The most likely sources of the ^{10}Be and ^{26}Al are the nuclear reactions ^{7}Li(α, p)^{10}Be ($Q = -2.56$ MeV, $E_{\text{th}} = 4.03$ MeV) and ^{23}Na(α, n)^{26}Al ($Q = -2.96$ MeV, $E_{\text{th}} = 3.48$ MeV). Table 2 lists the measured concentration of ^{10}Be and ^{26}Al in the various ores along with their lithium and sodium concentrations. Also listed are the calculated ^{10}Be and ^{26}Al concentrations assuming: (1) only 4 α-particles from the decay chains contribute to production; (2) the energy-averaged cross section is 50 mb,† (3) the effective target thickness is 2.5 mg cm^{-2}; and (4) the lithium and sodium are uniformly distributed throughout the ore. The last column lists the measured-to-calculated ratios. That these differ from unity is not surprising because assumptions (1)–(3) are subject to errors of at least ±50%. What is surprising is that the measured-to-calculated ^{10}Be and ^{26}Al ratios vary by almost a factor of ten. This strongly suggests that the lithium and sodium are not uniformly distributed throughout the ores.

Although it is evident from the foregoing that most of the ^{10}Be and ^{26}Al in the uranium ores is a result of α-particle-induced reactions, it is possible that some came from fission. To test this, we measured the ^{10}Be and ^{26}Al concentration in two samples of enriched (97.66%) ^{235}U and one sample of enriched (99.98%) ^{238}U that had been irradiated in a reactor. Because this work, which was performed in collaboration with R. Fleming and R. M. Lindstrom from the

TABLE 2. ^{26}Al AND ^{10}Be IN URANIUM AND THORIUM ORES

concentration (%)	Li (p.p.m.)[a]	^{10}Be atoms g^{-1}	^{10}Be$^{b}_{\text{calc}}$ atoms g^{-1}	^{10}Be : ^{10}Be$_{\text{calc}}$	Na (p.p.m.)[a]	^{26}Al atoms g^{-1}	^{26}Al$^{b}_{\text{calc}}$ atoms g^{-1}	^{26}Al : ^{26}Al$_{\text{calc}}$
1.0 U	13.6	3.1×10^6	4.9×10^6	0.63	199	2.3×10^7	1.0×10^7	2.3
1.64 U	11.6	2.78×10^6	6.7×10^6	0.41	104	1.1×10^7	8.7×10^6	1.26
7.69 U	12	6.98×10^6	33.0×10^6	0.21	250	1.02×10^2	9.8×10^7	10.4
57.6 U	16.9	24.9×10^6	35.0×10^7	0.07	1360	1.43×10^{10}	4.0×10^9	3.6
14.2 Th	36	1.45×10^7	5.9×10^7	0.24	95	9.1×10^7	2.2×10^7	4.1

[a] By mass.

[b] The same assumptions were made for ^{10}Be and ^{26}Al (see text).

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† 1 mb = 10^{-3} b = 10^{-31} m^2.

National Bureau of Standards and L. Brown and F. Tera from the Carnegie Institution of Washington will be reported elsewhere, only a summary of the results is presented here.

Table 3 summarizes the results of the ^{10}Be and ^{26}Al measurements in the two ^{235}U and the ^{238}U sample. ^{10}Be was very evident in the two ^{235}U samples and, as can be seen from the table, the measured numbers of ^{10}Be atoms per fission are in good agreement. Some fissions were observed in the ^{238}U sample (this received the same irradiation as the more highly irradiated ^{235}U sample), and are largely a result of the presence of 0.0175 % ^{235}U. This accounts for about 25 % of the small number of ^{10}Be atoms observed in the ^{238}U sample, the balance presumably being the result of nuclear reactions with light-element contaminants. If it is assumed that the light-element contamination of the ^{235}U and ^{238}U samples are similar (which is known to be the case) then less than 0.1 % of the observed ^{10}Be atoms from the ^{235}U are from this source. The present value of 7.8×10^{-6} atoms of ^{10}Be per fission is in reasonable agreement with the value 4.1×10^{-6} reported by Vorob'ev *et al.* (1969) particularly because the latter authors may have failed to observe ^{10}Be ions emitted with energies less than 13 MeV or greater than 31 MeV.

TABLE 3. ^{26}Al AND ^{10}Be IN IRRADIATED URANIUM ISOTOPES

sample	no. of fissions	^{10}Be†	^{26}Al†	^{10}Be per fission	^{26}Al per fission
^{235}U	7.69×10^{12}	5.88×10^{7}	—	7.6×10^{-6}	—
^{235}U	2.81×10^{14}	1.43×10^{9}	$< 3.5 \times 10^{5}$	7.9×10^{-6}	$< 1.9 \times 10^{-9}$
^{238}U	2.0×10^{10}	6.8×10^{5}	$< 1.6 \times 10^{6}$	3.4×10^{-5}	—

† Total atoms in sample.

The search for ^{26}Al in the ^{235}U and ^{238}U samples was unsuccessful and the zero counts obtained set an upper limit of 1.9×10^{-9} at the 95 %-confidence level on the ^{26}Al per fission ratio from ^{235}U. It is noteworthy that, because the mass of the ^{235}U sample was only 2.7 mg and the irradiation time was 2 h, the sensitivity of this experiment could readily be improved by at least 3 orders of magnitude. Indeed, AMS provides a new and very sensitive method of detecting very rare fission modes and a worthwhile experiment might be to search for neutron-rich ^{32}Si in fission.

