Half-life measurement of ^7Be in host beryllium metal

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Summary. The decay rate of an electron-capture nucleus is proportional to the electron density at the nucleus. To see how the decay rate is changed by artificially, we have measured the half-life of ^7Be in beryllium(Be) metal at room temperature ($T=293\text{ K}$) and at close to the temperature of liquid helium ($T=5\text{ K}$). We found that the half-life of ^7Be in Be metal at $T=293\text{ K}$ is $53.25 \pm 0.04\text{ d}$, which is slightly longer than those in the hosts of graphite, lithium fluoride and other minerals, surveyed so far. Furthermore, that at $T=5\text{ K}$ is $53.39 \pm 0.04\text{ d}$, which is 0.26% longer than that at $T=293\text{ K}$.

1. Introduction

The constancy of nuclear half-life has been firmly established experimentally as an exponential decay law. However, There is a long-standing debate about the degree to which nuclear decay rates can be changed artificially. In one of the β -decay modes, the electron-capture (EC) decay rate depends on the density of atomic electrons within the nucleus as first suggested by Segrè *et al.* [1, 2]. The general understanding is that the wave function of the initial and final states between atomic electrons and the nucleus, respectively, should be considered in the decay mode. The EC decay nucleus ^7Be has been often used to look for effects of external-electron density on decay half-lives, by virtue of its simple electronic structure ($1s^2 2s^2$) in neutral Be atom and the adequacy of its half-life for measurement. Therefore, some studies have reported on how external factors, such as chemical form [3–6], host metal [7–10], pressure [11, 12], and even temperature [13] alter the decay rate(half-life).

Recently, we have produced several radioactive fullerenes [14–16], such as the ^7Be endohedral C_{60} ($^7\text{Be}@\text{C}_{60}$) [14] and ^7Be in Be metal (Be metal(^7Be)). We have measured the half-life of ^7Be in the sample of $^7\text{Be}@\text{C}_{60}$ and Be metal(^7Be) by using a reference method and standard clock time. It was revealed that the half-life of ^7Be inside C_{60} at $T=5\text{ K}$ was almost 1.5% shorter than that in Be metal at room temperature [13, 17]. This implied that the ^7Be atoms are located in a unique environment inside C_{60} . This large change in the half-life is firmly caused by the electron dens-

ity at the nucleus position and the magnitude of the change has also been explained theoretically [18].

Because of the uniform lattice structure (*hcp*) included ^7Be in Be metal(^7Be), the EC decay rate of the ^7Be nucleus in the structure is also essentially intriguing and should be surveyed in order to compare any other situations in chemical forms, pressure, and even temperature. In the present study, we have measured the half-life of ^7Be in Be metal(^7Be) at both room temperature ($T=293\text{ K}$) and close to the temperature of liquid helium ($T=5\text{ K}$) by using a reference method. We compare the half-life of ^7Be in Be metal(^7Be) at $T=293\text{ K}$ and 5 K , as well as in other materials.

2. Experimental procedure

Be metal (*hcp* lattice structure) 10 mm in diameter and 0.3 mm in thickness was utilized to produce ^7Be uniformly in the Be metal. After being washed with weak HCl solution, the Be metal was sealed in a quartz tube (vacuum packed) that was 12 mm in diameter as a target. Irradiation with a bremsstrahlung (generated by 50 MeV electrons) was carried out at the Electron Linear Accelerator, Laboratory of Nuclear Science, Tohoku University. The experimental setup for the irradiation is shown in Fig. 1. The sample in a quartz tube was set in the middle of a sweep magnet placed on the axis of the electron beam. A platinum plate converter 2 mm in thickness was set in front of the sweep magnet in order to generate a bremsstrahlung. Then, the sample was irradiated only by the bremsstrahlung, and all the electrons were ruled out by the magnetic field (see Fig. 1). Therefore, the damage to the lattice of Be metal was confined to the minimum. ^7Be can be produced in Be metal uniformly by the photonuclear reaction $^9\text{Be}(\gamma, 2n)^7\text{Be}$. After irradiation, the sample was baked in an electric oven with vacuum packing at 1150 degrees C (the melting point of Be metal is 1278 degrees C) for a few hours to recover the lattice defect even if the defect occurs by nuclear reactions. Finally, the sample was washed again with weak HCl solution to clean the surface.

