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G.M. Raisbeck* and F. Yiou*, J. Klein** and
R. Middleton**, D. Brownlee***

* Laboratoire Rene Bernas, C.S.N.S.M.,
BP N°1, 91406 Orsay, FRANCE

** University of Pennsylvania, Philadelphia, PA 19104,U.S.A.

*** University of Washington, Seattle, Wa 98195, U.S.A.
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ABSTRACT. Using accelerator mass spectrometry the cosmogenic isotopes 26 Al (half-life 716,000 years) and 10 Be (1.5 x 106 years) have been measured in deep sea magnetic spherules. Because 26 Al can be abundantly produced by relatively low energy solar flare particles, while 10 Be is mainly produced by higher energy galactic cosmic rays, the ratio 26 Al/ 10 Be of a body irradiated in space increases as the size of the body decreases. Our measured ratios of 26 Al/ 10 Be in the spherules are much larger than found in meteorites, and indicate irradiation in interplanetary space of a parent body of $\lesssim 1$ cm diameter. Since most bodies of this size entering the atmosphere are associated with cometary orbits, our results strongly suggest that these spherules represent cometary debris. Our results also suggest an irradiation time in space of the order of 10^6 years for the parent bodies.

1. INTRODUCTION

The extraterrestrial origin of deep sea magnetic spherules has been suspected ever since their discovery, almost a hundred years ago (Murray and Renard, 1891). Modern studies have strongly supported this point of view (Brownlee, 1981), but the exact nature of the parent material has remained uncertain. There is considerable evidence to suggest that the spherules are ablation products of larger bodies which have impacted with the atmosphere (Brownlee, 1981). A minority view is that the spherules exist already in interplanetary space as melted objects from asteroidal collisions (Parkin et al., 1983).

If the spherules are ablation products, the size of the initial objects is of critical importance. Most meteoroids entering the earth's atmosphere weigh < 1 mg, have cometary type orbits, and presumably represent cometary debris. On the other hand, it is generally believed that at least a significant fraction of the larger size objects giving rise to meteorites come from asteroids (see references in Brownlee, 1981).

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On the basis of 53 Mm data, Nishiizumi (1983) and Nishiizumi and Arnold (1982) have argued that spherule parant bodies are meteorite size objects. The development of the technique of accelerator mass spectrometry now makes possible the measurement of other cosmogenic isotopes in these spherules. We earlier reported measurements of 10 Be (half-life 1.5 million years) and 26 Al (716,000 years) in three groups of chondritic type spherules (Raisbeck et al., 1983). We noted that the 26 Al/ 10 Be ratios suggested \lesssim 1 cm size objects as the parent bodies for at least some of these spherules. We also mentioned that similar measurements in individual spherules were highly desirable to test this hypothesis, as well as to give additional information on parent size and irradiation times. We report here our first results in a program to measure 10 Be and 26 Al in individual spherules.

2. EXPERIMENTAL PROCEDURE AND RESULTS

The chondritic spherules considered here were magnetically scavenged from the ocean floor using the "cosmic muck rake", at the same (KK2) or nearby (KK1) location as those reported as sample B-1 in Raisbeck et al. (1983). Their sizes and weights are given in Table I. After weighing, the spherules were dissolved, and an aliquot taken for ²⁷Al measurements by flameless atomic absorption spectrometry. Carrier solutions of 9 Be (0.3 or 0.5 mg) and 2 Al (0.5 mg) were then added, and the two elements separated on a Dowex 50 x 8 ion exchange column. After further purification, the samples were transformed into BeO and Al₂0, for measurement on the accelerator. The procedures for ¹⁰Be and ²⁶Al measurements on the University of Pennsylvania tandem accelerator are described in Klein et al. (1982) and Middleton et al. (1983), respectively. Because of the order of magnitude less material used, the uncertainties in the measurements are larger than in our earlier work. The uncertainty on the 26 Al measurements is mainly due to the statistics of the number of atoms detected. The $^{10}\mathrm{Be}$ measurements, on the other hand, were limited by interference from a 10B induced background. Because of this interference we were able to obtain only upper limits to 10Be concentrations for two of the spherules.

