Abstract—We present data for the cosmogenic nuclides $^{10}$Be and $^{26}$Al in a suite of 24 extraterrestrial spherules, collected from Antarctic moraines and deep sea sediments. All of the 10 large spherules collected in glacial till at Lewis Cliff are extraterrestrial. As in earlier work, the great majority of particles show prominent solar cosmic-ray (SCR) production of $^{26}$Al, indicating bombardment ages on the order of 10$^6$ years or even longer. These long ages are in direct contradiction to model ages for small particles in the inner Solar System and may require reconsideration of models of small particle lifetimes. A small fraction of the particles so far measured (6/42) possess cosmogenic radionuclide patterns consistent with predictions for meteoroid spall droplets. We believe that most of the spherules were bombarded in space primarily as bodies not much larger than their present size.

The content of in situ produced $^{10}$Be and $^{26}$Al in quartz pebbles in the same moraine suggests that these spherules may have on average a significant terrestrial age.

INTRODUCTION

Cosmic spherules and fragments in the size range 0.1–1 mm, originally extracted magnetically from deep-sea sediments (Brownlee et al., 1979; Millard and Finkelman, 1970; Murray and Renard, 1883; Murrell et al., 1980) have now been isolated from ice in Greenland and Antarctica (Harvey and Maurette, 1990; Koeberl and Hagen, 1989; Maurette et al., 1986).

Concentrations of cosmogenic radionuclides have been measured in a representative suite of deep-sea spherules and Greenland particles by accelerator mass spectrometry (AMS) (Nishizumi et al., 1991; Raisbeck and Yiou, 1987; Raisbeck et al., 1985b). Also, light noble gases were measured in similar particles by an ultrasonic noble gas mass spectrometer (Olinger et al., 1990). The data show that most of the particles had lifetimes in space of >10$^6$ years. These lifetimes based on measurements are much longer than collision lifetimes expected of similar sized particles from model calculations, which predict a few times 10$^4$ years or even longer. These long ages are in direct contradiction to model ages for small particles in the inner Solar System and may require reconsideration of models of small particle lifetimes. A small fraction of the particles so far measured (6/42) possess cosmogenic radionuclide patterns consistent with predictions for meteoroid spall droplets. We believe that most of the spherules were bombarded in space primarily as bodies not much larger than their present size.

Several questions seem important to us at this stage: Are these new spherules found in the glacial till of extraterrestrial origin? What is the relationship among the deep-sea sphcrules, Greenland particles, and those from the Antarctic? What is the distribution of micrometeoroid lifetimes in space? Since melt droplets from larger meteoroids must occur, how can they be identified and what is their proportion relative to spherules produced by atmospheric melting of initially small particles? We measured the cosmogenic radionuclides $^{10}$Be ($T_{1/2} = 1.5 \times 10^6$ years) and $^{26}$Al ($T_{1/2} = 7.05 \times 10^5$ years) in 16 individual spherules collected from glacial till in Antarctica along with 8 additional deep-sea spherules to investigate these questions. Results for 6 of the Antarctic particles were previously reported (Nishizumi et al., 1992).

SAMPLE DESCRIPTION AND EXPERIMENTAL METHODS

The concentrations of cosmogenic $^{10}$Be and $^{26}$Al and major element chemical compositions were measured in 10 individual large spherules (150–420 $\mu$m) from Antarctica. These spherules were collected from Pacific Ocean sediments (18°41’N, 123°47’W, 4280 m depth). After weights and diameters were measured, all particles were individually mounted in acrylic resin, and a small surface was polished flat with Al oxide followed by C coating. The quantitative elemental analysis of these polished surfaces was performed using a JEOL 733 Superprobe. Magnetism, Al, Si, Ca, Cr, Mn, Fe, and Ni were measured at 15 kV with a spot size of 30 $\mu$m. Results are shown in Table 1. The probe analysis errors are ±1% for Mg, Si, and Fe, ~2% for Al and Ca (because of their lower abundance), <10% for Cr and Mn, and relative errors for Ni range from 10% to 20%. After electron microprobe analysis, each particle was separated from the resin with chloroform and washed with ethanol and deionized water in an ultrasonic bath. The individual weights were measured again. We dissolved with HF-HNO$_3$ mixture along with Be (0.5 mg) and Al (1.0 mg) carriers. Beryllium and Al were chemically separated and purified for AMS. These procedures are similar to our earlier work (Nishizumi et al., 1991). The $^{10}$Be and $^{26}$Al concentrations were determined using the Lawrence Livermore National Laboratory tandem accelerator (Davis et al., 1990). The measured $^{10}$Be/Be ratios ranged from 1 x 10$^{-14}$ to 2 x 10$^{-13}$ and the measured $^{26}$Al/Al ratios from 1 x 10$^{-14}$ to 8 x 10$^{-13}$. After correcting for the $^{10}$Be background due to $^{11}$Be and for chemical blank (6 x 10$^{-15}$ for $^{10}$Be/Be and 1 x 10$^{-14}$ for $^{26}$Al/Al), the measured ratios were normalized to ICN $^{10}$Be and NBS $^{26}$Al standards, which were diluted at La Jolla. The corrected ratios were converted to atom concentration (atom/g sample) and activities (dpm/kg sample). The results are shown in Table 2 along with density of the samples.