5. FUTURE DIRECTIONS

The foregoing has been a celebration of the new opportunities afforded by the great increase in sensitivity possible with AMS. We have also tried to stress the usefulness of using two isotopes to solve problems that are insoluble by using only one.

We anticipate that AMS measurements of ^{10}Be and ^{26}Al produced *in situ* in terrestrial samples will soon be a valuable tool for understanding surficial geological processes. Before this is possible, a fair amount of ground work is necessary. Production rates of ^{26}Al and ^{10}Be will have to be determined in materials of geological significance, and reliable procedures will have to be developed to separate quartz (and perhaps other materials such as olivine) from a surrounding matrix containing atmospherically produced ^{10}Be. In all likelihood, measurements of ^{26}Al produced in the atmosphere will become easier as techniques for measuring ^{26}Al improve, especially as larger currents of Al$^-$ become available. This will make possible the use of ^{26}Al : ^{10}Be ratios in a number of other systems as well. Perhaps an understanding of the climatic factors affecting radionuclide distribution will be assisted by such measurements. Another possibility might be the dating of ice by using ^{26}Al : ^{10}Be ratios.

The measurement of ^{26}Al in extraterrestrial samples will, no doubt, continue to be of great interest. The use of SCR-produced ^{26}Al will find increasing application in unravelling histories of meteorites and cosmic spherules. ^{26}Al measured in mineral separates will help, when coupled with ^{10}Be to elucidate the irradiation histories of meteorites. Similarly, the histories of other special classes of meteorites, such as eucrites, and 'lunar' and 'martian' meteorites, may well be better understood from ^{26}Al and ^{10}Be measurements.

One of the great positive aspects of accelerator mass spectrometry is the way in which it fosters collaborations among scientists in many varied fields. The work we have done bears testimony to the success of these collaborations. Without the help of experts, we would not have been able to identify interesting projects, nor had the expertise to carry them out. We especially thank Grant Raisbeck and Francoise Yiou for bringing to our attention cosmic spherules, and with whom we made the first measurements of LDG. Kuni Nishiizumi and Jim Arnold are responsible for much of the work that we have done on meteorites and the Moon. Greg Herzog and Dave Alymer have suggested, and provided us with samples from, several interesting extraterrestrial problems. Robert Giegengack has been an important resource of information on all matters geological. Pankaj Sharma separated the Be and Al from the Libyan desert glass, and initiated the research on atmospheric ^{26}Al in marine samples. Louis Brown, Fouad Tera, and the rest of the group at Carnegie Institution of Washington, are of immeasurable and constant assistance on almost all problems we undertake.

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Discussion

W. KUTSCHERA (*Argonne National Laboratory, Illinois, U.S.A.*). Concerning the production of ^{26}Al by muon interaction in quartz, I assume that it is mainly the stopped muon on ^{28}Si that contributes to the ^{26}Al. The question is whether this might be used as a tool to trace the stopped-muon flux as a function of depth going down in rock.

J. KLEIN. We have made one measurement on a sample of quartz recovered from a depth of 10 m, in which we are confident that the ^{26}Al was produced by muons. From this single measurement, and the assumption that the muons contribute about 10% to the production at the surface, we estimate the muon attenuation length to be about 10 metre-water-equivalents. We would like to measure the ^{26}Al in several samples acquired at various depths to determine more accurately the muon attenuation length and its contribution to surface production. In October 1987 we will irradiate SiO_2 in the stopped-muon channel at LAMPF to measure the ^{26}Al production cross section.

M. PAUL (*Hebrew University, Jerusalem, Israel*). In the measurements of ^{10}Be and ^{26}Al in uranium and thorium ores, Professor Middleton showed the ratios of measured against calculated ^{26}Al and ^{10}Be. These numbers were not one, but they were consistently higher than one for the ^{26}Al and lower than one for ^{10}Be. What is the significance of this?

R. Middleton. The fact that the measured-to-calculated ratios for ^{26}Al were in general higher than one, and for ^{10}Be lower than one, is completely arbitrary. The cross sections we assumed were a bit outrageous, but the point is that neither the ratios for Al nor Be are consistent. Both show variations on the order of a factor of ten. Whatever assumptions one makes, it is impossible to get them all right. Therefore the only conceivable interpretation is that the lithium (in the case of ^{10}Be) and the sodium (in the case of ^{26}Al) are not uniformly distributed with respect to the uranium. The presence of ^{10}Be and ^{26}Al in the thorium ores (thorium does not undergo spontaneous fission) clinches the argument against the ^{10}Be and ^{26}Al resulting from highly asymmetric fission.

H. Margaritz (*Weitzmann Institute, Rehovot, Israel*). Did Professor Middleton try to separate the mineral phases so that he could establish a correlation between the uranium, lithium and ^{10}Be concentrations or the uranium, sodium, ^{26}Al concentrations? I presume that the sodium occurs in definite phases, whereas the lithium may not, making it easier to look for the ^{26}Al correlation than for the ^{10}Be.

R. Middleton. I think this was considered but thought to be very difficult. The point is that the range of the α-particle is only of the order of 5 μm, so one has to look within 5 μm of a concentration of uranium and this is very difficult, especially as we need on the order of 10^6 atoms to make a measurement.