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# Ab-initio study of the effects of pressure and chemistry on the electron-capture radioactive decay constants of $^{7}$Be, $^{22}$Na and $^{40}$K

Kanani K.M. Lee [a,b,*], Gerd Steinle-Neumann [b]

[a] Department of Physics, New Mexico State University, MSC 3D Box 30001, Las Cruces, NM 88003-8001, USA
[b] Bayerisches Geoinstitut, Universität Bayreuth, D-95440 Bayreuth, Germany

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**Abstract**

Using state-of-the-art ab-initio computations, we find that pressure and chemical environment have a small effect on the electron-capture component of the decay constants of isotopes of $^{7}$Be, $^{22}$Na and $^{40}$K. As expected, $^{7}$Be shows the greatest amount of change with ~0.1–0.2% increase in the decay constant at 25 GPa as a metal, chloride or oxide. In their metallic forms, $^{22}$Na and $^{40}$K show an increase of 0.06% and 0.025% change, respectively, at 25 GPa. Upon further compression, potassium and sodium undergo an s-to-d electronic transition of their valence electron, causing a significant change in the charge density at the nucleus. Our results suggest that the effect of pressure and chemistry on $^{40}$K, combined with the opposing effects of high temperatures, will have little, discernible effect on the heat production in the deep Earth as our predicted changes are smaller than the uncertainties in the total decay constant for $^{40}$K.


*Keywords:* electron-capture radioactivity; beta decay; beryllium; sodium; potassium; high pressure

## 1. Introduction

Radioactive decay plays a central role in planetary science as appropriate decay schemes are used to date geological and astronomical processes at all time and length scales (Dickin, 1995), and radioactivity provides an important source of heat in planetary bodies, both in their early history during accretion and differentiation (Dickin, 1995; Urey, 1955, 1956; Urey and Donn, 1956) and also over geological times (Fowler, 2005). In the Earth, approximately 21 TW (Fowler, 2005) of heat is currently thought to be produced by the decay of four isotopes, $^{40}$K, $^{232}$Th, $^{235}$U and $^{238}$U, almost half of the ~44 TW heat measured at the surface (Pollack et al., 1993). Roughly 20% of that heating is estimated to be generated in the crust ($<10^{-7}$ of the Earth's volume), and as much as 8 TW could be produced by $^{40}$K decay in the core alone (Lee and Jeanloz, 2003; Lee et al., 2004). Despite the small natural abundance of $^{40}$K (~0.01% of total potassium, averaged over the entire Earth: ~100–1000 ppm K (Verhoogen, 1980)) and the other isotopes (~80–100 ppb Th and ~15–25 ppb U averaged over the entire Earth (Fowler, 2005)), they are important in determining the thermal and tectonic evolution of the Earth.

There are two primary decay types: α and β. Many naturally occurring heavy nuclei, including $^{232}$Th, $^{235}$U and $^{238}$U, decay by emission of α-particles (Dickin, 1995). In contrast, β decay isotopes correct a single proton ($p$) or neutron ($n$) excess by directly converting a proton into a neutron or a neutron into a proton,

$$\begin{array}{ll} n \rightarrow p + e^- & \beta^- \text{ decay} \\ p \rightarrow n + e^+ & \beta^+ \text{ decay} \\ p + e^- \rightarrow n & \text{electron capture} \end{array} \quad (1)$$

In the positive and negative β decays, an electron ($e^-$) or positron ($e^+$) respectively is produced inside the nucleus and

* Corresponding author. Department of Physics, New Mexico State University, MSC 3D Box 30001, Las Cruces, NM 88003-8001, USA.
*E-mail address:* kanani@physics.nmsu.edu (K.K.M. Lee).

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Table 1
Electron-capture parent isotopes and daughter isotopes, total half-life values, electron-capture half-life, percent of decay scheme that are electron capture, characteristic gamma ray energies emitted with each decay. Values are from Krane (1988), Dickin (1995) and from additional sources as noted in the table

| Parent | Daughter | $\tau_{1/2}$ | $\tau_{1/2,ec}$ | Percent EC decay | γ-ray energy (MeV) | Additional references |
|---|---|---|---|---|---|---|
| $^{7}$Be | $^{7}$Li | 53.3 days | 53.3 days | 100% | 0.478 | Norman et al. (2001) |
| $^{22}$Na | $^{22}$Ne | 2.6 yr | 28 yr | ~9.4%, rest $\beta^+$ | 1.275 | Firestone et al. (1978) |
| $^{40}$K | $^{40}$Ar | 1.25 Gyr | 10.4 Gyr | ~12%, rest $\beta^-$, $\beta^+$ | 1.461 | (Norman et al., 2001; Steiger and Jager, 1977) |

immediately ejected. Another particle, either a neutrino or anti-neutrino, is also emitted during β decays. The electron-capture decay process occurs when an orbital electron is captured by a proton in the nucleus to form a neutron, neutrino and often a γ-ray (Krane, 1988). Table 1 shows the relative amount of electron-capture decay and respective total half-life $\tau_{1/2,ec}$ and $\tau_{1/2}$ for investigated isotopes $^{7}$Be, $^{22}$Na and $^{40}$K.