RESULTS AND DISCUSSION

Table 1 gives the diameter, mass, and major-element composition (wt%) of Antarctic spherules, LC-7-LC-16, and deep-sea spherules, LJ-36-LJ-2737, along with the results of previous...
that nearly half of the deep-sea spherules are oxidized Fe particles. Murrell et al. (1980) and our unpublished work indicate that the stony particlcs in deep-sea spherules, LC-1-LC-6, for comparison (Nishiizumi et al., 1992). Oxygen concentration is estimated by difference and shown in the table. Spherule LC-1 is extremely unusual; it is a black, glassy, pear-shape rather than rounded or elliptical. Even allowing for internal holes in LC-4 and LC-6, the mean density of these particles, average = 2.35 ± 0.56 g/cm³ (Murrell et al., 1980), consistent with their lower mean Fe content. Samples LJ-2722, 2730, and 2733 are Fe particles, which appear to be oxidized to a composition in the range of FeO-Fe₂O₃. Concentrations of Fe and O for both Antarctic spherules and deep-sea spherules are anticorrelated, as shown in Fig. 1 (R = 0.988). The plot includes all Lewis Cliff particles and both stony and Fe deep-sea spherules except LJ-36 and 2736. Nickel concentrations in all Lewis Cliff particles except LC-10 are depleted compared to deep-sea spherules.

A Mg-Si-Fe ternary diagram is useful to show how the Lewis Cliff compositions compare with the compositions of 410 magnetically collected deep-sea spherules (KK1) (Brownlee et al., 1983). As shown in Fig. 2, an abnormal fraction of the Mg-Si-Fe compositions are Si-rich or Fe-poor relative to the main cluster of KK1 compositions. This may be partly explained by the fact that the KK1 deep-sea spherules were collected magnetically, so there is a bias favoring Fe-rich particles in the KK1 collection. It is also possible that the Lewis Cliff moraine spherules are biased towards lower density materials that are more easily carried by wind.

Although compositions of Lewis Cliff particles are slightly different from deep-sea spherules, all 16 Lewis Cliff particles are stony type. Murrell et al. (1980) and our unpublished work indicate that nearly half of the deep-sea spherules are oxidized Fe particles.

In addition to the difference of collection methods, magnetic (deep-sea spherules) vs. hand picking (Lewis Cliff), it seems that the aridity and lack of liquid water of the Antarctic environment are more favorable for survival of stony particles than conditions on the ocean floor. This aridity also results in long terrestrial ages of Antarctic meteorites and low erosion rates of Antarctic surface rocks. Assuming the Lewis Cliff particles and deep-sea spherules had similar origins, the higher abundance of stony types among the Lewis Cliff particles indicates that the stony particles in deep-sea spherules may be destroyed comparatively quickly by weathering or
there is a strong collection bias against non-magnetic properties. It is also possible that the moraine spherules are highly selected by the aeolian process that concentrated them. We may have underestimated the flux of stony particles based on deep-sea spherule results (Mauger et al., 1987).

So far as the stony particles are concerned, the cosmogenic radionuclide data are consistent with all samples being members of the same population. Unquestionably, they are all extraterrestrial, validating the collection and selection methods employed.

