Environmental effects on nuclear decay rates*

ZHOU Shu-Hua(周书华)[1)]

China Institute of Atomic Energy, P. O. Box 275(18), Beijing 102413, China

Abstract: The possible change of nuclear decay rates in different environments has long been an interesting topic due to its importance not only in nuclear physics but also in astrophysics, geological dating, condensed matter physics, etc. The progress in the investigation of variations in nuclear decay rates are reviewed.

Key words: decay rate, environmental effect, radioactive nuclide

PACS: 21.10.Tg, 23.20.Lv, 23.40.-s **DOI:** 10.1088/1674-1137/35/5/008

1 Introduction

Although in textbooks and handbooks the decay rate of a radioactive nuclide is depicted by the decay constant, the possible decay rate perturbation by the change in atomic electrons due to the physical or the chemical state of the atom has been an interesting subject for more than one hundred years. A large volume of work in this subject have been published since the discovery of radioactivity. The reviews of earlier theoretical and experimental investigations have been presented by several authors [1–6]. In the last decade, some remarkable results in both theoretical and experimental investigations of the environmental effects on nuclear decay rates have appeared. It may be useful to have a review of recent progresses.

2 Early investigations

Soon after the discovery of radioactivity, starting from Bequerel's experements with uranium cooled to the boiling point of liquid air(−185 ℃) [7], a large number of studies were conducted, trying to interfere with the decay rate of a radioactive nuclide by different physical and chemical environments, such as temperature as low as −255 ℃ and as high as 1350 ℃, pressure from 300 atm to 2000 atm, a magnetic field of 4.5 T, strong electric fields, different altitudes, acceleration to 970000 g, radiation by X-, β-, γ-rays, etc. In one experiment, the radioactive substance was put in a steel encased Cordite bomb where a temperature of 2500 ℃ and a pressure of 1200 atm were expected to occur. Although more than 80 papers on the subject appeared in the first three decades of the last century, none of them could unambiguously show the variation in decay rates.

In 1947, Segrè [8] and Daudel [9] pointed out independently that the radiative decay constant by electron capture is proportional to $|\psi|^2$ of the electrons near the nucleus, and that in the case of light elements like ^{7}Be, it may be possible to change this quantity by an appreciable amount by putting the ^{7}Be into different chemical compounds, and then the half-life of ^{7}Be in different compounds would be altered accordingly. Daudel [9] mentioned also that the atomic electron density may affect the internal conversion and hence the decay rate of an isomeric state. Their work stimulated interest in this field. Experimental results from different laboratories provided supporting evidence for their predictions, especially Johlige et al. [10], using seven pairs of Be compounds, each with a different chemical composition. They found a large difference in the decay constants of ^{7}Be $\Delta\lambda/\lambda$(Be)=$(1.852\pm0.082)\times10^{-3}$ between $Be_4O(CH_3COO)_6$ and $(BeF_2)_{amorph}$. Hensley et al. [11] reported that when applying a high pressure of up to 270 kilobars to ^{7}BeO, the decay rate of ^{7}Be increased by 0.59%. The influence of the shift of electronic environments on the decay rate of an isomer was first examined by Bainbridge et al. [12] with ^{99m}Tc. Meanwhile Slater [13] provided a theoretical

Received 12 August 2010

∗ Supported by National Natural Science Foundation of China (10305020)

1) E-mail: shzhou@ciae.ac.cn

explanation and supporting evidence for the work of Bainbrige et al.

The rates of nuclear decay by α and β emissions might be influenced by the perturbation of the Coulomb barrier caused by the change in the electron density near the nuclei. Some diverged theoretical estimations of the rate change in α decay were published [14, 15]. The chemical effects on the β decay calculated by Alder et al. [16] produced the values lying in the range 10^{-4} to 10^{-5}. However, no concrete experimental results were available.

3 High precision measurements

In previous experimental studies, ionization chambers or NaI detectors were used in the ^{7}Be decay rate measurements. The reported errors of about 1%–0.1% resulting from the measurements with those detectors prohibited the precision of the subtle decay rate variation of ^{7}Be in different physical and chemical environments. Due to the importance of the decay rate of ^{7}Be in many areas of science, such as nuclear physics, astrophysics, geology, environment and condensed matter, and the availability of high purity germanium detectors, after about two decades of silence, experimental investigations on the variation of the decay rate of ^{7}Be in different environments revived.

