J Radioanal Nucl Chem (2015) 303:1581–1583
DOI 10.1007/s10967-014-3570-7

Comparison of the decay constants of ^{51}Cr with metal, oxide, and chromate chemical states

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Received: 31 August 2014 / Published online: 9 September 2014

Abstract Chromium-51 decays through electron capture, the probability of which is perturbed by its electronic state. We have precisely measured the decay constants (λ) of ^{51}Cr with metal (Cr^0), oxide (Cr^{3+}), and chromate (Cr^{6+}) to investigate the effects of chemical states on the decay constants of ^{51}Cr. The value of $\left\{\lambda\left(Cr^{6+}\right)-\lambda\left(Cr^0\right)\right\}/\lambda\left(Cr^0\right)$ was determined to be $(6.4 \pm 3.7) \times 10^{-4}$, whereas the difference less than 1.4×10^{-4} was observed for $\left\{\lambda\left(Cr^{3+}\right)-\lambda\left(Cr^0\right)\right\}/\lambda\left(Cr^0\right)$.

Keywords Half-life · Decay constant · Chemical effect · Radionuclide Cr-51

Introduction

The decay constants of more than ten nuclides from ^{7}Be to ^{235}U^m have been found to be changed with changing environmental factors such as its chemical state [1]. Kakiuchi and Mukoyama [2] reported the change in the decay constant of the electron capture decay nuclide ^{51}Cr between the two chemical forms $CrCl_3$ (valence state +3) and Na_2CrO_4 (valence state +6). They also estimated the value for the relative change in the decay constant among Cr^0, Cr^{3+}, and Cr^{6+} valence states with a simple theoretical model. The estimation shows that the relative change in the decay constant between Cr^0 and Cr^{3+} state is approximately equal to that between the Cr^{3+} and Cr^{6+} state. However, there is no experimental value of the decay constant of Cr^0 state so far. In this study, the decay constants of ^{51}Cr have been precisely measured with Cr^0, Cr^{3+}, and Cr^{6+} valence states to investigate the effects of chemical states on the decay constants of ^{51}Cr.

Experimental

The decay constants of ^{51}Cr were measured for three chemical forms: chromium metal (Cr^0), chromium (III) oxide Cr_2O_3 (Cr^{3+}), and potassium chromate K_2CrO_4 (Cr^{6+}). The isotope ^{51}Cr was produced in the $^{nat}Cr(\gamma, xn)^{51}Cr$ reaction.

Target materials were about 100 mg each of Cr metal, Cr_2O_3, and potassium dichromate $K_2Cr_2O_7$. Each target material was sealed in a quartz tube and irradiated with bremsstrahlung photons. The irradiation was carried out with the electron linear accelerator at Tohoku University. The accelerator was operated at an electron energy of 30 MeV with a mean current of around 0.12 mA during the 8 h irradiation.

After the irradiation, the metal and Cr_2O_3 targets were maintained at 800 °C for 5 h in argon and atmosphere, respectively, with an electric furnace. The $K_2Cr_2O_7$ target was mixed into 250 mg of a non-radioactive $K_2Cr_2O_7$ reagent and then dissolved in 3 mL of distilled water. The solution was heated on a hot-plate and alkalified with potassium carbonate to produce CrO_4^{2-}. The solution was

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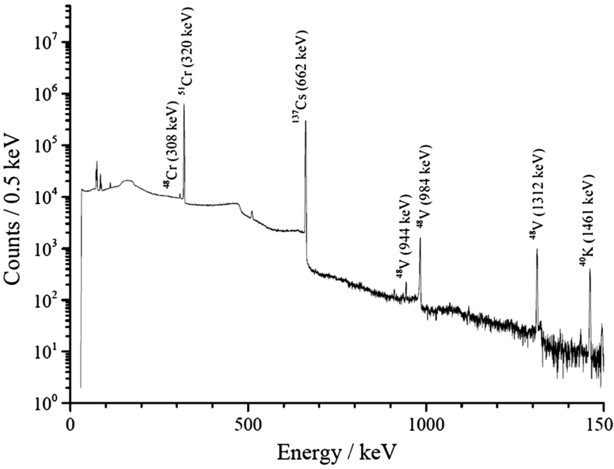

