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Pressure Dependence of the Radioactive Decay Constant of Beryllium-7

Abstract. *Diamond anvil presses of a new design were used to compress samples of beryllium-7 oxide to 120, 210, and 270 kilobars. The decay constant for the conversion of beryllium-7 to lithium-7 by electron capture was measured for compressed and uncompressed samples. A least-squares fit of the equation* $(\lambda_C - \lambda)/\lambda = K_P P$ *to the experimental data, where* λ_C *and* λ *are the decay constants of the compressed sample and an uncompressed sample, respectively, and* P *is pressure, yields a value of* $(2.2 \pm 0.1) \times 10^{-5}$ *kbar*$^{-1}$ *for the constant* K_P.

Twenty-six years have passed since the independent suggestions of Segrè (*1*) and Daudel (*2*) that the decay constants of certain radioactive nuclides might be altered by varying the electron density in the vicinity of the nucleus. Since that time, several groups have placed nuclides that decay by electron capture and internal conversion in different chemical and physical environments in order to observe these effects (*3*). The two nuclides most frequently used for these studies have been ^{7}Be and ^{99m}Tc.

The first attempts to observe the effect of high pressure on the half-life of a nuclide were carried out by comparing the measured activities before and after compression of a radioactive sample. Bainbridge (*4*) observed that the difference in the decay rate between ^{99m}Tc compressed to 100 kbar and ^{99m}Tc at 1 bar was about 0.02 percent. A few years later Gogarty *et al.* (*5*) used a similar technique to measure the effect of pressure on the decay rates of ^{7}Be and ^{131}Ba. The technique of measuring the effect while the sample remained at a high pressure was used first by Cooper (*6*) on the decay of ^{90m}Nb and again by Mazaki *et al.* (*7*), who repeated the ^{99m}Tc measurement.

The diamond anvil press (*8*) permits studies on small samples at high pressures without the use of large hydraulic devices. We report here results obtained with the diamond anvil press on the change in the decay constant of ^{7}Be in BeO at pressures up to 270 kbar. Beryllium-7 decays to ^{7}Li when a captured electron converts a proton to a neutron.

The values of $\Delta\lambda/\lambda$ measured at 120, 210, and 270 kbar are shown in Fig. 1; the line is a least-squares fit to the equation $(\lambda_C - \lambda)/\lambda = K_P P$, where λ_C and λ are the decay constants of the compressed and uncompressed samples, P is pressure, and K_P is the constant of proportionality. The value obtained for K_P is $(2.2 \pm 0.1) \times 10^{-5}$ kbar^{-1} when P is in units of kilobars. Using the data of Gogarty *et al.* (*5*) for the compression of $^{7}BeCO_3 \cdot Be(OH)_2$ we calculate a K_P value of $(2 \pm 3) \times 10^{-5}$ kbar^{-1}, in agreement with our value of K_P within experimental error. Diffraction patterns produced when an x-ray beam traverses the entire pressure range in a single sample of pure BeO provide evidence that no phase transition occurs. The maximum pressure in this sample calculated from the measured BeO lattice parameters is 300 or 240 kbar, based on the Birch equation with the elastic constants of Anderson *et al.* (*9*) or the shock compression data of Cline and Stephens (*10*), respectively.

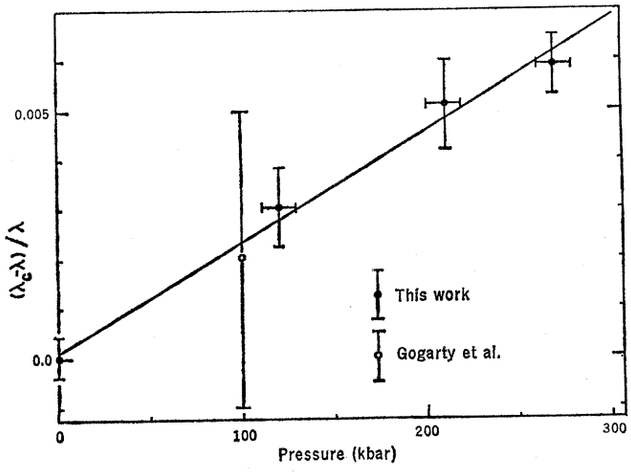

Fig. 1. Fractional increase in the total decay constant of ^{7}Be in ^{7}BeO as a function of pressure; the line is a least-squares fit of our data (see text). Error bars represent one standard deviation. The data point of Gogarty *et al.* (*5*) is calculated from a least-squares fit of 20 measurements near 100 kbar.

The press, which was modified from the one described by Bassett *et al.* (*8*), is a simple piston and screw device constructed from stainless steel. The body of the press is basically cylindrical, 2.5 cm in diameter and 5 cm long. Radiation emitted by the sample emerges through an opening which has a solid angle of approximately 2 steradians. The anvils are 25-mg (⅛ carat) brilliant-cut single crystal diamonds which have had their culet faces enlarged to 0.3 and 0.4 mm.

Samples of BeO were prepared in the conventional radiochemical manner (*11*) from the carrier-free isotope in 0.5*N* HCl (*12*). The pressure samples were made by first compressing a layer of NaCl between the diamond faces. Then the piston with the smaller anvil face was removed from the press and a piece of the ^{7}BeO in the shape of a thin platelet about 0.1 mm in diameter was transferred to the center of the large anvil. The piston was then reinserted into the press, and the press was loaded to its final pressure.