Using an HPGe γ-ray spectrometer system, Huh et al. [17] measured with a high precision (of $\pm$0.01%) the half life of ^{7}Be in $Be^{2+}(OH_2)_4$, $Be(OH)_2$ and BeO to be $T_{1/2}$=53.69 d, 53.42 d and 54.23 d, respectively. So the observed variation of the decay rates of ^{7}Be is as much as 1.5%. The same researchers also reported increases at the 1% level in the ^{7}Be decay constant for $Be(OH)_2$ gel exposed to pressures of up to 400 kbar [18]. Tossell [19] calculated the electron densities at the Be nucleus in a number of oxidic compounds and found that the variations in the electron densities are only at the 0.1%–0.4% level. This result raised doubts about experimental results by Huh et al.. However, Tossell in the same work estimated the effects of Be-O bound compression in $Be(OH)_4^{2-}$ on the energy and the electron density at the nucleus, and indicated that several hundred kbar of pressure could increase the electron density by 1% or more, qualitatively consistent with the results obtained by Liu et al. with the $Be(OH)_2$ gel.

Considering that if ^{7}Be is implanted in a medium having high effective electron affinity (EA), the beryllium should lose a significant fraction of its 2 s electrons as a result of its interaction with nearby medium atoms, Ray et al. [20] measured the difference in the decay constant of ^{7}Be implanted in Au, which has a high electron affinity, and that in Al_2O_3, which has lower electron affinity, and found the former is smaller than the latter by (0.72$\pm$0.07)%. They also analyzed this result as well as others using the linear muffin-tin orbital (LMTO) method and Hartree's calculations to quantitatively explain the results. In the same paper, Ray et al. claimed that the results suggest the predicted ^{8}B solar neutrino flux should be 1.9% lower than the standard value.

Norman et al. [21] measured the decay rate of ^{7}Be implanted in host materials of graphite, boron nitride, tantalum, gold, lithium fluoride and aluminum and obtained the half-lives 53.107$\pm$0.022, 53.174$\pm$0.037, 53.195$\pm$0.052, 53.311$\pm$0.042, 53.12$\pm$0.07 and 53.17$\pm$0.02, respectively. The resulting decay rates in those host materials show changes of up to 0.38%, which is about half of that seen between Al_2O_3 and Au by Ray et al. Norman et al. also argued that the size of the possible correction for the decay rate of ^{7}Be due to the measurement in different host materials was considerably smaller than the experimental uncertainty in the rate for the competing ^{7}Be (p, γ) ^{8}B reaction, so the observed variations in the ^{7}Be decay rate should not warrant changing the predictions for the flux of ^{8}B solar neutrinos [21]. In response to Norman et al., Ray et al. [22] pointed out that the apparent disagreement between the two sets of experimental results could be due to the choice of different reference samples, and the irradiation damage to the lattice structure in the case of using heavy ion beams for ^{7}Be implantation might also be responsible.

To test the decay rate variation of ^{7}Be in host materials with different electron affinities, Liu Z et al. [24] implanted the ^{7}Be ejected from ^{7}Li(p,n)^{7}Be reactions in natural beryllium (EA=$-$0.19 eV) and gold (EA=2.308 eV) and found no difference in the decay rate of ^{7}Be in the two hosts with the experimental precision of 0.12%. To explain the result, the author suggested that the different lattice structures of Be and Au should be taken into account along with the electron affinity.

Souza et al. [23] measured the difference in the decay rate for ^{7}Be implanted in Li and Ta. The result $\Delta\lambda/\lambda$=(0.71$\pm$0.31)% was smaller than that expected from the comparison of the electron densities of tantalum and lithium (ρ_e(Ta) $\approx$30 ρ_e(Li)). They postulated that the small decay rate variation that might be due to the change in density of valence electrons at the nucleus could be balanced by the change in the exchange and overlap correction factors as Boruta and Makariunas [25] had calculated.

Nir-EI et al. [26] measured the decay rate of ^{7}Be implanted in four materials: copper, aluminum, sapphire and PVC. They found no host material dependence within 1σ.

There are two papers published concerning the ^{7}Be decay rate in C_{60}. Ohtsuki et al. [27] measured the ^{7}Be decay rates in C_{60} and in metal Be. They observed that the half-life obtained for ^{7}Be in C_{60}, $T_{1/2} = 52.68 \pm 0.05$ days, is 0.83% shorter than in Be metal. They attributed this result to the special environment in the C_{60} cage such as the many π electrons in C_{60} and the special dynamic conditions of the electrons inside the C_{60} cage. Ray et al. [28] measured the change in the ^{7}Be decay rate in exohedral and endohedral C_{60} fullerene compounds and found that the half-life of the endohedral ^{7}Be@C_{60} complex was shorter than that of the exohedral complex by 1%.