Fig. 1 A typical γ-ray spectrum for a Cr metal sample measured for 7,150 s

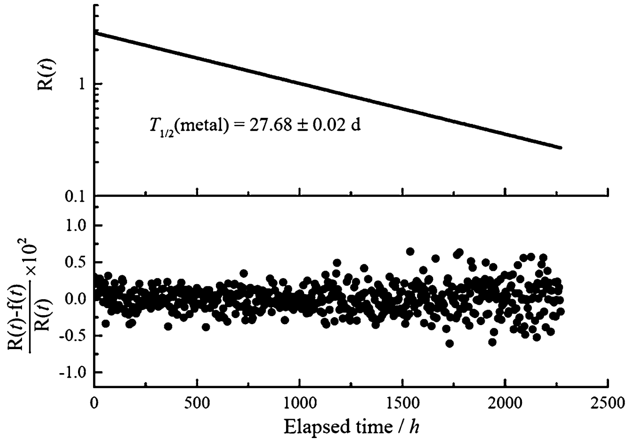

Fig. 2 An example of the decay curve for the 320 keV γ line of ^{51}Cr (*upper panel*) and the normalized residuals (*lower panel*)

filtered and the filtrate was evaporated to less than 1 mL on a hot-plate. Finally, a K_2CrO_4 sample was prepared by recrystallization from the solution. The metal, Cr_2O_3, and K_2CrO_4 samples were placed in aluminum cups separately and sealed with an epoxy resin adhesive.

These samples were measured in pairs of Cr_2O_3-metal and K_2CrO_4-metal to reduce the influence caused by the difference of detectors. The sample pairs were set in automated sample changers [3] and alternately placed in front of a high-purity Ge (HP-Ge) detector at intervals of 7,200 s. The procedures were repeated over at least 95 days, which is longer than 3.4 times of the half-life of ^{51}Cr (27.702 days [4]). A ^{137}Cs source was positioned close to the HP-Ge detector as a reference source to correct for influential factors for determination of the half-life such as pile-up effects [5]. The dead-time of the measurement system was from 4 to 8 % at the beginning of the measurement. The internal clock time of the computer for data acquisition was constantly calibrated by a time-standard signal distributed via a long-wave radio transmission station in Japan.

The experiments for each of the sample pair were separately conducted two times.

Results and discussion

The decay constant of ^{51}Cr was determined based on a reference method using a ^{137}Cs source. The ratio $R(t)$ is given by the following equation:

$$R(t) = \frac{C_{\mathrm{sample}}(t)}{C_{\mathrm{ref}}(t)},$$

$$C_{\mathrm{sample(ref)}}(t) = \frac{\lambda_{\mathrm{sample(ref)}} N}{\left(1 - e^{-\lambda_{\mathrm{sample(ref)}} t_{\mathrm{R}}}\right)} \frac{t_{\mathrm{R}}}{t_{\mathrm{L}}},$$

where $C_{\mathrm{sample(ref)}}(t)$ and $\lambda_{sample(ref)}$ are count rates of a sample (reference source) at the beginning of each data acquisition and decay constant, respectively. N is the net counts in the objective peak. t_{R} and t_{L} are real time and live time, respectively. The decay constant t_{sample} is described in the following equation:

$$\lambda_{\mathrm{sample}} = \lambda_{\mathrm{ref}} - a_{\mathrm{slope}},$$

where a_{slope} is the slope of the graph of ln $R(t)$ against time.

A typical γ-ray spectrum for a Cr-metal sample measured for the first 7,150 s is shown in the Fig. 1. The ^{51}Cr γ peak at $E\gamma = 320.1$ keV and the ^{137}Cs γ peak at $E\gamma = 661.7$ keV can be observed as two prominent peaks. Other peaks in the spectrum are ascribed to ^{48}Cr, which is produced in the ^{50}Cr(γ, 2n) reaction, ^{48}V, in the ^{50}Cr(γ, pn) reaction, and natural background radiations.

A typical decay curve for the $R(t)$ obtained using a least-squares fitting procedure and their residuals are shown in the upper and lower panel of Fig. 2, respectively. The residuals of the fit of all the data are within the limit of approximately 0.6 %. The half-life of ^{51}Cr with the metal form is determined to be 27.68 ± 0.02 days by the weighted average of four samples, which is in good agreement with literature value 27.702 ± 0.004 days [4].

