RADIOCHEMISTRY AND RADIOPHARMACEUTICALS

Tantalum-178—A Short-Lived Nuclide for Nuclear Medicine: Production of the Parent W-178

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The physical decay characteristics of the short-lived radionuclide Ta-178 (half-life 9.3 min) appear to be suitable for use in conjunction with low-energy detection systems such as the multiwire proportional camera. This camera is inefficient for emissions with energies greater than 100 keV. The gamma-ray spectrum of Ta-178 is dominated by the characteristic hafnium x-rays (55–65 keV), emitted as a result of electron-capture decay. The parent nuclide, W-178 (half-life 21.7 d), was produced in the Michigan State University cyclotron by proton bombardment of stacked natural tantalum-foil targets. Optimum production was found to occur with an incident proton energy of 34 MeV at an effective activity of 1.1 mCi/μA-hr per MeV of target thickness. Tungsten-178 was chemically separated from the Ta foils with a yield of 98%.

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The high-pressure multiwire proportional camera (MWPC) has recently attracted attention as a possible alternative to the gamma camera because it provides high spatial resolution at count rates more than twice those of present detectors (*1*). This system is designed for use with low-energy gamma emitters: for example, the efficiency is typically 25% at 140 keV, 70% at 80 keV and 80% at 60 keV (*2*). This paper reports on a short-lived radionuclide, tantalum-178 (Ta-178), which may be suitable for use with such a camera.

Tungsten-178 ($T_{1/2}$ = 21.7 d) decays entirely by electron capture to the 9.3-min Ta-178 daughter (Fig. 1) without feeding the high-spin Ta-178 isomer ($T_{1/2}$ = 2.2 hr). The short-lived Ta-178 then decays to stable hafnium-178, 99.2% of the time by electron capture and 0.81% by positron emission. Electron capture results in a 61.2% branch to the ground state of Hf-178 and 33.7% to the first excited state at 93.1 keV; a 93.1 keV photon is emitted in 5.9% of the disintegrations (Fig. 2). The remaining 4.3% feeds hafnium levels between 1175 and 1772 keV. Since the 93-keV transition is heavily converted, the most prominent features of the Ta-178 emission spectrum are the characteristic radiation peaks of hafnium, with energies between 54.6 and 65.0 keV (*3,4*).

This communication deals with the production of the parent radionuclide, W-178.

MATERIALS AND METHODS

The reaction chosen was $^{181}Ta(p,4n)^{178}W$. This is the one used by Siddiqi and Emery (*5*) to produce samples for spectroscopic studies, but little information was available concerning production rates. Preliminary calculations based on the nuclear evaporation code ALICE (*6*) predicted an energy-dependent cross-section, approximately Gaussian in shape, centered at an incident proton energy E_p = 34 MeV. For comparison, the results of Rao et al. (*7*) and Birattari et al. (*8*) indicated that optimum produc-

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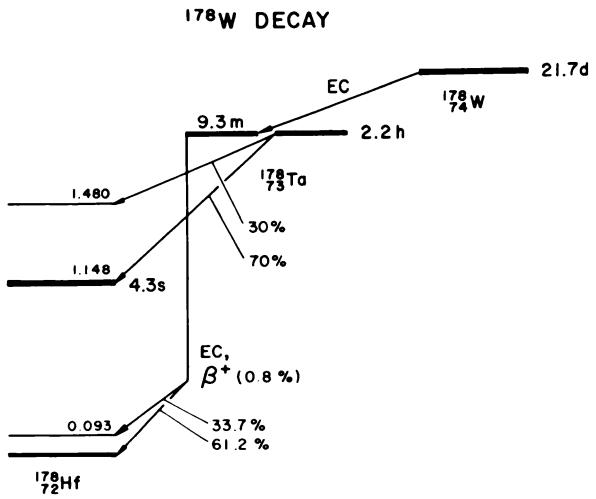

FIG. 1. Simplified decay scheme of W-178.

tion would be found at $E_p = 36$ MeV, with a full width at half maximum of 10 MeV and a maximum cross section of 760–780 mB. Based on the ALICE code, the two strongest competing reactions at 34 MeV are $^{181}Ta(p,5n)^{177}W$ and $^{181}Ta(p,3n)^{179}W$, which have cross sections lower than the (p,4n) reaction by factors of 11 and 60, respectively. At higher energies, the (p,5n) cross section increases and the (p,3n) decreases in such a way that, for example, at 40 MeV the (p,5n) reaction is 3.3 times stronger than the (p,4n) and the (p,3n) becomes insignificant. Thus optimal conditions were predicted to be met at 34–36 MeV, allowing a maximal production of W-178, while avoiding that of W-177. The latter radionuclide decays to Hf-177 via Ta-177 ($T_{1/2} = 57$ hr), giving rise to a 113-keV gamma ray in Hf-177.