Two samples of Be metal(^7Be) were prepared in the present experiment. To measure the half-life at $T=293\text{ K}$ and 5 K , one sample of Be metal(^7Be) was placed at the top of a helium (He) closed-cycle cryostat, and the other was placed at the top of a sample holder, in an automated sam-

Fig. 1. Setup for irradiation of Be metal using the Electron Linear Accelerator at Tohoku University.

Fig. 2. Measurement system; computer controlled sample changer.

ple changer, respectively. The samples could then be moved precisely in front of a γ -ray detector. This allowed the decay rates of the two samples to be measured in a comparable way. The system is shown in Fig. 2. The ^7Be decays directly to the $3/2^-$ ground state of ^7Li with a branching of 89.6%, and goes to the first excited state in $^7\text{Li}(1/2^-$ at 478 keV) with that of 10.4% which decays by γ emission to the ground state [19]. The activities of the 478 keV γ -rays emanating from ^7Be were measured with a high-purity ger-

manium (HPGe) detector (its resolution was 1.7 keV with 50% of the efficiency of a standard NaI detector) coupled to a 4096-channel pulse-height analyzer. Due to the excellent energy resolution of the HPGe-detector, a good signal-to-noise ratio was obtained. The background was reduced by a lead shield. Therefore, the background peaks did not impair the determination of the half-life of ^7Be in the present experiment. The radioactivities of ^7Be were uniquely detected by means of its characteristic γ -rays, and any other sources were ruled out. Here, we set the measurement duration to $T_d=21\,600$ seconds (21 480 seconds for the live measurement time and 120 seconds for the dead time of the measurement system plus the sample exchange time) for one data point. The total measuring time was 166 d, which is more than three half-lives of ^7Be . The start time was taken from a time standard signal distributed *via* a long-wave radio center in Japan. Therefore, the uncertainty in the time measurements can be neglected. Furthermore, the low-temperature and high-temperature samples were alternatively measured each for approximately 6 h by turns. Therefore the systematic uncertainties are properly reduced in the measurements.

3. Results and discussion

A typical γ -ray spectrum obtained in the sample of Be metal(^7Be) at $T=293$ K is shown in Fig. 3. The decay scheme of ^7Be is also shown in the figure. The expected γ line at $E_\gamma=478$ keV and the natural background of ^{40}K γ line at $E_\gamma=1461$ keV can be seen as two giant peaks. No peaks were seen at around $E_\gamma=478$ keV when the ^7Be source was absent. The exponential decay curve of the ^7Be activities for the sample of Be metal(^7Be) at $T=293$ K is also shown as a function of time (days) in Fig. 4. The decay curve obtained in the present measurement was fitted including statistical errors by a Minuit program distributed from the CERN Program Library. The statistical error dominates the uncertainty in each data point in Fig. 4. The uncertainty of our meas-

Fig. 3. Typical γ -ray spectrum for the ^7Be in the sample of Be metal(^7Be) and decay scheme of ^7Be nucleus.

Fig. 4. Exponential decay line of ^7Be in a sample of Be metal(^7Be). Background count rate of radioactivities for ^{40}K are also shown.

urement is given by the uncertainty of the exponential line fitted to the counts (*i.e.* counts per second) of the decay spectrum. The change in the dead time in the data acquisition system is evaluated to be about 8–15 sec for all running case. Therefore, the uncertainty due to the dead time is estimated to be less than 0.04% and this value is smaller than the fitting errors of the half-life of ^7Be . For the sample of Be metal(^7Be) at $T=293\text{ K}$, the half-life was also obtained not only for this run but also as a reference run for that of ^7Be in the sample of $^7\text{Be}@C_{60}$ [13, 17]. Therefore, the averaged half-life of $T_{1/2}=53.25\pm0.04\text{ d}$ was precisely determined for the sample of Be metal(^7Be) at $T=293\text{ K}$. In the sample of Be metal(^7Be) at $T=5\text{ K}$, a half-life of $T_{1/2}=53.39\pm0.04\text{ d}$ was obtained in the present experiment. The counting rates of the natural background emanating from ^{40}K (the 1461 keV γ -rays) are also shown in Fig. 4. The obtained value in counts/s for ^{40}K was also fitted using the same procedures. It was found that the fitted line infinitely corresponds to a horizontal line. This also secures the reliability of our measurements.

