

**Radioactive 7Be Materials Flown on LDEF**

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Final Report

The following article is the draft of a presentation prepared for the 2nd LDEF Post-Retrieval Symposium, held June 1-5, 1992. This paper was presented by J.C. Gregory, and contains a report of the work done up to the date of the conference on the detection and quantification of cosmogenic isotopes on the LDEF.

COSMOGENIC RADIONUCLIDES ON LDEF:  
AN UNEXPECTED  $^{10}\text{Be}$  RESULT

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SUMMARY

Following the discovery of the atmospheric cosmogenic radionuclide  $^7\text{Be}$  on the LDEF<sup>1)</sup>, we began a search for other known nuclides produced by similar mechanisms. None of the others have the narrow gamma-ray line emission of  $^7\text{Be}$  decay which enabled its rapid detection and quantification. This paper describes a search for  $^{10}\text{Be}$  atoms on LDEF clamp plates using accelerator mass spectrometry. An unexpected result was obtained.

## INTRODUCTION

During the first few weeks after return of LDEF to the Kennedy Space Center, the presence of the radio-isotope  $^7\text{Be}$  was discovered during surveys of the spacecraft for cosmic-ray-induced radio-nuclides. It was clear from those results that there was more of the isotope on the leading side than on the trailing. From further experiments at the Marshall Space Flight Center the activity of  $^7\text{Be}$  was found to be  $6 (\pm 2) \times 10^5$  atoms per  $\text{cm}^2$  (corrected to date of the orbital retrieval in January, 1990) on a variety of leading edge metal surfaces including stainless steel and aluminum.<sup>1)</sup> The activity associated with similar substrates from the trailing edge was of the order of 100 times less. The hypothesis proposed in the Nature article recognized production of  $^7\text{Be}$  in the atmosphere by the action of cosmic ray protons and secondary neutrons with atmospheric oxygen and nitrogen nuclei, and the transport of the Be upwards to orbital altitudes. The oxygen density at these altitudes is too low by 3 or 4 orders of magnitude to account for production in situ. The Be atoms then stuck to the LDEF surfaces until they decayed ( $t_{1/2}$  for  $^7\text{Be}$  is 53 days).

We have subsequently described<sup>2)</sup> a mechanism by which Be species were transported upwards as positive ions (at least above 100km). By analogy with meteoritic metal ion chemistry,  $\text{Be}^+$  is the most probable form in that environment, rather than Be, BeO, or  $\text{BeO}^+$ . Simple diffusion of mass 7 particles (without invoking electrostatic effects) from the turbopause to 300km has been shown<sup>3)</sup> adequate to explain our results in a general way.

From the point of view of surface chemistry, the front surfaces of stabilized vehicles like LDEF are scoured clean of hydrocarbon contamination by the ambient atomic oxygen. Surfaces of aluminum and stainless steel would be covered with a thin layer of oxide;  $\text{Al}_2\text{O}_3$ ,  $\text{Cr}_2\text{O}_3$ ,  $\text{Fe}_2\text{O}_3$ , etc. This may be modified in some cases by the presence of  $\text{SiO}_2$ . In all cases these are ionic lattices into which an  $8 \text{ km s}^{-1}$   $\text{Be}^+$  ion could easily be absorbed. The sticking probability at present is unknown, but because its value on stainless steel and aluminum were found to be similar, we have assumed it to be 1.

Nuclear spallation reactions are rather indiscriminate, and all possible products are formed, though with different cross-sections and half-lives. The stable isotopes produced generally cannot be measured by quantitative chemical analysis either because the number of atoms is too few, or more usually, because at these levels all materials are contaminated with most stable nuclides. A few unstable isotopes are produced, however, including  $^{14}\text{C}$  and  $^{10}\text{Be}$  from atmospheric oxygen and nitrogen and  $^x\text{Cl}$  and  $^y\text{Al}$  from argon. Such nuclides may be detected and quantified by the technique of accelerator mass spectrometry (AMS) and we describe here the results of a search for  $^{10}\text{Be}$ .

$^{10}\text{Be}$  is of most interest in our present study because of its chemical similarity to  $^7\text{Be}$ . This allows us to assume similarity in atmospheric oxidation and ionization characteristics, and in the chemistry of the surface interaction at the satellite. Since the source function of the two isotopes are known, their chemistry the same, differences in steady state concentration at orbital altitudes and consequently in measured surface density on LDEF will be dependent on differences in mass and half-life. The measurement would be a useful check on models of atmospheric diffusion or transport at these altitudes.

### Experimental Approach

- a) The University of Pennsylvania Center for AMS
- b) Preparation of the Samples

AMS samples are normally in the form of a few milligrams of metal oxide (*or chloride?*) which is placed into the Cu cathode of the AMS ionization source. They are obtained by dissolving rock or mineral samples in suitable acids, extracting the Be, and precipitating the hydroxide. In this case it was necessary to remove a thin layer of the surface of the LDEF material sample by chemical means. The samples available to us were anodized aluminum tray clamps. Since about  $10^7 - 10^8$  atoms of  $^{10}\text{Be}$  are required per sample by the AMS technique, and since the cross-section for  $^{10}\text{Be}$  production from O or N is similar (somewhat less) than that for  $^7\text{Be}$ , we need ca.  $100\text{ cm}^2$  of surface area per sample. This is based on analogy with the measured density of  $^7\text{Be}$ . Since  $t_{1/2}$  for  $^{10}\text{Be}$  decay is 1.5 million years, corrections for decay since LDEF retrieval are unnecessary.