Relative differences in the decay constant of ^{51}Cr obtained from this work and the previous research [2] are shown in the Fig. 3. The relative difference $\left\{\lambda\left(\mathrm{Cr}^{6+}\right) - \lambda\left(\mathrm{Cr}^{0}\right)\right\}/\lambda\left(\mathrm{Cr}^{0}\right)$ was determined to be $(6.4 \pm 3.7) \times 10^{-4}$. On the other hand, the difference less than 1.4×10^{-4} at a 68 % confidence level was observed for $\left\{\lambda\left(\mathrm{Cr}^{3+}\right) - \lambda\left(\mathrm{Cr}^{0}\right)\right\}/\lambda\left(\mathrm{Cr}^{0}\right)$. Kakiuchi and Mukoyama [2] reported the value of $(5.3 \pm 2.1) \times 10^{-4}$ for $\left\{\lambda\left(\mathrm{Cr}^{6+}\right) - \lambda\left(\mathrm{Cr}^{0}\right)\right\}/\lambda\left(\mathrm{Cr}^{0}\right)$, which is in good agreement with the our value of $\left\{\lambda\left(\mathrm{Cr}^{6+}\right) - \lambda\left(\mathrm{Cr}^{0}\right)\right\}/\lambda\left(\mathrm{Cr}^{0}\right)$. It follows from the results that the magnitude of the correlation of the decay constants of ^{51}Cr with Cr metal, Cr_2O_3,

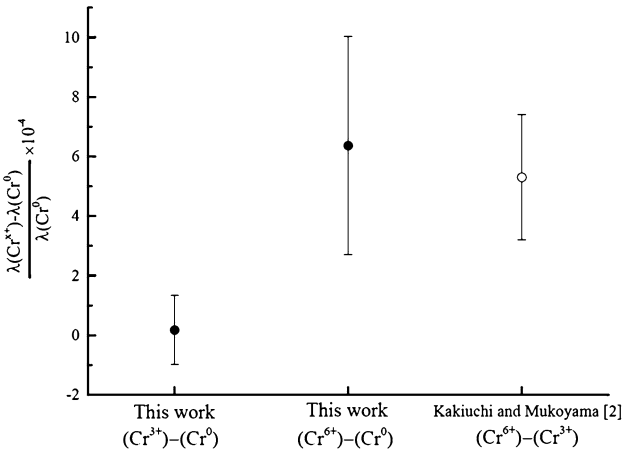

Fig. 3 Relative differences in the decay constant of ^{51}Cr between Cr metal, Cr_2O_3, and K_2CrO_4 chemical forms, respectively

and K_2CrO_4 chemical forms, respectively, are determined to be $\lambda(Cr^0) \approx \lambda(Cr^{3+}) < \lambda(Cr^{6+})$. In Ref. [2], the relative difference $\{\lambda(Cr^{6+}) - \lambda(Cr^0)\}/\lambda(Cr^0)$ and $\{\lambda(Cr^{3+}) - \lambda(Cr^0)\}/\lambda(Cr^0)$ were estimated to be 2.63×10^{-3} and $(1.22–1.65) \times 10^{-3}$, respectively, based on a Hartree–Fock-Slater approximation. There is a discrepancy between the simple theoretical estimation and our results not only of quantitative degree but also the qualitative tendency.

In conclusion, we measured the decay constants of ^{51}Cr with the Cr metal, Cr_2O_3, and K_2CrO_4 chemical form. The difference less than 1.4×10^{-4} at a 68 % confidence level was observed for $\{\lambda(Cr^{3+}) - \lambda(Cr^0)\}/\lambda(Cr^0)$, which disagrees with the theoretical estimation in Ref. [2]. For further discussion on the relation between the decay constant and the electron state, higher-accuracy experimental data and more realistic model are required.

Acknowledgments The authors would like to thank the technical staff of Research Center for Electron Photon Science, Tohoku University for their excellent operation of the accelerators. This work was supported by JSPS KAKENHI Grant Number 24740136.

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