The production rates of $^{10}$Be and $^{26}$Al in cosmic spherules for various shielding conditions and sizes have been discussed (Nishiizumi et al., 1991) using improved Reedy-Arnold model calculations (Reedy, 1987, 1990; Reedy and Arnold, 1972). Recently, Reedy and Masarik (1995) updated their model calculations. The results are summarized below. We used the average chemical composition of stony type deep-sea spherules for these calculations: 17% Mg, 1.4% Al, 16.5% Si, 1.4% Ca, 0.2% Mn, 22.0% Fe, 0.5% Ni, and 40.7% O. The average chemical compositions of 16 Lewis Cliff particles are similar to these values, with slightly higher concentrations of Al and Si and lower concentration of Fe compared to deep-sea spherules. Because of the variety of chemical compositions for individual particles and the possible effects of terrestrial weathering, we used the same average chemical composition for cosmogenic nuclide calculation of Lewis Cliff particles.

The major target element for $^{10}$Be production is O. The O concentrations of Lewis Cliff particles are 42.4 ± 2.4%, and the $^{10}$Be production rate calculation for stony particles would not be sensibly affected if individual compositions were used. For $^{26}$Al production, the major targets are Si and Al. Although the production rate of solar cosmic-ray-produced $^{26}$Al is sensitive to the Mg and Al concentrations, the average chemical composition was adopted. The uncertainty of depth and size of individual particles has a much larger influence than composition differences for the solar cosmic-ray calculation.

The saturation activity of $^{10}$Be in small stony type objects (less than a few mm in diameter) is estimated to be ~8 dpm/kg (Reedy, 1987). The saturation value of $^{10}$Be on the surface of a large body (24 kyr irradiation), such as an asteroid, is somewhat higher, 10–12 dpm/kg. In ordinary-sized chondrites (radius ≤ 40 cm), the
The observed $^{10}\text{Be}$ activities of Lewis Cliff particles are ~20 dpm/kg or less. These Lewis Cliff spherules show clear evidence of exposure to galactic cosmic-ray bombardment on time scales from ~10^6 up to as much as 10^7 years. Many Lewis Cliff spherules contain high $^{26}\text{Al}$, up to ~240 dpm/kg, which is produced by solar cosmic-ray bombardment of small objects in space or very near the surface (<1 g/cm^2) of larger objects. Cosmicogenic nuclide contents in Lewis Cliff particles are very similar to those in deep-sea spherules and, although the data set is small, also for Greenland particles. To date, the $^{10}\text{Be}$ and $^{26}\text{Al}$ concentrations in 42 individual particles (16 Lewis Cliff particles from Antarctica, 2 Greenland particles and 24 deep-sea spherules), which have some chemical analysis in addition to cosmicogenic nuclide concentration, are known (Nishiizumi et al., 1991, 1992; Raisbeck et al., 1985b). These data can now be used to study their lifetimes in space and the conditions under which they were bombarded by galactic cosmic-ray and solar cosmic-ray particles. Figure 3 illustrates $^{26}\text{Al}$ vs. $^{10}\text{Be}$ in 41 stony particles. One stony deep-sea spherule (KK2-5) contains high $^{10}\text{Be}$ (50 ± 7 dpm/kg) (Raisbeck et al., 1985a) and is not shown in the figure. Beryllium-10 concentrations in five deep-sea spherules are shown only as upper limits. The solid line and dashed lines indicate $^{26}\text{Al}$ and $^{10}\text{Be}$ in typical sized meteorites and their ranges.

In previous studies, the high $^{26}\text{Al}$ values were thought to reflect cosmic-ray irradiation of small objects (4x bombardment) or of the surface of asteroids that are meters or more in diameter (2x bombardment) (Nishiizumi et al., 1991; Raisbeck et al., 1985b). However, such a model cannot explain the high $^{10}\text{Be}$ (~15 dpm/kg) found in 8 out of 42 particles, including those described in previous work (Nishiizumi et al., 1991; Raisbeck et al., 1985b). These levels of $^{10}\text{Be}$, which are similar to ordinary chondritic levels, suggest that this population of particles may have received galactic cosmic-ray bombardment in objects of size often encountered in meteorites (a few cm to 40 cm). The combination of solar cosmic-ray produced-$^{26}\text{Al}$ and high $^{10}\text{Be}$ can be produced at the surface of such bodies. Two possible sources are (1) surface erosion products from meteoroids in their orbits, or (2) melt droplets from larger meteoroids during atmospheric entry. The latter is an old idea for the origin of deep-sea spherules. Even though a large fraction of the preatmospheric mass of meteoroids is lost to ablation during passage through the atmosphere (Bhandari et al., 1980), for meteorites near the smaller end of the size range, a significant fraction of this ablated material would have been exposed to cosmic rays at <1 cm depth. The very high $^{10}\text{Be}$ in one particle, KK2-5, measured by Raisbeck et al. (1985a) cannot be explained by any of these models.