The results are given in Table I. For convenience we also give the earlier results from Raisbeck et al. (1983) for the group of spherules (B-1) recovered by the same technique, and at the same place, as the spherules studied here.

TABLE I $^{26}\mathrm{Al}$ and $^{10}\mathrm{Be}$ in deep sea magnetic spherules

Sample	Diameter (microns)	Weigh (µg)	t ²⁷ Al (%)	²⁶ Al (10 ⁹ atoms/g)	10 Be (10 ⁹ atoms/g)	²⁶ Al/ ¹⁰ Be	Irradiation age (My)
B-1a		1780	1.12 ±.11		5.6 ± 0.8	8.2 ± 1.7	
KK1-1	465	140	$0.83 \pm .08$	61 ± 9	10.7 ± 3.6		
KK1-2	445	133	$0.61 \pm .06$		2.7 ± 1.6	21 ± 13	0.4
KK1-3	445	118	$0.47 \pm .05$	25 ± 5	< 1.5	> 17	< 0.2
KK1-4	485	152	$0.88 \pm .09$	31 ± 7	5.0 ± 2.0	6.1 ± 3.4	0.8
KK2-3	620	316	$1.08 \pm .11$	33 ± 6	< 1.0	> 25	< 0.2
Chondritic Meteorites (saturated)			0.87	20-35 ^b	20 - 22 ^C	1 - 1.5	
Chondritic (c2) cosmic dust (saturated)				189 ^d	₁₄ d	~ 14	

a From Raisbeck et al. (1983)

3. DISCUSSION

The results for the individual spherules in Table I tend to support our earlier interpretation. We include in Table I the saturation values found for 10 Be and 26 Al in chondritic meteorites, and calculated values for very fine chondritic dust irradiated to saturation in interplanetary space at 1 AU. For irradiation times short compared to the ²⁶Al half-life, the ²⁶Al/¹⁰Be ratio would be about twice these values. In Fig. 1 we show a curve for the $^{26}\,\mathrm{Al}/^{10}\,\mathrm{Be}$ ratio as a function of depth from the surface of a body of chondritic composition irradiated in space. This curve has been estimated in the following way. The concentrations at zero depth have been taken from the calculations of Reedy (1984) for chondritic dust. It can be noted that the corrected value of Michel et al. (1982) gives a similar result for 26 Al. The values at large depths are taken from observed meteorite results. The shape of the curve between these points has been estimated from calculated ²⁶ Al depth profiles in lunar soils and rocks (Reedy et al., 1984, and private communication) and chondritic meteorites (Michel et al., 1982) assuming 10 Be remains relatively constant. It is realized that there are significant uncertainties in such a procedure. Among other things, the calculations are quite

b From Heymann and Anders (1967);

c From Moniot et al. (1982) and Sarafin et al. (1984)

d From Reedy (1984)

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sensitive to the assumed solar flare spectrum, chemistry and size of the parent body, and erosion effects. However, such calculations have given reasonable fits to observed ²⁶Al profiles, particularly in lunar samples (Reedy et al., 1983) and we believe Fig.1 should be sufficiently accurate for the purpose of the discussion that follows.

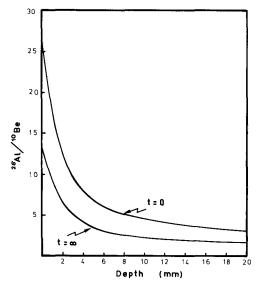


Figure 1. Calculated $^{26}\text{Al}/^{10}\,\text{Be}$ as a function of depth (assuming a density of 3 g/cm³) for chondritic matter irradiated in interplanetary space at 1 AU for a time very short (t = 0) or very long (t = $^{\infty}$) compared to ^{26}Al half-life (716,000 years). See text for details.

