Effect of pressure on the decay rate of ^{7}Be

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Abstract

Beryllium-7 in Be(OH)$_2$ gel was compressed in diamond-anvil pressure cells up to 442 kbar at room temperature. By counting the activity of ^{7}Be, the decay rate for the conversion of ^{7}Be to ^{7}Li via electron capture was measured. The decay constant of ^{7}Be, λ, was found to increase, but the rate of increase decreased with increasing pressure. A quadratic regression of the data yields $(\lambda - \lambda_0)/\lambda_0 = (4.87 \times 10^{-5})P - (5.9 \times 10^{-8})P^2$, where the subscript zero denotes zero pressure and P stands for pressure in kilobar. Thus, λ of ^{7}Be increases by about 1% at 400 kbar. The observed data set can be rationalized by an increase in electron density near the nucleus of ^{7}Be at high pressures. This result may bear some implications for the conversion of ^{40}K to ^{40}Ar, which has been widely adopted to date geological events. © 2000 Elsevier Science B.V. All rights reserved.

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1. Introduction

The suggestion that the decay constants of certain radioactive nuclides might be altered by varying the electron density near their nuclei was proposed independently by Daudel [1] and Segré [2] more than half a century ago. Attempts have been made to observe this effect, by changing either the chemical or the physical environment of nuclides that decay by electron capture or internal conversion. As the lightest radioactive nuclide undergoing electron capture decay and with a half-life of $\sim$ 53 days, ^{7}Be is the most suitable nuclide for such study. There have been several studies of the decay rate of ^{7}Be using various chemical forms,

and the results have recently been summarized and updated by Huh [3]. However, there have been only two studies involving high pressures and both were carried out more than 25 years ago [4,5]. The technologies of both detecting γ-rays and measuring pressures have greatly improved since then. Using the present state-of-the-art technology, we have made much a more accurate and precise measurement of the decay rate of ^{7}Be at high pressures.

2. Experimental procedure

Carrier-free ^{7}Be (in 1 ml of 0.1 N HCl) with an activity of 1.1 mCi as of 9 November 1998 was purchased from the Hot Lab of Brookhaven National Laboratory (S/M number: 054911). Be(OH)$_2$ in gel form was prepared from 100 μl of the ^{7}Be stock solution by adding 5 mg of ^{9}Be

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(initially dissolved in 1 ml of 1 N HCl) as the [7]Be carrier followed by NH_4OH to form $Be(OH)_2$ precipitate. After centrifugation to separate the precipitate from the solution, the supernatant water was removed. About one half of the precipitate was used for this study. Aliquots of the sample were loaded into four different types of diamond-anvil pressure cells (A, B, C and D in Table 1), each covering a certain range of pressures. A hardened stainless steel gasket with a hole about 100–150 μm in diameter and 60–80 μm in depth was sandwiched between the two diamond anvils in each cell. Fine powders of ruby were then placed inside the hole, and extreme care was taken to load the sample inside the hole to ensure that none of the sample was left outside the pressure chamber. No pressure-transmitting medium other than $Be(OH)_2$ gel itself was used, and the whole assembly was sealed by driving the diamond anvils together. Two of the samples recovered after the high-pressure experiment (A and B) were later examined by X-ray diffraction and Raman spectroscopy. These samples appeared to be amorphous. An in situ Raman study of a newly prepared $Be(OH)_2$ gel at 250 kbar showed that the spectrum of the sample at high pressure was similar to that of the starting material, implying that the sample remained amorphous at high pressure. The latter phenomenon is in line with the pressure-induced amorphization of silicates [6] and oxides [7] generally observed at room temperature.

Pressures were measured using the ruby-fluorescence technique. The hydrostaticity of the sample at $P \leq 130$ kbar, as estimated from the shape of fluorescence bands of ruby, is close to that when a water pressure medium is used, and it is judged to be acceptable in the range 130–450 kbar. Based on 10–25 measurements evenly distributed in the pressure chamber of each sample, the experimental uncertainty in pressure varies from ± 1.5 kbar around 30 kbar to ± 14 kbar around 440 kbar (Table 1).

In order to accommodate the geometry and weight of the different pressure cells, a special stage was built to hold the pressure cells a few millimeters above the HPGe γ-ray detector (EG&G ORTEC GEM-150230), which was interfaced with a digital γ-ray spectrometer (EG&G ORTEC DSPec™). The detector offers a counting efficiency of 150% (relative to 3×3 NaI) and a resolution of 1.23 keV (FWHM) for the [7]Be photon peak centered at 477.56 keV. The counting system provides extended live-time correction according to the Gedcke–Hale method [8] and automatically sets a threshold to reject pile-up of pulse pairs at 0.5 μs resolution. The outline and orientation of each pressure cell were clearly marked on the stage so that the position, geometry and sample-to-detector distance remained constant from run to run for each pressure cell. The sample-to-detector distance ranged from ~5 to ~15 mm above the center of the detector for different pressure cells.