For this and an earlier group of Lewis Cliff and deep-sea spherules, in six cases (LC-6, LC-10, LC-13, LC-15, LJ-527, LJ-2373) the contents of $^{10}\text{Be}$ and $^{26}\text{Al}$ are consistent with their being spall droplets from a depth >3 cm in meteoroids. Thus far the $^{10}\text{Be}$ and $^{26}\text{Al}$ data would allow 6 out of the 42 stony objects measured to be explained with an ablation hypothesis. In all other cases, clear evidence is present for solar cosmic-ray bombardment (i.e., $^{26}\text{Al}/^{10}\text{Be}$ greater than the ratio produced in meteorites by galactic cosmic-ray). Although some $^{26}\text{Al}$ might be produced by solar cosmic-rays at the ablation surface of meteorites, the distribution of $^{10}\text{Be}$ exposure ages in these particles is shifted toward shorter ages than those of ordinary chondrites.

An earlier paper (Nishiizumi et al., 1991) discussed two acceptable models (4x and 2x) for the bombardment history of the particles bearing a clear solar cosmic-ray signature. In our view, the present data and other arguments favor the 4x model, but these data are not yet conclusive (see below). Measurements of other nuclides, such as $^{53}\text{Mn}$ and $^{59}\text{Ni}$, can provide further useful discriminants.

Quartz-containing terrestrial pebbles were also collected from the same Lewis Cliff glacial moraine. The quartz separated from the pebbles contains in-situ produced $^{10}\text{Be}$, (2.74 ± 0.09) × 10^6 atom/g, and $^{26}\text{Al}$, (1.18 ± 1.1) × 10^6 atom/g, respectively (Nishiizumi et al., 1992). The $^{26}\text{Al}/^{10}\text{Be}$ ratio, 4.34 ± 0.42, is slightly lower than for a pure surface exposure and may correspond to a burial of the

![FIG. 3. The correlation between $^{10}\text{Be}$ activity (dpm/kg) and $^{26}\text{Al}$ activity (dpm/kg) for Antarctic, Greenland, and deep-sea spherules. The solid line and dashed lines indicate $^{10}\text{Be}$ and $^{26}\text{Al}$ concentrations in typical sized chondrites. Exposure age increases along the line to the right.](image-url)
moraine beneath ice or soil for ~0.5 Ma. If Lewis Cliff particles had similar terrestrial histories, 26Al and 10Be concentrations in these particles would be about 60% and 20–30% higher, respectively, when these particles fell to Earth than they are at present. However, there is insufficient basis now for meaningful corrections.

MODELS

There are a number of qualitative arguments which favor bombardment of these objects mainly or entirely as small bodies (radius < 1 cm or even < 1 mm; Raisbeck et al., 1985). One is simply the relative number or mass flux of such bodies in interplanetary space, relative to that of meteoroids or larger bodies (Love and Brownlee, 1991, McDonnell, 1978). Another is the overwhelming predominance of particles showing high solar cosmic-ray production. Still, the two major model calculations of lifetime of small grains (Dohnanyi, 1978; Grün et al., 1985) so far made are in flat contradiction with our data since they suggest a mean lifetime of 2 x 10^3 years. A full exploration of the alternatives requires, in our view, a new model taking into account such constraints as the erosion rates of lunar rocks. However, this is beyond the scope of the present paper.

CONCLUSIONS

We confirm that nearly all spherules show unambiguous solar cosmic-ray effects. They have been bombarded in space for periods on order of 10^5–10^7 years, at depths of generally <1 mm. The range of exposure ages of particles is in good agreement with previous work. 26Al and 10Be lifetimes range from 2 x 10^4 years. A full exploration of the alternatives requires, in our view, a new model taking into account such constraints as the erosion rates of lunar rocks. However, this is beyond the scope of the present paper.

REFERENCES


