Enhanced Electron-Capture Decay Rate of ^{7}Be Encapsulated in C_{60} Cages

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The decay rate of ^{7}Be electron capture was measured in C_{60} and Be metal with a reference method. The half-life of ^{7}Be endohedral C_{60} (^{7}Be@C_{60}) and ^{7}Be in Be metal (Be metal (^{7}Be)) is found to be 52.68 ± 0.05 and 53.12 ± 0.05 days, respectively. This amounts to a 0.83% difference in electron-capture decay half-life between ^{7}Be@C_{60} and Be metal (^{7}Be). Our result is a reflection of the different electron wave functions for ^{7}Be@C_{60} inside C_{60} compared to the situation when ^{7}Be is in a Be metal.

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There is a longstanding interest in how nuclear decay rates, in particular β-decay rates, can be changed artificially because it leads to information about the electron wave functions of the medium surrounding the decaying nucleus and because of the (somewhat remote) possibility of changing the decay rates of radioactive waste products. As first suggested by Segré *et al.* [1–3], electron-capture (EC) decay rates depend on the density of atomic electrons within the nucleus. External factors such as chemical form and pressure may alter the electron overlap densities with the nucleus and thus affect the electron-capture decay rates. The nucleus ^{7}Be is a good candidate to use to look for external electron density effects on decay half-lives because of its simple electronic structure, $1s^2 2s^2$, in the EC-decaying atom. The ^{7}Be atom decays directly to the $3/2^-$ ground state of ^{7}Li with a branching of 89.6%, and to the first excited state in ^{7}Li ($1/2^-$ at 478 keV) with the remaining branching of 10.4%. This excited state decays by γ emission to the ground state [4]. In recent studies, there have been several observations and/or calculations of variations in half-life with the host metals [5–10], chemical forms [11–14], and pressure [15,16]. In most of these environments, the half-life is longer than 53.10 days.

After the discovery of C_{60} and the subsequent successful production of large amounts of fullerenes [17,18], it was found that large endohedral fullerenes, such as M@C_{82} (M = metal), can be created simultaneously with ordinary fullerenes through arc-discharge vaporization of composite rods made of graphite and the metal compounds [19,20]. However, the production rate of endohedral C_{60} is quite low compared to ordinary C_{60} and even to M@C_{82}. An alternative way of producing endohedral C_{60} is to insert foreign atoms into the cages of the preexisting C_{60} after their formation [21–27]. We have examined the formation of endohedral fullerenes by a nuclear recoil implantation of several foreign atoms following nuclear reactions[24–27]. We found that ^{7}Be can be endohedrally doped to create the ^{7}Be endohedral C_{60} (^{7}Be@C_{60})[27].

Because of the unique chemical form of C_{60}, the electron contact density on ^{7}Be nuclei is affected significantly by the electron density of C_{60}. Therefore, it is interesting to investigate the question: "how does the electron-capture decay rate in ^{7}Be change inside the C_{60} cage relative to other situations?" Here, we compare the half-life of ^{7}Be when it is encapsulated in C_{60} to that of ^{7}Be in Be metal as a reference. To accurately calibrate our time measurements, we rely on a standard clock time radio signal. We find a surprisingly short half-life of ^{7}Be in C_{60}.

To allow us to measure the EC-decay rate of ^{7}Be inside C_{60}, we produce ^{7}Be@C_{60} by a nuclear recoil implantation technique (method (1)). As to a reference sample, another method (2) was employed to produce Be metal (^{7}Be) samples.

(1) To produce ^{7}Be@C_{60}, Li_2CO_3 was used in powder form. The grain size of the material was smaller than 100 meshes (20 μm). Purified fullerene (C_{60}) was carefully mixed with Li_2CO_3 (weight ratio = 1:1) in an agate mortar, adding a few ml of carbon disulfide (CS_2). After drying up, about 50 mg of the mixture sample was wrapped in a pure aluminum foil of 10 μm in thickness for irradiation. Proton irradiation with a beam energy of 16 MeV was performed at the Cyclotron Radio-Isotope Center, Tohoku University. The beam current was typically 3 μA and the irradiation time was about eight hours. The reaction ^{7}Li(p, n)^{7}Be then leads to the production of ^{7}Be. After the kinetic energy of ^{7}Be has decreased in the sample to an appropriate value, it then penetrates into the C_{60} cage to produce the endohedral fullerene [27]. After irradiation, the sample was dissolved in CS_2 and filtered through a millipore filter (pore size = 0.2) to remove insoluble materials. The soluble portion was injected into a High Performance Liquid Chromatography (HPLC) device equipped with a 5 PPB (Cosmosil) (silica-bonded with a pentabromobenzyl group) column of 10 mm (in inner diameter) and 250 mm (in length) at a flow rate of 2 ml/min. To confirm the presence of fullerenes and their derivatives, a UV detector was installed with a wavelength of 400 nm. Eluent fractions were collected for

