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## LETTERS TO NATURE

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## Observation of <sup>7</sup>Be on the surface of LDEF spacecraft

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THE Long Duration Exposure Facility (LDEF), an orbiting unmanned satellite, was recently returned to Earth after almost six years in space. From radioactivity measurements, we have found significant quantities of the isotope <sup>7</sup>Be on the leading edge (but only on the leading edge) of LDEF. Although the absolute atmospheric concentration of <sup>7</sup>Be needed to explain this detection is extremely small (10<sup>-7</sup> atoms cm<sup>-3</sup>), its concentration at LDEF's altitude (310 km) must be several orders of magnitude higher than in the stratosphere below, where it is produced by cosmic-ray reactions with atmospheric nitrogen and oxygen nuclei. To explain the presence of <sup>7</sup>Be on the surface of LDEF, it must first be rapidly and efficiently transported to high altitudes, and then adsorbed onto the surface of the spacecraft. Neither process had been expected. Our detection may therefore lead to the use of <sup>7</sup>Be as an exo-atmospheric tracer, as well as to studies of surface interactions in space.

The LDEF spacecraft was launched by the space shuttle Challenger on  $7$  April 1984 into a nearly circular orbit with an inclination of 28.5° and an altitude of 480 km. It was retrieved by the space shuttle Columbia on 12 January 1990 at an altitude of 310 km. Because of its large mass, long space exposure and the wide variety of materials onboard, the LDEF provided a unique opportunity for induced radioactivity studies. These measurements are still in progress and will be reported elsewhere.

The LDEF spacecraft is a twelve-sided cylindrical aluminium structure, 9.1 m long by 4.3 m in diameter (see Fig. 1). It consists of an open grid to which were attached various experiment trays designed to measure the effects of long space exposure on spacecraft materials and components. Throughout its orbital lifetime, the spacecraft was passively stabilized about all three axes of rotation, allowing one end of the spacecraft to point

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always toward the Earth, and fixed leading and trailing sides with respect to the orbital motion.

After its return to the Kennedy Space Center, gamma-ray spectra were obtained along each side of the spacecraft using a germanium detector array provided by the Naval Research Laboratory. Measurements were also made of selected components from the spacecraft. The gamma-ray line at 478 keV from the radioactive decay of <sup>7</sup>Be was unambiguously observed to emanate from the leading side of the spacecraft, as shown in Fig. 1. The weaker signal observed from the trailing side of the spacecraft can be traced to the attenuated gamma-ray flux from the leading surfaces.

Individual components were brought to the Marshall Space Flight Center to quantify the residual radioactivity on the LDEF. A high-purity germanium detector inside a low-level background facility was used to obtain spectra of small aluminium and steel samples taken from the leading and trailing sides. In Figs 2 and 3, gamma-ray spectra of two identical aluminium plates and two steel trunnion end pieces taken from the leading and trailing sides of the spacecraft are shown. A clear <sup>7</sup>Be signal was seen on the leading side, with little or no signal above background on the trailing side.

A polished aluminium plate, used as a thermal control surface in LDEF experiment AO114 (ref. 1), was subjected to several tests to determine the depth of penetration and the form of deposition of the <sup>7</sup>Be. The surface was coated with collodion, stripped to remove all loose particles, then wiped firmly with xylene. Less than 10% of the surface activity was removed by this process, indicating that the  $7Be$  was neither associated with dust particles nor other soluble surface contaminants. An acid etch, without a stable beryllium carrier, removed several tens of micrometres of the aluminium surface, and most of the <sup>7</sup>Be activity (the remainder was assumed to be re-deposited). This suggests that the <sup>7</sup>Be ions are trapped in the metal oxide surface layer, indicative of a chemical interaction with the surface. Such a process was previously unknown, with the exception of the atomic oxygen effect<sup>1</sup>.