In the present study, the half-lives of ^7Be in the samples of Be metal(^7Be) at both $T=293\text{ K}$ and 5 K were determined as $T_{1/2}=53.25\pm0.04\text{ d}$ and $T_{1/2}=53.39\pm0.04\text{ d}$, respectively, as seen in Fig. 5. The observed change is $0.14\pm0.06\text{ d}$. This difference is almost a one sigma. The half-life values obtained for ^7Be in several other host materials such as graphite and boron nitride have been reported so far [7–9]. In Fig. 5, the half-lives previously measured are also plotted as a comparison. The values stay in the range of about 53.1–53.3 d. We find that the half-life value of ^7Be in Be metal(^7Be) by the measurement at $T=293\text{ K}$ may be almost in agreement with the available data, but can be slightly longer than the value for graphite, boron nitride *etc.* Furthermore, the figure clearly shows that the half-life of ^7Be in the Be metal(^7Be) at $T=5\text{ K}$ is 0.26% longer than that in the Be metal(^7Be) at $T=293\text{ K}$, and is also longer than any ^7Be half-

Fig. 5. Half-life values of ^7Be in Be metal(^7Be) as determined with a least-squared fit (solid circle for $T=293\text{ K}$ and solid square is for $T=5\text{ K}$). Half-lives previously measured are also shown for comparison [7–9].

Fig. 6. Schematic view of the CRIB facility installed by the Center of Nuclear Science, University of Tokyo, at RIKEN [20].

life in metals reported in any environment up to now. We suppose that this can be due to the temperature dependence of a lattice constant in the structure (*hcp*) of Be metal. We are investigating this by theoretical calculations.

We previously measured the half-life of ^7Be in the sample of $^7\text{Be}@C_{60}$. We found that the half-life of ^7Be in a sample of $^7\text{Be}@C_{60}$ at a temperature close to $T=5\text{ K}$ is $52.47\pm0.04\text{ d}$, this value is 1.5% shorter than that ^7Be in the Be metal(^7Be) at $T=293\text{ K}$ [13]. Therefore, we realized 1.75% of the difference between the half-life of ^7Be in $^7\text{Be}@C_{60}$ and that in the Be metal(^7Be) under $T=5\text{ K}$.

It is further intriguing to study the temperature dependence of the half-life of ^7Be inside several metals. In our future study, in order to find the effects of temperature inside metals (*e.g.* superconducting metals), we will measure the half-life of ^7Be in the sample of Nb metal that is cooled to a temperature close to liquid He ($T=5\text{ K}$). The superconducting critical temperature of Nb metal is known to be 9 K. The ^7Be can be produced uniformly by the $^1\text{H}(^7\text{Li},n)^7\text{Be}$ reaction. The ^7Be beams produced by the reaction are directly utilized to implant ^7Be uniformly in Nb metal. The Center for Nuclear Study (CNS), of the University of Tokyo, has been developing an extensive low-energy RI beam facility (CRIB, see Fig. 6) of the in-flight method in a joint venture with RIKEN [20]. The Nb metal is set at the target position of the CRIB beam line. The implantation is carried out with ^7Be secondary beam of over $\sim 24\text{ MeV}$. The research program got under way in 2010.

4. Conclusion

We have produced the radioactive ^7Be nucleus in Be metal by the $^9\text{Be}(\gamma, 2n)^7\text{Be}$ reaction caused by a high energy bremsstrahlung. The half-life of ^7Be in Be metal(^7Be) was measured using a HPGe detector taking into account a standard time. We found that the half-life of ^7Be in Be metal(^7Be) at $T=293\text{ K}$ was $T_{1/2}=53.25\pm0.04\text{ d}$, and that of ^7Be in Be metal(^7Be) at $T=5\text{ K}$ was $T_{1/2}=53.39\pm0.04\text{ d}$. The difference in the half-life values of ^7Be in Be metal(^7Be) between $T=293\text{ K}$ and 5 K is 0.26%. The half-life value of ^7Be in Be metal(^7Be) at $T=5\text{ K}$ is longer than any ^7Be half-life in metals reported in any environment.

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