PHYSICAL REVIEW C VOLUME 37, NUMBER 5 MAY 1988

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Depopulation of the isomeric state $^{180}Ta^m$ by the reaction $^{180}Ta^m(\gamma,\gamma')\,^{180}Ta$

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(Received 27 October 1987)

The irradiation of an enriched sample of $^{180}Ta^m$ with bremsstrahlung from a linear accelerator having an end-point energy of 6 MeV has excited a strong channel for the reaction $^{180}Ta^m(\gamma,\gamma')\,^{180}Ta$, which requires a total spin change of $8\hbar$. An integrated cross section of 4.8×10^{-25} cm^{2}keV has been found. This is a large value exceeding by two orders of magnitude known cross sections for (γ,γ') reactions producing isomers of other species.

The isotope $^{180}Ta^m$ carries a dual distinction. It is the rarest stable isotope occurring in nature[1] and it is the only naturally occurring isomer.[2] The actual ground state of ^{180}Ta is 1^+ with a half-life of 8.1 h, while the tantalum nucleus of mass 180 occurring with 0.012% abundance is the 9^- isomer $^{180}Ta^m$. It has an adopted excitation energy of 75.3 keV and half-life in excess of 1.2×10^{15} yr.[2]

The stellar s process[3,4] for nucleosynthesis has steadily gained favor for the production of $^{180}Ta^m$ and the role of the most critical intermediary $^{180}Hf^m$ has been well established.[2,5] However, the viability of this cosmic mechanism rests upon the absence of any reactive channel $^{180}Ta^m(\gamma,\gamma')\,^{180}Ta$ which could destroy the isomeric population in the photon bath present in the stellar interior at the time of creation. Prior experiments[6,7] have failed to show such a channel having any gateway for excitation below 1332 keV, but the rarity of the target material limited the sensitivity of those measurements. Reported here is the measurement of a very large cross section for the photonuclear deexcitation of $^{180}Ta^m$ through a gateway level at an energy $E \gtrsim 1.4$ MeV. This definitive observation of such a strong radiative coupling between isomeric and ground states of ^{180}Ta may affect explanations for the natural occurrence of $^{180}Ta^m$.

The energy-level diagram of ^{180}Ta and its daughters is shown in Fig. 1, together with a schematic representation of the individual steps in the excitation and detection of the $^{180}Ta^m(\gamma,\gamma')\,^{180}Ta$ reaction. As can be seen in Fig. 1, the principal means for the detection of the ^{180}Ta ground state lies in observing the $K\alpha$ lines of its daughter ^{180}Hf, following the decay by electron capture of the parent ^{180}Ta. The efficiency for the emission of $K\alpha$ photons relative to the number of ^{180}Ta decays is about 57%.[8]