Table 1
Variation of the decay constant (λ) of [7]Be in $Be(OH)_2$ gel with pressure

Type of pressure cell	Pressure	Number of pressure measurements	Period of counting	λ	$T_{1/2}$	$\lambda-\lambda_0/\lambda_0$
	(kbar)		(days)	(day^{-1})	(days)	
None[a]	1 atm	–	110	0.0129769 ± 0.0000007	53.414 ± 0.003	0
A	28.2 ± 1.5	10	72	0.0129934 ± 0.0000023	53.346 ± 0.009	0.00127 ± 0.00018
A	46.8 ± 4.7	10	77	0.0130048 ± 0.0000046	53.299 ± 0.019	0.00215 ± 0.00036
B	92.3 ± 5.7	15	78	0.0130287 ± 0.0000040	53.202 ± 0.016	0.00399 ± 0.00031
B	131.5 ± 10.7	9	72	0.0130461 ± 0.0000036	53.131 ± 0.015	0.00533 ± 0.00028
C	145.3 ± 6.4	25	91	0.0130512 ± 0.0000010	53.110 ± 0.004	0.00572 ± 0.00010
D	233.5 ± 6.1	15	53	0.0130848 ± 0.0000046	52.973 ± 0.019	0.00832 ± 0.00036
D	311.3 ± 9.5	14	55	0.0131000 ± 0.0000047	52.912 ± 0.019	0.00949 ± 0.00037
C	441.7 ± 13.8	18	87	0.0131068 ± 0.0000053	52.884 ± 0.022	0.01001 ± 0.00041

[a] By mounting and covering a [7]Be–$Be(OH)_2$ source on a glass slide.

Activities of ^{7}Be in the sample at various pressures were assayed on a daily basis for one to two half-lives of ^{7}Be (see Table 1). To compensate for the decrease of count rate with time and maintain compatible counting statistics between runs, the counting time increased from ~ 20 min at the beginning to 3–4 h near the end of the experiment. The accumulated counts from ^{7}Be decay for each run were generally in the order of 10^{6}–10^{7}, resulting in typical counting errors of ~ 0.03–0.1% ($\pm 1\sigma$) for the data points. The starting sample at ambient conditions was also counted in a well-type HPGe detector (EG&G ORTEC GWL-100230) [3]. The decay rate of ^{7}Be in uncompressed sample obtained from these two different detectors is identical (0.0129769 ± 0.0000007 vs. 0.0129765 ± 0.0000014 day^{-1}), lending unequivocal support to the accuracy and precision of our experiment. The stability of the counting system was ensured by weekly calibration of the detector using a uraninite standard solution. Throughout the course of this experiment there were no signs of any changes in energy, resolution and efficiency. The electronic stability of the counting system was also monitored in some runs using the much longer-lived ^{152}Eu which was placed just outside the pressure chamber as a real-time calibration standard.

3. Results and discussion

A typical exponential decrease of ^{7}Be activity at high pressure (A_t), together with that of ^{152}Eu, with time is shown in Fig. 1. To simplify the plot, A_t is normalized against A_0 (^{7}Be activity at $t = 0$). Weighted least-squares fitting of the data sets results in the decay constant λ and the associated uncertainty ($\pm 1\sigma$). As mentioned earlier, uncertainties of our data points are $< 0.1\%$ ($\pm 1\sigma$) based on counting statistics. With 54–138 data points on the decay curve, the resulting uncertainty for λ is $< 0.04\%$ ($\pm 1\sigma$). The experimental conditions and results are summarized in Table 1. The fractional increase in the decay constant of ^{7}Be in Be(OH)$_2$ gel as a function of pressure obtained in the present study is shown in Fig. 2, in which the data of earlier studies are also

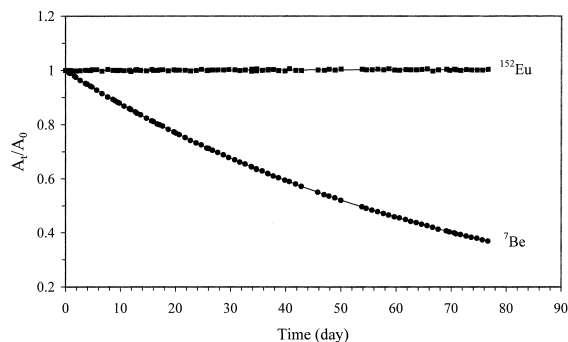Fig. 1. Exponential decay of ^{7}Be in Be(OH)$_2$ gel at 46.8 ± 4.7 kbar. The horizontal line ($A_t/A_0 = 1$) is decay-corrected and normalized activity of ^{152}Eu which was placed outside the pressure chamber of the diamond-anvil cell as a real-time calibration standard. Error bars for the A_t/A_0 values are actually much less than the size of the data points.