Since radioactive decay is a nuclear process, it is often considered to be insensitive to external factors, such as ionization, temperature, pressure, or chemical environment. This is due to screening provided by the orbital electrons surrounding the nucleus and has been shown to be essentially true for α- and positive and negative β decays (see references within Hahn et al., 1976). However, for electron-capture processes, external forces can influence the stability of an unstable nuclide because the decay rate is directly related to total electron charge density $\rho_e$ at the nucleus (Daudel, 1947; Segre, 1947; Emery, 1972). $\rho_e$ is affected by the shape of the orbital: only s atomic orbitals, or s-like states in condensed phases, have finite electronic densities at the nucleus. Therefore the electron-capture decay constant, $\lambda_{ec}\left(=\frac{\ln 2}{\tau_{1/2,ec}}\right)$, can be influenced by external factors that affect the s-like components of the electronic wave functions such as ionization (Bosch, 1999), temperature (Emery, 1972; Hahn et al., 1976), pressure (Gogarty et al., 1963; Mukoyama and Shimizu, 1974; Bukowinski, 1979a; Liu and Huh, 2000; Tossell, 2002), physical state (Norman et al., 2001) and chemical state (Daudel, 1947; Huh, 1999; Tossell, 2002). Of these, only the effects of chemical and physical (e.g., solid versus aqueous solution) state and pressure have been explored in detail for $^{7}$Be, with a few studies on other very short-lived electron-capture isotopes: $^{64}$Cu, $^{97}$Ru, $^{85}$Sr and $^{89}$Zr (see references in Hahn et al., 1976).

The change in electron-capture decay constant, $\delta\lambda_{ec}$ is given by (Bukowinski, 1979a; Tossell, 2002):

$$\delta\lambda_{ec} = \left[\frac{\rho_e}{\rho_{e,ref}} - 1\right]\lambda_{ec,ref} \quad (2)$$

where $\lambda_{ec,ref}$ is the reference decay constant for electron-capture decays and $\rho_{e,ref}$ is the reference electronic charge density, both at 0 GPa (Table 2).

Here we investigate the effects of pressure and chemistry on the electron-capture portion of the decay constants of $^{7}$Be, $^{22}$Na and $^{40}$K (Table 1) by means of ab-initio computations of the electron charge density $\rho_e$ at the nucleus. We add new estimates of $\delta\lambda_{ec}$ for $^{7}$Be, $^{22}$Na and $^{40}$K as metals, oxides and chlorides and compare to existing estimates of $\delta\lambda_{ec}$ due to pressure for metallic $^{40}$K (Bukowinski, 1979a) and crystalline $^{7}$BeO (Hensley et al., 1973). We also compare effects due to the chemical environment on $^{7}$Be compounds.

## 2. Previous work

### 2.1. $^{7}$Be

$^{7}$Be is naturally produced by cosmic ray bombardment of atmospheric nitrogen and oxygen and decays completely by electron capture to $^{7}$Li (Table 1). Once formed, $^{7}$Be is removed from the atmosphere and is incorporated into sedimentary material and water. The short half-life, rapid removal from the atmosphere and affinity for sediments make this nuclide useful in determining recent sediment formation. The external environment is expected to have the largest effect on the decay constant of $^{7}$Be due to its simple electronic structure that offers little electronic shielding of the nucleus. Experiments indicate

Table 2
The reference total electron charge density $\rho_{e,ref}$ for metal and simple compound forms of Be, Na and K at zero pressure for this study unless otherwise indicated. The structures used are those stable at zero pressure. $R_{MT}$ values (in Bohr) are kept constant through compression and are 1.23 for beryllium, 1.39 for sodium and 1.96 for potassium compounds. Uncertainties, where available, are given in parentheses

| Isotope | Compound | Structure | $\rho_{e,ref}$ (electrons/Bohr$^3$) | Predicted difference in $\lambda_{ec}$ at 0 GPa from metal form (%) |
|---|---|---|---|---|
| $^{7}$Be | Be | hcp | 35.3901 | |
| | $BeCl_2$ | orth | 35.3672 | −0.065 |
| | BeO | hcp | 35.4017 | +0.033 |
| | BeO [a] | | | +0.091 |
| | BeO [b] | | | −0.991 |
| | BeO [c] | | | −0.037 (0.008) |
| | BeO [d] | | | −0.030 (0.018) |
| $^{22}$Na | Na | bcc | 888.388 | |
| | NaCl | fcc | 887.970 | −0.047 |
| | $Na_2O$ | fcc | 888.180 | −0.023 |
| $^{40}$K | K | bcc | 5393.79 | |
| | KCl | fcc | 5393.14 | −0.012 |
| | $K_2O$ | fcc | 5393.45 | −0.006 |
| | $Fe_{31}K$ [e] | hcp | | +0.029 |

[a] For a model of BeO and with respect to the $\rho_{e,ref}$ for $Be(OH_2)_4^{2+}$ (Tossell, 2002).
[b] Experimental value for difference between BeO and $Be(OH_2)_4^{2+}$ (Huh, 1999).
[c] Experimental value for difference between BeO and $Be(OH_2)_4^{2+}$ (Johlige et al., 1970).
[d] Experimental value for difference between BeO and Be (Segre and Wiegand, 1949).
[e] These computations used a different $R_0$ than the other computations, thus the absolute values of $\rho_{e,ref}$ cannot be directly compared with that listed for K in the bcc structure. Instead, we compare $\rho_{e,ref}$ (bcc K: 5220.71 $e$/Bohr$^3$, $Fe_{31}K$: 5222.22 $e$/Bohr$^3$) for the same value of $R_0$.