Each tray clamp was made of anodized aluminum alloy, and of dimensions 2 in. x 5 in. x 0.125 in., providing about  $60\text{ cm}^2$  of surface area on the exposed side. Clamps used were from LDEF locations F-9, A-10, B-2 and F-4 (9 and 10 being leading edge rows and the others close to the trailing edge). All surfaces of each clamp which were not to be etched were coated with Petropoxy 154 epoxy cement and cured for a few minutes at  $150^\circ\text{C}$ . The first plate (B2) was etched with a few mL of 3:1 v/v solution of 9M HCl and 20%  $\text{HNO}_3$  (high purity). Pre-existing scratches were attacked vigorously but most of the surface did not appear to react. We were concerned that a thin layer of  $\text{Si O}_2$  a few nm thick, formed in orbit, may have been protecting the Al surface from attack. This  $\text{Si O}_2$  layer may also contain some of the  $^{10}\text{Be}$  of interest. We abraded the surface with  $5\mu\text{m Al}_2\text{O}_3$  powder and later with stainless steel wool.  $^7\text{Be}$  carrier solution containing  $1.484\text{ mg Be mL}^{-1}$  was added. The other plates were treated with steel wool

and a mixture of acids including HF. Two successive etchings were performed on each of the last three plates and these samples, each of volume 40 to 80 mL, were then processed and analyzed separately. The steps in this processing were:

1. Evaporation of solution to dryness; redissolution in 9M HCl and filtering of solid residue.
2. Removal of Fe and Co using Dowex 1-X8 ion exchange column.
3. Removal of Al in a large ion-exchange column (Dowex 50W - X8). Elution of Be with 300 mL 1N HCl.
4. Precipitation of  $\text{Be}(\text{OH})_2$ , and some residual  $\text{Al}(\text{OH})_3$ , using ammonium chloride solution; washing of the precipitate.
5. Redissolution and passage through cation exchange column.
6. Redissolution and precipitation of  $\text{Be}(\text{OH})_2$  with  $\text{NH}_4\text{OH}$ .
7. Calcining of  $\text{Be}(\text{OH})_2$  in quartz crucibles at  $850^\circ\text{C}$ .
8. Loading of Be oxide into AMC copper cathodes.

## Results

The results of these assays are shown in Table 1. In terms of areal density it may be seen that the absolute numbers  $\text{cm}^{-2}$  for  $^{10}\text{Be}$  are of the order of  $10^6$ , a reasonable value in light of the  $^7\text{Be}$  number densities. However, there is clearly no ram/wake effect and a second etch of both ram (A-10) and wake (F4) plates produced similar values. These results are inconsistent with our hypothesis of atmospheric origin. The last column expresses the same data in terms of  $^{10}\text{Be}$  atoms per gram of Al. A mean value is about  $10^8$  atoms of  $^{10}\text{Be}$   $\text{g}^{-1}$  Al. Note that this is based on an assumed constant etch depth of  $100\mu\text{m}$ .

## Discussion

We have considered the following explanations for the result:

- 1) *Laboratory contamination with  $^{10}\text{Be}$ .* These assays were performed in laboratories familiar with the risk of ambient contamination (the isotope is normally present in surface dirt in concentration of  $\sim 10^8$  atoms  $\text{g}^{-1}$ ). Controls are used in various ways to catch the presence of such contamination. It was not considered a plausible cause for the result.

2) *Production in orbit by spallation of Al with energetic protons .*

$$\text{No. } ^{10}\text{Be atoms g}^{-1}\text{Al} = \int \Phi (E) t. N_A/A.\sigma (E) dE$$

$$\text{for } t(\text{flight}) \ll t_{1/2} (^{10}\text{Be})$$

where:  $\Phi (E) t$  is the mission proton fluence given by Watts<sup>4</sup>),  $N_A$  is Avogadro's no.,  $A=27$ , and  $\sigma (E)$  is the energy dependent cross-section for the reaction:  
 $p + ^{27}\text{Al} \rightarrow ^{10}\text{Be} + \dots$

This approximate calculation gave the LDEF mission production as:

$$n(\text{mission}) ^{10}\text{Be} \sim 1.3 \times 10^5 \text{ atoms/g Al}$$

The mechanism is clearly incapable of explaining our measurements.

3) *<sup>10</sup>Be is naturally present in industrial aluminum.* It is clear that all the results are explained if this is the case. This was not known to us, and is quite unexpected.

**TABLE 1**

<b>Clamp Plate No.</b>	<b>No. Be-10 atoms/cm<sup>2</sup></b>	<b>No. Be-10 atoms/g of plate</b>
F9-7 (1st etch)	$0.53 \times 10^6$	$0.64 \times 10^8$
A10-1 (1st etch)	$0.96 \times 10^6$	$1.15 \times 10^8$
(2nd etch)	$0.91 \times 10^6$	$1.1 \times 10^8$
B2-3 (1st etch)	$7.1 \times 10^6$	$8.5 \times 10^8$
F4-2 (1st etch)	$1.2 \times 10^6$	$1.44 \times 10^8$
(2nd etch)	$2.2 \times 10^6$	$2.64 \times 10^8$

**Table 1.** Measured <sup>10</sup>Be densities on LDEF clamp plates. Data are presented both as area density of <sup>10</sup>Be atoms cm<sup>-2</sup> of exposed side of clamp plate and as volume density of <sup>10</sup>Be atoms per gram of aluminum plate assuming a common etch depth of 100μm for all plates. Note that plate B2-3 was etched more deeply than the others.



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