In Table 1, the measured number of <sup>7</sup>Be atoms per unit area on various spacecraft surfaces is shown. The results are corrected to the retrieval date of 12 January 1990 and for the offset angle from the leading direction. The areal density for <sup>7</sup>Be on the aluminium and steel is the same within the experimental uncertainty, and is apparently not a strong function of the type or surface condition of the metal. The Teflon thermal coating, however, which was used on many LDEF experiment trays, has a density of <sup>7</sup>Be an order of magnitude lower than that found on the aluminium surface. The reason for this apparent difference in uptake efficiency is unknown, but could be related to the material's covalence-bond structure. The explanation may be complicated, also, by the observed erosion of the Teflon surface by atomic oxygen.<br>The appearance of <sup>7</sup>Be on the leading surfaces, as shown in

Figs 1-3, rules out direct production of the isotope within the spacecraft by the incident radiation flux. In striking contrast to the distribution of <sup>7</sup>Be, the increased flux of geomagnetically trapped protons from the west in this type of orbit<sup>2</sup> results in higher spallation-induced activities on the trailing side of the spacecraft (see Figs 2 and 3). Such induced activity is also not



Corrected for decay since recovery and for surface orientation relative to spacecraft ram direction

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confined to the surface. Although we do observe a small <sup>7</sup>Be signal from aluminium samples taken from the trailing side of the spacecraft, this contribution is at a level we expect from reactions with the incident flux, and is about two orders of magnitude smaller than the surface activity of the leading side. This leads us to conclude that the  $7B$ e must have accumulated



FIG. 2 Gamma-ray spectra of aluminium plates on the leading and trailing sides of LDEF. The isotope <sup>7</sup>Be is identified by the strong line at 478 keV seen on the leading side but not on the trailing side. The strong line at 511 keV is produced by positron-annihilation gamma rays from the spallation product <sup>22</sup>Na, as well as laboratory background. Both spectra represent 48-h counts.

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on the spacecraft surfaces from the ambient atmosphere at orbital altitudes.

The short-lived isotope <sup>7</sup>Be was first detected in the atmosphere by Arnold and Al-Salih in 1955<sup>3</sup>, and later mapped by others as a function of altitude and latitude<sup>4-8</sup>. It is produced in the atmosphere by high-energy cosmic-ray interactions with air as are other radioisotopes such as <sup>14</sup>C and <sup>3</sup>H. Once formed, <sup>7</sup>Be ions are presumed to oxidize rapidly and attach to small aerosol particles, providing a downward transport mechanism from peak production regions of the atmosphere<sup>9-16</sup>. The primary removal process for  ${}^{7}$ Be, which occurs on a timescale comparable to its mean lifetime ( $\sim$ 77 days), is the washout of the aerosol-attached <sup>7</sup>Be in rain water<sup>3-6</sup>.

At a given latitude above  $\sim$  20 km, the production rate of <sup>7</sup>Be varies vertically in proportion to the oxygen-nitrogen gas density. Peak production per unit volume occurs in the lower stratosphere, at  $\sim$ 20 km, below which the cosmic-ray flux is substantially attenuated. At higher altitudes, the number of 'Be atoms produced per unit volume decreases rapidly, but the number of <sup>7</sup>Be atoms per unit mass of air, or concentration, should be essentially constant. Balloon and aircraft measurements<sup>6,15</sup> are in approximate agreement with this, although few measurements extend much above the peak production altitudes.

We can calculate the concentration of  ${}^{7}$ Be at 310 km from the data in Table 1, assuming the trapping efficiency on the metal surfaces is near unity. Using the LDEF orbital velocity of 7.8 km  $s^{-1}$  and the 77-day mean lifetime of  $7$ Be, we find a density of  $1.1 \times 10^{-7}$  atoms cm<sup>-3</sup>, or a relative concentration of  $3.8 \times 10^{6}$ atoms per gram of air. In the peak production region, at altitude 20 km, previous measurements<sup>4-8</sup> yield a concentration of  $10^3$ <sup>7</sup>Be atoms per gram of air, or  $\sim 0.1$  atoms cm<sup>-3</sup>, in agreement with a simple calculation using the known cosmic-ray flux. Thus, the measured concentration of  $7$ Be per unit mass of air at 310 km is three to four orders of magnitude in excess of the concentration at 20-50 km.

The concentration of  ${}^{7}$ Be in the 300-km range, far in excess of what can be produced in air at that altitude, is evidence of an unknown mechanism transporting <sup>7</sup>Be from the stratosphere. Although the amount being transported (or removed) from the peak production regions is a minute fraction of the total atmospheric burden of  ${}^{7}$ Be, the observed density is contrary to assumptions of conventional atmospheric transport and mixing, and in particular, to the assumed attachment to aerosol particles<sup>16</sup> Whatever the mechanism, however, the transport to orbital altitudes must take place on a timescale similar to the mean lifetime of  ${}^{7}$ Be.