Suggesting that the enhanced electron screening, which had been observed in d(d, p)t reactions in metallic environments [29], may also affect the decay rates for radioactive nuclei, Zhou et al. [30] measured the decay rate variation of ^{7}Be implanted in Pd and Au host materials. Pd and Au have similar crystal structure but different electron affinity ($EA_{\rm Pd}$=0.56 eV, $EA_{\rm Au}$=2.31 eV) and different screening potentials ($U_{\rm e}$(Pd)=800±90 eV, $U_{\rm e}$(Au)=280±50 eV), and the extracted effective electron number surrounding each implanted deuteron in Pd is larger (6.3±1.3) than that for Au (0.9±0.3). Both the differences in electron affinity and the effective electron density favor a larger decay rate of ^{7}Be in Pd than in Au. In that experiment, 28.4 MeV radioactive ^{7}Be beams were used to implant the ^{7}Be ions into the Pd and Au hosts. The annealing process was performed to remove the damage in the lattice structure of the host materials. The 478 keV γ-rays resulting from the decay of ^{7}Be in Pd and Au were simultaneously measured with two sets of HPGe detectors. The measurements were conducted for 54 days, and then the positions of the two source samples were interchanged and the measurements were conducted again for 81 days. The observed decay rate of ^{7}Be in Pd was larger than that in Au by (0.8±0.2)%.

4 Test of the Debye model prediction

Using the plasma Debye model for α-decay, Raiola et al. [31] predicted that the half-life of the α-decay of ^{238}U in a cooled metal environment could be deduced by a factor of 3.2. Kettner et al. [32] predicted that for the α-decay and β^{+}-decay, one might observe a shorter half-life due to the acceleration mechanism of the Debye electrons for these positively charged particles similar to the protons, deuterons or ^{3}He in the fusion reactions, while for the β^{-}-decay and electron capture process, one could observe a longer half-life due to the deceleration for the negatively charged particles, and that if the α-decay ^{210}Po$\rightarrow \alpha+^{206}$Pb ($T_{1/2}$=138 days) happens in a metal cooled to T=4 K, the half-life would be shortened to 0.5 days; for ^{226}Ra $\rightarrow \alpha + {}^{222}$Rn ($T_{1/2}$=1600 years) would become $T_{1/2}$=1.3 years.

For the electron screen effect on α-decay, there are some other theoretical predictions. Zinner [33] using quantum mechanical tunneling arguments showed that although the electron screening effect causes the effective screened potential lower than the pure coulomb barrier and thus reduces the tunneling path, the screening effect also makes the α-decay energy lower and thus increases the tunneling path. So the screening effect on reducing the α-decay half-life is likely very small, if present at all. Liolios [34] predicted that ultrastrong magnetic fields and dense astrophysical plasmas can reduce the half-life of α-decaying nuclei by many orders of magnitude and in that kind of environment the conventional Geiger-Nuttall law is modified so that all relevant half-lives are shifted to dramatically lower values.

Encouraged by the possible benefit to the radioactive waste disposal management, several experiments on the measurements of half-lives of radioactive nuclei implanted in metals cooled to around 12 K were performed or planned to test the plasma Debye model predictions. Wang et al. [35] measured the half-life of ^{7}Be in metallic and non-metallic environments cooled to T =12 K. The observed half-life is increased by (0.9±0.2)% (^{7}Be in Pd) and (0.7±0.2)% (^{7}Be in In), while the half-life is unchanged for ^{7}Be in Li_2O. The trend of the results accords with the expectation from the Debye plasma model, but the value is much smaller than expected. The possible contribution from the Debye electrons themselves and the oxygen or hydrogen contamination in the metal hosts were discussed. In addition, the β^{-} decay half-life of ^{198}Au in Au cooled to T = 12 K was observed to be longer by (4.0±0.7)% than the literature value [36]. The β-decay half-life of ^{22}Na implanted in the palladium host cooled to 12 K was found to be shorter by (1.2±0.2)% [37] and when cooled to 15 K by (0.46±0.14)% [38], while implanted in aluminum cooled to 90 K was found to be shorter by (0.70±0.45)% [39].