30 sec intervals (0–30, 30–60, 60–90,...sec). Figure 1 shows for materials inserted into C_{60} samples, a radiochromatogram measured with a γ detector (solid circles) and a chromatogram measured with an UV detector (solid line), both plotted versus retention time after injection. A clear correlation between the UV-absorption intensity and the γ counting rate in the 8.5–10.5 min interval is seen in Fig. 1. From the correlation of the elution behavior between the UV chromatogram and the radioactivities of the ^{7}Be atoms, we found that the atom-doped fullerene ^{7}Be@C_{60} was indeed produced by nuclear recoil implantation. The successful insertion of Be atom into C_{60} with an *ab initio* molecular simulation was also inserted in the figure [27]. The fraction that corresponded to C_{60} was deposited on a sample holder where the CS_2 solvent was allowed to dry up so that a measurement sample was obtained.

(2) The production of Be metal (^{7}Be) also involved a series of different procedures. Be metal, a hexagonal close-packed (hcp) structure, of 10 mm (in diameter) $\times$0.3 mm (in thickness) was used to produce ^{7}Be uniformly in the metal. The Be metal was sealed in a quartz tube of 12 mm in diameter which was then used as a target. The irradiation with bremsstrahlung (50 MeV electrons) was carried out at the Electron Linear Accelerator, Laboratory of Nuclear Science, Tohoku University. The sample in the quartz tube was set in the middle of a sweep magnet placed on the axis of the electron beam. A 2 mm thick platinum converter was set in front of the sweep magnet to generate bremsstrahlung. Then, the sample was irradiated only by the bremsstrahlung (all electrons were diverted by the magnetic field). Therefore, the damage to the Be metal lattice was minimal. The ^{7}Be isotope is produced in the ^{9}Be(γ, 2n)^{7}Be reaction in the Be metal. After irradiation, the sample was baked in an electric oven with 1100 °C for 1 hour in an attempt to repair the lattice defects that might be induced by the (γ, 2n) reaction. Finally, the sample was washed with HCl solution to clean the surface.

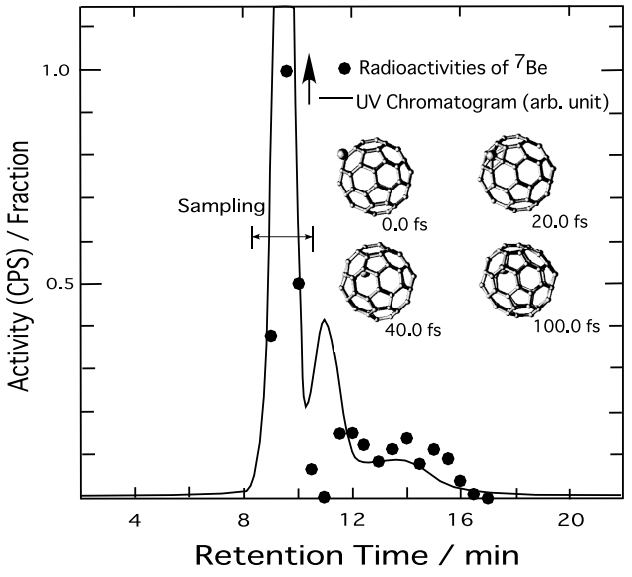

FIG. 1. HPLC elution curves of the soluble portion of the crude extracted in the proton irradiated sample of ^{7}Li and C_{60}. The vertical axis shows the radioactivities of ^{7}Be in each fraction (solid circles) and an absorbance of UV chromatogram of C_{60}. The inserted figures are time-ordered snapshots of a simulation taken from Ref. [27], where ^{7}Be hits the center of a six-membered ring of C_{60} with 5 eV kinetic energy. One can see successful production of ^{7}Be@C_{60}.