The purpose of including a layer of NaCl between the anvils of the press is twofold. First, it serves as a pressure-transmitting medium, that is, a substance which surrounds and provides a uniform pressure on the entire sample. This results in part from the high compressibility and low shear strength of the NaCl. Second, it serves as an internal pressure standard. Because of the design of the press, it is possible to use standard x-ray diffraction techniques to determine the lattice parameter of

NaCl. This value, in turn, can be used to calculate pressure.

A slightly modified Debye-Scherrer powder camera 114.59 mm in diameter was used for this work. The geometry of the press and camera is such that the x-rays pass through the sample perpendicular to the compression axis. As a result, the x-ray record contains information about the pressure over the entire sample area.

Figure 2 shows portions of two films that were exposed in the manner described above. The patterns, which are both from NaCl, were obtained from samples loaded to pressures of 120 and 270 kbar. The BeO is located in the central portion of the high-pressure region but has too low a scattering power to produce lines of its own. The strongest line in each pattern is from the 200 plane of the high-pressure portion of the NaCl. The line from the 200 plane at a pressure of 1 bar is inside the corresponding 200 line at high pressure and clearly separated from it. This is because the pressure gradient is concentrated near the edges of the anvils, where the sample is very thin, and as a result there is not enough NaCl between the two pressure extremes to provide the intensity necessary to produce a broad band. Since the BeO is located in the center of the sample area, it is subjected to a pressure that is the same as or greater than that of the NaCl immediately surrounding it. The spots on the patterns are Laue reflections from the single crystal diamond anvils.

To calculate the pressure we used the isothermal compression data for NaCl given by Weaver *et al.* (*13*). We measured the diffraction pattern and calculated the lattice parameter of the NaCl unit cell. From this we calculated V/V_0, the ratio of the compressed volume to the volume at 1 bar, and then determined the sample pressure from the Weaver equation (*13*). The overall uncertainty in the pressure is estimated to be approximately ± 10 kbar.

After preparation and preliminary pressure determination, the presses were fixed to special mounts for counting. In order to provide a set of reference standards for the system, samples of BeO at 1 bar were counted along with the compressed samples. In addition, two of the experiments included a pair of identical chemical samples which provided a null measurement to check the electronic and geometric stability of the system. In each of these cases the $\Delta\lambda/\lambda$ values were found to

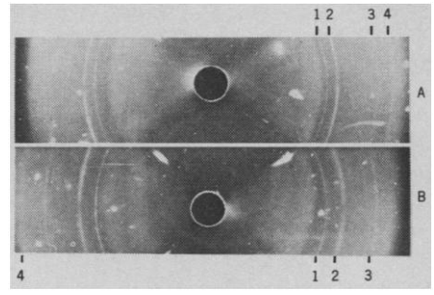

Fig. 2. X-ray diffraction photographs obtained from ^{7}BeO-NaCl samples compressed to (A) 120 kbar and (B) 270 kbar. Lines from the 200 and 220 planes of NaCl were produced by using filtered copper radiation. Lines 1 and 3 are reflections from the portion of the sample at 1 bar; lines 2 and 4 were produced from the high-pressure portion of the sample.

be less than $(4 \pm 4) \times 10^{-4}$. This value is considerably smaller than the value of $\Delta\lambda/\lambda$ measured for the compressed and uncompressed samples. The 477-kev gamma ray was used to monitor the decay of ^{7}Be. The samples were counted with Ge(Li) detectors (*14*), which were connected through a field-effect transistor preamplifier and a linear amplifier to a 4096-channel analyzer with computer-compatible output. On the average, activities were measured every other day for a period of three half-lives, about 6 months. Statistical fluctuations in each of the measured activities of the sample are of the order of ± 0.1 percent. After the raw data were acquired they were stored on magnetic tape, and the activities were calculated later.

The method of integrating over a constant energy region bracketing the photopeak was used to determine the activity of the counted sample. We chose the energy region between 455 and 485 kev to represent the total activity. After the activities were calculated, they were used as input to a standard least-squares code which fits an exponential decay curve to the data to determine the value of the decay constant for each sample. Values of $(\lambda_C - \lambda)/\lambda$ were calculated directly from the fitted decay constants. This method of analysis was chosen because it is insensitive to efficiency changes in the gamma-ray detection system.

The increase in the decay constant of ^{7}BeO with pressure has been estimated with the electron wave functions of the free Be atom (*15*). In our model calculation the fractional amount of electron density in the outermost portion of the Be atom (a radius of 0.89 Å was used) containing 10 percent of the volume is redistributed inside the new volume after compression ($0.9V_0$) with the original electron density distribution. This gives a 0.35 percent increase in the electron density at the nucleus for a free Be atom. We assume the same enhancement in λ of 0.0035 for BeO at a value of V/V_0 of 0.9, which corresponds to a pressure of 240 kbar when use is made of the equation of state of Cline and Stephens (*10*). The experimental values of $(\lambda_C - \lambda)/\lambda$ measured at 210 and 270 kbar are 0.0051 and 0.0059, respectively. Interpolating between these two points, we observe that the calculation outlined above gives an enhancement in λ which is two-thirds of the experimental value. However, this calculation is based on several assumptions and more rigorous theoretical calculations need to be made with more realistic models.

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