Riola et al. [40] measured the α-decay half-life of ^{210}Po inside the metal Cu at room temperature and cooled to T=12 K. The result shows that the half-life for 12 K is shorter by (6.3±1.4)% than at room temperature.

5 Summary

The attempts to change the half-lives of radioactive nuclei can be classified into two categories: changing the electron density at the nucleus; and accelerating/decelerating the decay processes by the plasma electrons in a cooled metallic environment. The experimental results from the measurements of ^{7}Be in different chemical forms and implanted in host materials with different effective electron affinities have shown the variation in the decay rate around the 1% level. While the linear muffin-tin orbital method can be used for a rough estimate of the variation of the electron capture decay rate in different hosts, the plasma Debye model unreasonably over-estimates the variations of the α-decay rates by the electron screening effect in cooled metals.

References

1 Daudel R. J. Phys. Rad., 1952, **13**: 557
2 DeBenedetti F, Barros F Des, Hoy G R. Ann. Rev. Nucl. Sci., 1966, **16**: 31
3 Emery G T. An. Rev. Nucl. Sci., 1972, **22**: 165
4 Freedman M S. Ann. Rev. Nucl. Sci., 1974, **24**: 209
5 HaHn H P, Born H J, Kim J I. Radiochimica Acta, 1976, **23**: 23–37
6 Dostal K P, Nagel M, Pabst D. Z Naturforsch, 1977, **32a**: 345–361
7 Bequerel H. Compt. Rend. Acad. Sci. 1901, **133**: 199
8 Segrè. Z. Physik, 1947, **71**: 274
9 Daudel. Rev. Sci., 1947, **85**: 162
10 Johlige H W, Aumann D C, Bron H J. Phys. Rev. C, 1970, **2**: 1616
11 Hensley W K, Basset W A, Huizenga J R. Science, 1973, **181**: 1164
12 Bainbridge K T, Goldharber M, Wilson E. Phys. Rev., 1951, **84**: 1260
13 Slater J C. Phys. Rev., 1951, **84**: 1261
14 Alder K, Baur G, Raff U. Phys. Lett. A, 1971, **34**: 163
15 Rubinson Wand Perlman M L. Phys. Lett. B, 1972, **40**: 352
16 Alder K, Baur G, Raff U. Helv. Phys. Acta. 1971, **44**: 514
17 Chih-An Huh. Earth and Planary Science Letters, 1999, **171**: 325–328
18 LIU Lin-Gun, Chih-An Huh. Earth and Planary Science Letters, 2000, **180**: 163–167
19 Tossell J A. Earth and Planary Science Letters, 2002, **195**: 131–139
20 Ray A, Das P, Saha S K et al. Phys. Lett. B, 2007, **455**: 69–76
21 Norman E B, Rech G A, Browne E et al. Phys. Lett. B, 2001, **519**: 15–22
22 Ray A, Das P, Saha S K et al. Phys. Lett. B, 2002, **531**: 187–189
23 Souza D J et al. Jour. of Nucl. Sci. and Tech., 2002, **2**(Suppl): 470–473
24 LIU Z et al. Chin. Phys. Lett., 2003, **20**: 829–831
25 Boruta J, Makariunas K. Phys. Lett. A, 1979, **71**: 47
26 Nir-EI Y et al. Phys. Rev. C, 2007, **75**:012801
27 Ohtsuki et al. Phys. Rev. Lett., 2004, **93**: 112501
28 Ray A et al. Phys. Rev. C, 2006, **73**: 034323
29 Raiola et al. Phys. Lett. B, 2002, **547**: 193–199
30 ZHOU S et al. Chin. Phys. Lett., 2005, **22**: 565
31 Raiola et al. Eur. Phys. J. A, 2005, **31**: 1141
32 Kettner et al. J. Phys. G, 2006, **32**: 489–495
33 Zinner N T. Nucl. Phys. A, 2007, **781**: 81–87
34 Liolis T E. Phys. Rev. C, 2003, **68**: 015804
35 Wang B et al. Eur. Phys. J. A, 2006, **28**: 375–377
36 Spillance et al. Eur. Phys. J. A, 2007, **31**: 203–205
37 Limata B et al. Eur. Phys. J. A, 2006, **28**: 251–252
38 LIian G et al. Chin. Phys. Lett., 2008, **25**: 70–72
39 Ruprecht G et al. Proceedings of Science (International Symposium on Nuclear Astrophysics: Nuclei in the Cosmos IX) 171, CERN, 2006
40 Riola F. Eur. Phys. J. A, 2007, **32**: 51–53