Although the uncertainties are large, it can be seen from Table I that the $^{26}{\rm Al/}^{10}{\rm Be}$ ratios of all the spherules are considerably larger than found in meteorite size objects. In fact, in two of the spherules (KK1-1, KK1-2), the $^{26}{\rm Al}$ concentrations themselves are much larger than ever found in chondritic type meteorites. This supports our earlier interpretation that a significant fraction of the $^{26}{\rm Al}$ in the spherules has been formed by solar flare particles. From Fig.1 we can see that the $^{26}{\rm Al/}^{10}{\rm Be}$ ratios in the spherules are consistent with irradiation in parent bodies having sizes of $\lesssim 1$ cm. Since most bodies of this size entering the atmosphere are associated with cometary like orbits, it seems reasonable to conclude that the spherules represent cometary debris.

As can be seen in Table I, the concentration of $^{10}\mathrm{Be}$ in meteoritic and dust size chondritic matter is estimated to vary only modestly. It is for this reason that we felt justified in assuming an approximately constant $^{10}\mathrm{Be}$ concentration in calculating $^{26}\mathrm{Al}/^{10}\mathrm{Be}$ for Fig.1. Using this same assumption, and for irradiation times of the order of the $^{10}\mathrm{Be}$ half-life or shorter, it is possible to estimate the irradiation times $(T_{\underline{i}})$ from the observed $^{10}\mathrm{Be}$ concentrations using the expression:

$$T_i = -\frac{1}{\lambda} \ln (1 - R)$$

where λ = the ¹⁰Be decay constant (4.62 x 10⁻⁷ years) and R is the ratio of the ¹⁰Be concentration to the saturation value (which we take as being (17 ± 3) x 10⁹ atoms/g). The irradiation times calculated in this way are given in Table I. Although these estimates are probably only good to a factor of 2 or so, the implied exposure ages are considerably shorter than found for most meteorites (Reedy et al., 1983).

It must be emphasized that the above irradiation times assume no loss of 10 Be from the spherules, either during their entry into the atmosphere, or during the time they spent in contact with sea water. Similarly, the conclusions based on the 26 Al/ 10 Be ratios assume no preferential loss of either of the isotopes during the same stages. The 27 Al concentrations in the spherules, are in reasonably good agreement with concentrations in carbonaceous chondrite meteorites, suggesting that any loss of this element must be quite modest. Unfortunately a similar test for 9 Be is very difficult because of its very low abundance. It should, however, be possible to test for this possibility in the future by studying similar spherules recently recovered from polar ice (M. Maurette, private communication). These spherules are much less severely etched than those found in sea sediments, and the volatile element contents of some of them suggest a much milder heating in the atmosphere.

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Walker: To clarify the question of the distinction between ablation which would remove the outer skin with a high 26Al/10Be compared to the interior - and irradiation in space as small objects: what is the max. 26 Al/10 Be seen in surfaces of lunar rocks? In short, what is the dividing line in terms of ²⁶Al/¹⁰Be? Answer: Although there exist detailed ²⁶Al profiles, I am not aware of any experiments that have measured ²⁶Al/¹⁰Be in the very surface layers of lunar rocks. Even if they were done, however, a direct comparison with the spherules would not be possible because the lunar material has a higher concentration of ²⁷Al, which is an important progenitor for production of ²⁶Al from solar flares. However, our interpretation of the ²⁶ Al/¹⁰ Be results implying irradiation in small bodies is not based on the high value alone. The preablation surface of a chondritic meteorite should indeed have a high 26 Al/10 Be ratio. However most chondritic meteorites are observed to be saturated in 10 Be, and therefore presumably their surface layers would be also, contrary to what is observed in the spherules.

<u>Lamy</u>: Your conclusion for parent bodies \lesssim 1 cm may not necessarily imply a cometary origin. The sporadic background, which is only partly connected to comets, is far superior to the contribution by streams associated to comets.

<u>Answer</u>: Our suggestion that small parent objects ($\lesssim 1$ cm) imply a likely cometary origin is based simply on the fact that most bodies of this size entering the atmosphere (>95% according D.W. Hughes, private communication at this meeting) are estimated to have comet like orbits, even if only a much smaller fraction are associated with specific streams.