given. The uncertainty of the fractional increase in our measurement is denoted by the vertical error bars, which are sometimes smaller than the symbol size, and that of pressure is denoted by the horizontal error bars. It should be noted that the single datum point of Gogarty et al. [4] was calculated from a least-squares fit of 20 measurements near 100 kbar using ^{7}BeCO$_3$·Be(OH)$_2$ as the starting sample, and the three data points of Hensley et al. [5] were obtained using BeO. There-
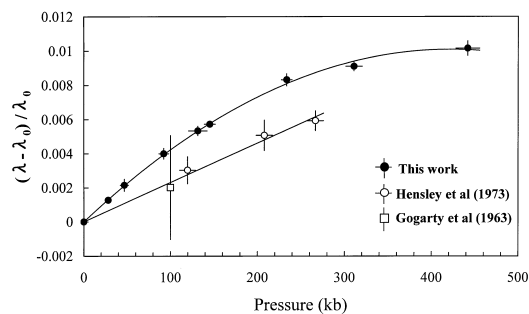

Fig. 2. Fractional increase of the decay constant of ^{7}Be in Be(OH)$_2$ gel as a function of pressure. The line through our data is the quadratic regression (see text). The data of Gogarty et al. [4] and those of Hensley et al. [5] are also shown for comparison. According to Huh [3], there is a small, but real, difference in the decay constant of ^{7}Be in different chemical forms. The present data were normalized using $\lambda_0 = 0.0129769 \pm 0.0000007$ day^{-1} for Be(OH)$_2$ gel. It is assumed that the data of Hensley et al. [5] were normalized using λ_0 for BeO.

fore, the effect of chemical forms on λ should be considered while comparing the data sets [3], as discussed below.

In the present study, not only were more measurements made, but the pressure spanned was more than 1.5 times that covered in the previous studies and much more accurate measurements of both decay constant and pressure were obtained. Fig. 2 shows that the initial fractional increase in the decay constant of ^{7}Be observed in the present study is about twice that reported earlier. However, in contrast to the linear behavior reported by Hensley et al. [5], the rate of increase was observed to decrease with increasing pressure in the present study. A quadratic regression of the data yields $(\lambda-\lambda_0)/\lambda_0 = (4.87\times 10^{-5})P-(5.9\times 10^{-8})P^2$ with the regression coefficient $R^2 = 0.999$, where the subscript zero denotes zero pressure and P stands for pressure in kilobar. Thus, λ of ^{7}Be increases by about 1% at 400 kbar.

The increase in decay constant in ^{7}Be at high pressures can be rationalized in terms of an increase in electron density near the nucleus at high pressures as predicted more than half a century ago. A rough calculation between the observed data and the compressional behavior of BeO at high pressures was carried out by Hensley et al. [5], though more rigorous theoretical calculations using more realistic models remain to be made. The different results between the present study and that of Hensley et al. may be interpreted as follows. If the electron density is inversely proportional to the volume, the lower rate of increase in the decay constant of ^{7}Be observed by Hensley et al. [5] may be due to the smaller compressibility of BeO, relative to Be(OH)$_2$ gel, at high pressure. Consequently, the observed difference in the results between Hensley et al. [5] and this study is not unexpected.

The linear behavior of the data of Hensley et al. [5] may be explained by the smaller range of pressures spanned in their experiment. Alternatively, considering the uncertainties in measuring both the decay constant and pressure in the experiment of Hensley et al. [5], their data could also be fitted with a quadratic regression. It is well known that compressibility of solids decreases with increasing pressure. The decrease in the rate of increase of the decay constant of ^{7}Be at higher pressures observed in the present study is most likely a manifestation of the decrease in compressibility of Be(OH)$_2$ gel at higher pressures.

The conversion of ^{40}K to ^{40}Ar by electron capture has been widely adopted to date geological events (e.g., [9–11]). If the effect of pressure on the decay rate of ^{7}Be observed in the present study also occurs in ^{40}K, and K-containing minerals were subjected to high pressures during their geological history, the ages of these materials determined by the conventional dating method might be overestimated. However, since λ of ^{7}Be increases by about 1% at 400 kbar, it would be expected that a similar effect on larger nuclides such as ^{40}K would be smaller. We would like to note here that, following our experiment on ^{7}Be, another experiment was performed on ^{83}Rb, a much bigger nuclide undergoing electron-capture decay. For a nuclide of this size, no measurable changes were observed up to 420 kbar at room temperature.

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