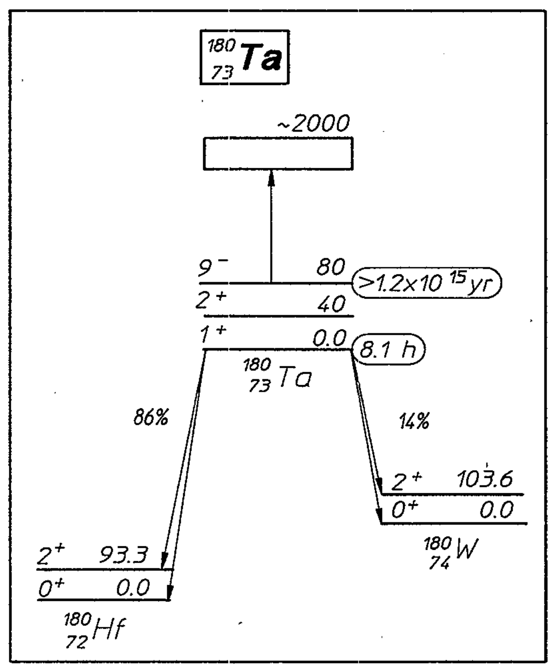

FIG. 1. Schematic energy-level diagram of ^{180}Ta and its daughters. Half-lives are shown in ovals to the right of the ground and isomeric levels. Energies are in keV. The initial transition of the (γ,γ') reaction is shown by the arrow pointing upward to the broad state represented by the rectangle. Cascade through the levels of ^{180}Ta is not known, but leads finally to the ground state. Electron capture to the left and beta decay to the right are indicated by the diagonal downward arrows. The final debris from pumping down the isomer is found principally in the $K\alpha$ fluorescence from the ^{180}Hf characterized by the 8.1 h lifetime of its ^{180}Ta parent.

Two targets were used in these experiments. One consisted of a disk 5 cm in diameter of tantalum in natural isotopic abundance. It contained about 0.5 mg of $^{180}Ta^m$ in the surface layer of thickness equal to the mean distance for escape of a 55 keV x-ray photon. The second target was enriched to contain 1.3 mg of $^{180}Ta^m$ in 24.7 mg of ^{181}Ta. Deposited as a dusting of oxide near the center of the surface of a 5 cm disk of Al and overcoated with a 0.25 mm layer of Kapton, this second sample was believed[9] to be free from self-absorption of the x-rays from the daughter Hf.

The samples were exposed to bremsstrahlung radiation from a Varian Clinac 1800 linear accelerator (LINAC) operated with an end-point energy of 6 MeV. This device has been well characterized,[10,11] and its output dose rate has been calibrated with an accuracy of $\pm 3\%$. After irradiation, the samples were counted with an N-type, HPGe spectrometer having a beryllium entrance window. Conventional techniques were used to calibrate the counting system with isotopic standards.

Figure 2 shows the spectra of the enriched target before and after 4 h irradiation. The spectrum from the other target was entirely similar with the Hf signal reduced by the ratio of the masses of the $^{180}Ta^m$ and the background increased by the appearance of K lines of Ta excited in the large mass of diluent ^{181}Ta by the decay of natural activity in the counting shield.

Figure 3 shows the dependence upon time of the counting rate observed in the Hf($K\alpha$) peaks after irradiation. Data points are plotted at the particular times at which the instantaneous counting rate equals the average counting rate measured over the finite time interval shown. The figure shows the close agreement of the measured rates to the decay expected for a half-life assumed to be 8.1 h.

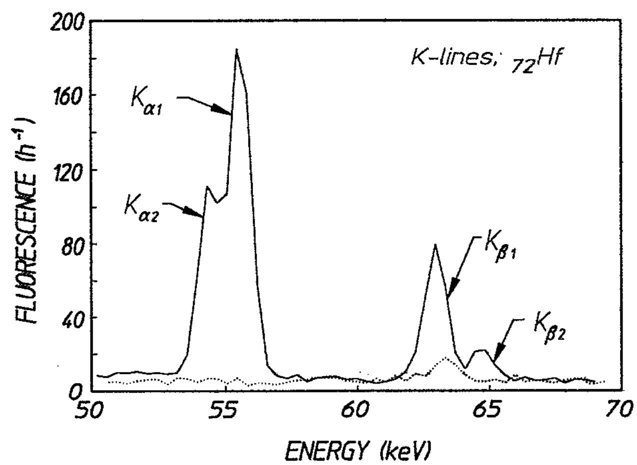

FIG. 2. Dotted and solid curves show, respectively, the spectra obtained before and after dumping some of the isomeric $^{180}Ta^m$ contained in a target sample enriched to 5%. An HPGe detector was used to obtain the dotted spectrum before irradiation. The feature at 63 keV is from traces of natural activity in the counting shield. The solid curve shows activity resulting from the transmutation of the pumped ^{180}Ta measured in the same sample and counting system after irradiation. The prominent additions are the $K\alpha$ and $K\beta$ hafnium x-ray lines resulting from electron capture in the ^{180}Ta.