The two samples of ^{7}Be@C_{60} and Be metal (^{7}Be) were placed in an automated sample changer, which vertically moved the samples in front of a γ-ray detector. This allowed the decay rates of the two samples to be measured in a comparable way. The ^{7}Be@C_{60} sample was moved in front of the detector and its γ rays were measured for a specific time T_d, precisely $T_d = 21\,570$ sec. Then the samples were interchanged and the Be metal (^{7}Be) sample was moved in front of the detector (T_m = 30 sec) and measured for exactly the same length of time. The $T_d + T_m$ is exactly 21 600 sec (6 hours). This procedure was then repeated 330 times over 170 days. The internal clock time of a computer for data acquisition was always calibrated by a time-standard signal distributed via a long-wave radio center in Japan. (*i.e.*, The starting time for each run was correlated to a time distributed publicly.) Therefore, the uncertainty in the time measurements is negligibly small. The system was completely controlled by a computer to obtain precise measurement positions. The reproducibility of each position was determined to be within 0.1 mm. Systematic errors were reduced by using the metal (^{7}Be) as a reference. We prepared the two samples so that we obtained similar and uniform concentrations of ^{7}Be. Furthermore, the temperature was kept constant at 20 °C by air conditioner. The activities associated with ^{7}Be, in particular, the 478 keV γ rays emanating from the EC-decay daughter of ^{7}Be, were measured with a high-purity germanium (HPGe) detector (ΔE_{FWHM} is 1.8 keV and 50% relative efficiency) coupled to a 2048-channel pulse-height analyzer. The excellent energy resolution of the HPGe detector resulted in a good signal-to-noise ratio. The background was reduced by a lead shield. Therefore, the background peaks do not impair the determination of the half-life of ^{7}Be in the present experiment. The activities associated with the decay of ^{7}Be could be uniquely detected through the identification of characteristic γrays, and all other sources could be ruled out.

A typical γ-ray spectrum obtained in a measurement of ^{7}Be@C_{60} decay as a function in keV is shown in Fig. 2. The expected γ line at $E_\gamma = 478$ keV and a natural background line at $\gamma = 1461$ keV can be seen as two giant peaks. No peak was seen at around $E_\gamma = 478$ keV when the ^{7}Be sources were absent. A direct summation

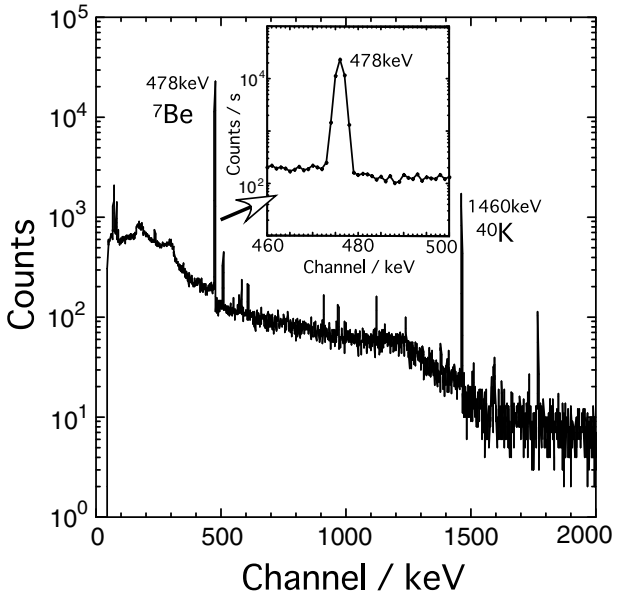

FIG. 2. Typical γ-ray spectrum of ^{7}Be in the ^{7}Be@C_{60} fraction.

method, which is normally used in activation analysis, was applied to obtain the peak area ($E_\gamma = 478$ keV) and the background was taken to be a straight line between the average counting number/channel on both sides of the peak. In Fig. 3, the two measured exponential decay curves for the ^{7}Be@C_{60} and Be metal (^{7}Be) activities are plotted versus time. Red and blue circles, respectively, indicate the radioactivities of ^{7}Be@C_{60} and Be metal (^{7}Be) samples. The initial radioactivities of the ^{7}Be in each sample were almost identical, *i.e.*, at time zero we obtained around 2.7 counts/s (cps) for ^{7}Be@C_{60} and 2.5 cps for Be metal (^{7}Be). So that the two decay curves can be compared, the data for Be metal (^{7}Be) shown in Fig. 3 were normalized to that for ^{7}Be@C_{60} by use of an adjustment procedure on a few initial points of the decay curve. The decay constants for the two samples were obtained by fitting straight lines to the measured data points by use of a MINUIT program distributed from the CERN Program Library. This program takes into account the statistical error associated with each data point in Fig. 3. This statistical error is by far the dominating uncertainty. The uncertainty of our measurement is given by the uncertainty of the slope of the straight line fitted to the logarithm of the counts. The results for the sample ^{7}Be@C_{60} and Be metal (^{7}Be) are $T_{1/2} = 52.68 \pm 0.05$, and $T_{1/2} = 53.12 \pm 0.05$ days, respectively. The dead time in the data acquisition system has been found to be about 8 or 9 sec for each 21 570 sec run. Therefore, the uncertainty due to the dead time is slightly less than 0.04%. (In order to reduce the dead time of the measurement system, the amount of the activities and the distance between a γ detector and the source were prepared suitably in the measurements.) This systematic error is about half of the statistical error quoted above.