Systematic low-level induced radioactivity measurements of



FIG. 3 Gamma-ray spectra of two stainless steel trunnion pin-end pieces from the leading and trailing directions of the spacecraft. As in Fig. 2, the <sup>7</sup>Be isotope is seen only on the leading side of LDEF. The 835-keV line from<br>the spallation product <sup>54</sup>Mn is observed on both sides of the spacecraft, but is stronger on the trailing side due to increased proton flux from the west. Both spectra represent 12-h counts.

LDEF materials have not revealed other nuclides with similar surface segregation behaviour; in particular, we have not found the heavier ones that would suggest a meteoritic origin<sup>17</sup> Whereas <sup>7</sup>Be will decay in orbit, there are other non-radioactive light isotopes produced by cosmic rays with similar altitude distributions. Their concentrations should be much higher than that of <sup>7</sup>Be, although more difficult to measure. We are currently attempting to detect other atmospheric cosmic-ray-produced isotopes on LDEF surfaces, such as <sup>14</sup>C and <sup>10</sup>Be, using accelerator mass spectrometry (A. J. T. Jull, personal communication).

The use of satellite surfaces to sweep up rare atmospheric species may prove to be a new method of investigating atmospheric mixing processes at orbital altitudes.  $\Box$ 

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## Low surface resistance in  $YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub>$  melt-processed thick films

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A LOW surface resistance,  $R_s$ , is the key to successful development of radio-frequency and microwave applications of hightemperature superconductors. Here we report the  $R_s$  of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> thick films on yttria-stabilized zirconia substrates at frequencies up to 50 GHz. Films processed below the peritectic temperature are fine grained, have  $R_s$  similar to bulk YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub>, generally have low critical current density  $(J_c)$  and exhibit little preferred orientation of crystallographic axes. Films processed above the peritectic temperature exhibit preferred orientation in large spherulitic grains, have higher  $J_c$  and far lower  $R_s$ . For these films the crossover frequency at which  $R_s$  equals that of copper is 50 GHz, a factor of two higher than the best bulk material or thick film yet reported and only a factor of  $\sim$  4 lower than highquality thin films. At the frequencies used for mobile communications (900 MHz and 1.8 GHz), the superconductor losses would be two orders of magnitude lower than those of normal metals. The particular advantages of the thick-film route are the speed and low cost of the process, and the ability to apply the films on curved surfaces and on large areas, the largest so far being  $>200 cm^2$ .

Because the of the quadratic frequency dependence of  $R_s$  in superconductors<sup>1</sup>, as opposed to the square-root dependence in normal metals, there is a great incentive to reduce  $R<sub>s</sub>$  in superconductors so that operation of devices below the cross-over frequency benefit from a lower  $R_s$  than that of a normal metal. Measurements of  $R_s$  so far can be divided into two broad groups: those made on bulk<sup>2</sup> or thick-film samples prepared from powder and those on thin films prepared by sputtering, laser ablation or co-evaporation. The thin films, which are usually grown epitaxially with the  $c$  axis normal to the plane of the substrate, show losses that are typically lower by two orders of magnitude relative to bulk materials<sup>2</sup>. It is believed that the increased losses in bulk material arise from the weak links between the random array of crystallites which also reduce critical currents'.

Our work was motivated by the need to provide an inexpensive, rapid and effective alternative to thin films for certain applications, and which can be used when thin-film methods are not possible for reasons of size or curvature. The thick-film process also has inherent advantages in that polycrystalline substrates of large size ( $\sim$ 300 mm square) are readily available. The best thick films so far have been formed on silver substrates by electrophoretic deposition<sup>4-6</sup>, and have an  $R_s$  at 21.5 GHz of  $18 \pm 3$  m $\Omega$  at 77 K. In that work, however, problems arose because of thermal mismatch with the substrate, causing spalling of the films; also, because silver was used as the substrate, the maximum processing temperature could not exceed  $\sim$ 930 °C. Our technique avoids these problems and gives extremely adherent films which have the lowest  $R_s$  for thick films yet reported.

The substrates used were comprised of polycrystalline 3 mol%

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