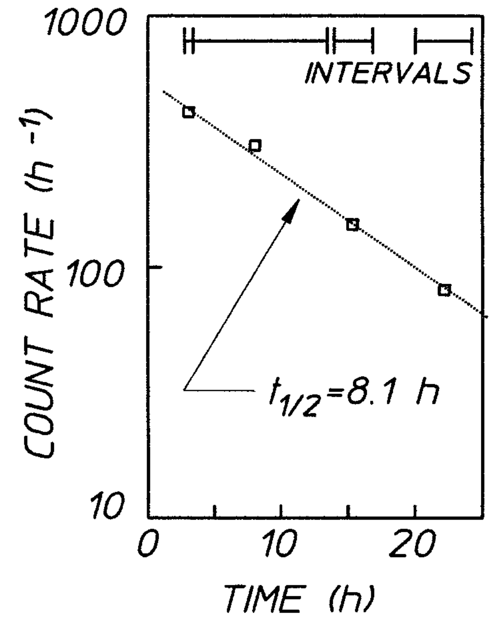

FIG. 3. Plot of the counting rates measured for the Hf ($K\alpha$) fluorescence from the target fabricated from natural tantalum as functions of the time elapsed from the end of the irradiation. The vertical dimensions of the data points are consistent with 1σ deviations of the measured number of counts accumulated during the finite counting intervals shown at the top of the graph. The dotted line shows the rate expected for a half-life of 8.1 h.

The spectrum of the bremsstrahlung pumping the fluorescence seen in Fig. 2 was taken from the literature[10] and was normalized to the total dose measured in this experiment. In this way, the time-integrated spectral intensity producing the fluorescence was found to be constant[12] to within a factor of 2 over the range 1–5 MeV at a value of 2×10^{14} keV/(keV cm^2). The number of counts observed in the Hf $K\alpha$ lines were corrected for finite irradiation and counting times, the absolute counting efficiency of the spectrometer, and the 57% emission intensity from the parent ^{180}Ta to obtain the total number of nuclei pumped to the ground state. Assuming self-absorption in the enriched target to be negligible, the integrated cross section for the deexcitation of the isomer can be readily calculated if the reaction is assumed to occur through a gateway state narrow in comparison to the range of energies spanned by the irradiation. A value of $\sigma\Gamma = 4.8 \times 10^{-25}$ cm^{2}keV is obtained for the integrated cross section if the gateway energy is arbitrarily assumed to be near the lowest value consistent with prior[7] negative results, 2.0 MeV. Even larger cross sections would result from the assumption that the gateway lies at higher energies where the pumping flux is decreased or from the inclusion of an exact self-absorption correction. Once the gateway energy is fixed, experimental error in the integrated cross section is bounded on the lower side by a total uncertainty of 15% contributed by the calibrations of source and detector and on the upper side by a factor of 2 arising from the possible loss of signal because of self-absorption of the Hf x-rays.

The results of this work show a radiative connection between the isomer ^{180}Ta^{m} and the ^{180}Ta ground state of remarkable strength. Comparative values for the deexcitation of other isomers are not available as it appears this is the first such measurement. However, the inverse process for the excitation of isomers by (γ,γ') reactions typically proceeds[13-15] with integrated cross sections at least two orders of magnitude smaller. The value reported here for the reaction ^{180}Ta$^{m}(\gamma,\gamma')^{180}$Ta is inexplicably large and may have several consequences. If the gateway level through which it proceeds is not sufficiently above the thermal energies expected to characterize the *s* process of nucleosynthesis, current models of the stellar production of ^{180}Ta^{m} will be severely affected.

The authors gratefully acknowledge the support of this work by the Innovative Science and Technology Directorate of the Strategic Defense Initiative Organization through direction by Naval Research Laboratory. Sincere appreciation is conveyed to Dr. P. P. Antich and E. C. Scarbrough of the University of Texas Southwestern Medical Center at Dallas for use of the linear accelerator and to the Oak Ridge National Laboratory isotope facility for the loan of the enriched ^{180}Ta^{m} sample.

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