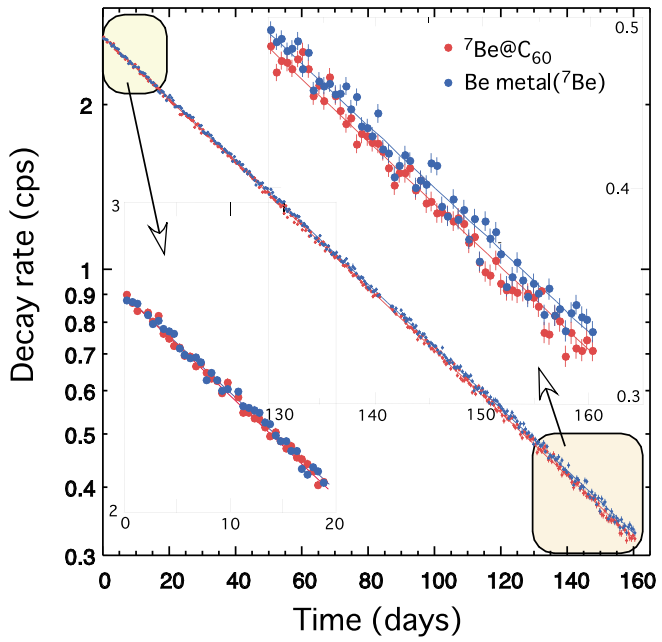

FIG. 3 (color). Exponential decay curves of ^{7}Be in samples of ^{7}Be endohedral C_{60} and Be metal (^{7}Be). Insets corresponding to the decay intervals of 0 ~ 20 days and 130 ~ 160 days are displayed with an expanded scale.

The half-life obtained in the sample of Be metal (^{7}Be) is $T_{1/2} = 53.12 \pm 0.05$ days. The half-life values obtained for ^{7}Be in several other host materials such as graphite and boron nitride have been presented by Jaeger and Norman *et al.* [5,8]. The value ($T_{1/2}$) is always in the range 53.1 to 53.2 days except for the case of gold (53.31 days)[8]. In Table I, half-lives previously measured are also shown as a comparison. Here, only the half-lives obtained using a standard time distributed publicly (stated in the text) are listed[5,8]. Therefore, we find that our reference measurement of ($T_{1/2}$) of ^{7}Be for Be metal (^{7}Be) is in satisfactory agreement with other available data.

TABLE I. The half-life ($T_{1/2}$) of ^{7}Be in the host of Be metal as determined with a least-squared fit. Half-lives previously measured in several host materials are also shown as a comparison.

Host materials	$T_{1/2}$ (days)	References
C_{60}	52.68 ± 0.05	This work
Beryllium metal	53.12 ± 0.05	This work
Lithium fluoride	53.12 ± 0.07	[5]
Graphite	53.107 ± 0.022	[8]
Boron nitride	53.174 ± 0.037	[8]
Tantalum	53.195 ± 0.052	[8]
Gold	53.311 ± 0.042	[8]

It is surprising to observe that the half-life obtained for ^{7}Be in the sample ^{7}Be@C_{60}, $T_{1/2} = 52.68 \pm 0.05$ days (see Table I), is as much as 0.83% shorter than for the Be metal (^{7}Be) sample, where we define the percentage difference by $[100 \times (\lambda(C_{60}) - \lambda(\text{Be metal})]/\lambda(\text{Be metal}))$. This difference in half-lives is sufficiently large so that it is clearly visually discernible when the data are displayed on the extended scale of the right inset in Fig. 3. The half-life of ^{7}Be in the C_{60} is shorter than any previously reported for any material or pressure. This implies that the ^{7}Be atoms are located in a unique environment inside the C_{60} cages. Several factors contribute to give rise to this unique environment; for example, the many π electrons of C_{60} and the special dynamic conditions of the electrons inside the C_{60} cage, which include ratchet and/or tumbling motion [28–30], all which affect the contact electron density at the ^{7}Be nucleus. Here, a magnitude of the average charge-transfer from $2s$ electrons of ^{7}Be atom, *e.g.*, K and L capture [31,32], can play an important role for such a large variation in the ^{7}Be decay constant between ^{7}Be@C_{60} and Be metal (^{7}Be).

In summary, we have measured the half-life of ^{7}Be (1) encapsulated in C_{60} and (2) incorporated in Be metal using a HPGe detector with a time reference from a standard-time radio signal. We found that the half-life of ^{7}Be in C_{60} and Be metal was $T_{1/2} = 52.68 \pm 0.05$ and $T_{1/2} = 53.12 \pm 0.05$ days, respectively. This 0.83% difference between the EC rate in C_{60} and in Be metal represents a strong environment effect on the ^{7}Be EC capture rate, caused by the different electronic wave functions near the ^{7}Be nucleus inside a C_{60} cage and inside Be metal. Since we have found a record-large environment effect on the decay constant, it is our belief that these results will be very useful in further experimental and theoretical studies of environmental effects on radioactive decay and, for example, help identify other favorable environments for large decay-constant changes.

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