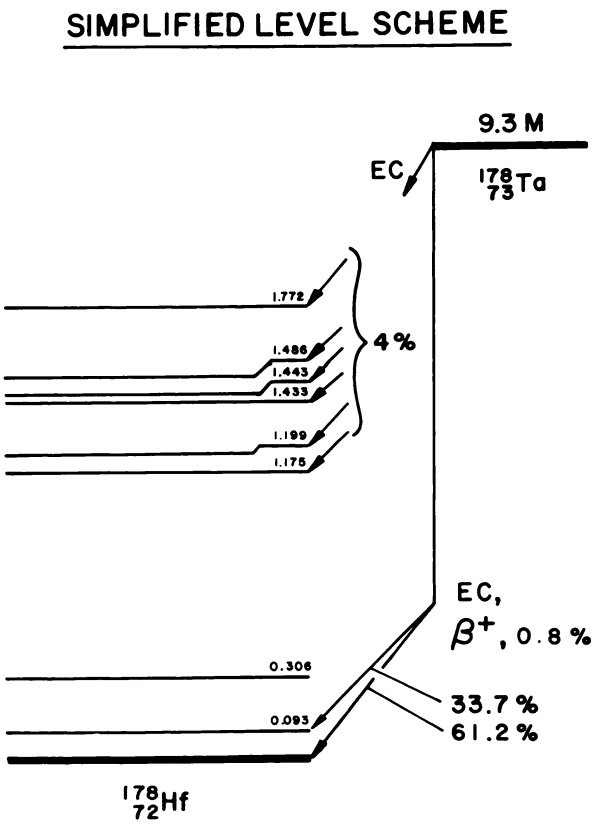

FIG. 2. Simplified decay scheme of Ta-178.

Based on the evaporation code, the production rate was calculated to be 0.22 mCi/μA-hr in a target thickness corresponding to a 1 MeV beam energy loss (7.6×10^{-3} cm of tantalum metal). For a practical target thickness of 8 MeV, therefore, the expected production rate was approximately 1.5 mCi/μA-hr.

With these calculations as a guide, five 0.005-in.-thick tantalum foils mounted in a pneumatic rabbit assembly were irradiated in the Michigan State University 50-MeV variable-energy sector-focussed cyclotron at an incident proton energy of 40 MeV. With an energy loss of 8 MeV across the target, the energy range of the incident protons thus spanned from 40 to 32 MeV.

Gamma-ray spectra were obtained using a Ge(Li) detector approximately 30 hr after bombardment, when most short-lived activities were negligible.

Separation of W-178. The irradiated portions of the tantalum foils were cut out and dissolved in concentrated hydrofluoric acid containing nitric acid. This solution was evaporated to dryness, treated with concentrated hydrochloric acid and again evaporated to dryness. The residue was dissolved in 6N HCl–1.5N HF solution and eluted through a pre-equilibrated 200–400 mesh anion-exchange column*. Spectra of the separated equilibrium samples were obtained using a Ge(Li) low-energy-photon spectrometer (LEPS), a planar crystal of 0.6 cc active volume.

RESULTS

Maximum yields of W-178 activity occurred at $E_p = 34$ MeV, in reasonable agreement with previous experiments and the evaporation-code calculations. The activities at $E_p = 39$ MeV and 32 MeV were each 83% of the maximum, indicating less dependence on energy than predicted.

Ge(Li) gamma-ray spectra taken 30 hr after the cyclotron bombardment showed that the relative intensity of the 113-keV gamma ray from Hf-177 decreased rapidly in intensity with decreasing beam energy, varying from 2.4 times that of the 93-keV peak in Hf-178 at $E_p = 39$ MeV to 0.16 at $E_p =$

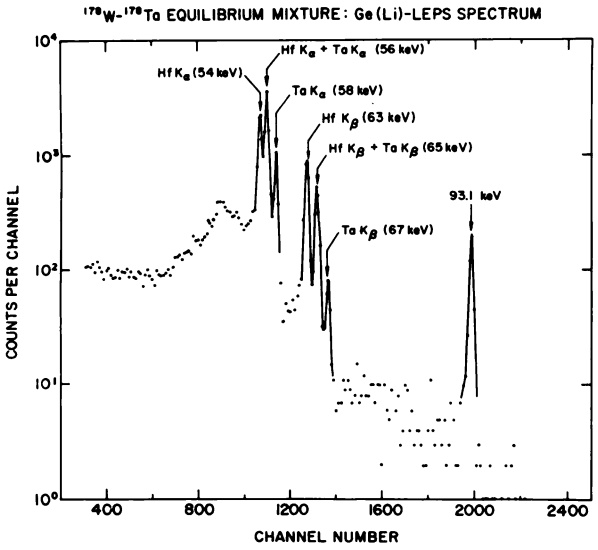

FIG. Ge(Li) pulse-height spectrum of ^{178}W-^{178}Ta equilibrium mixture after separation from irradiated Ta-181 target.

TABLE 1.

	Energy (keV)					
	54–56 63–65	(Ta-178)	80		140	
	HVL*	%T†	HVL	%T	HVL	%T
Muscle	3.12	80	3.61	83	4.32	85
Bone	1.21	56	1.75	67	2.37	75

* Half-value layer (cm).
† Transmission percentage through 1 cm of tissue.

36 MeV. At $E_p = 35$ MeV and below, the 113-keV peak was practically negligible.

The carrier-free chemical separation of W-178 from the irradiated tantalum targets resulted in yields of about 98%. Figure 3 shows a Ge(Li)-LEPS spectrum obtained from one such sample. The dominant features are the intense tantalum and hafnium characteristic radiations following electron-capture decay of W-178 and Ta-178, respectively. Tantalum K_α and K_β lines are seen at 56–58 and 65–67 keV, and the corresponding hafnium lines are at 54–56 and 63–65 keV. The 93-keV peak in Hf-178 can be seen at about 1/12 the intensity of the characteristic x-ray radiation, which is in agreement with the estimates of Nielsen (*4*). The transition is 83% internally converted, leading to an effective incidence of 6.3% per Ta-178 disintegration. Using this figure, a production figure of approximately 1.1 mCi/μA-hr per MeV of target thickness was achieved.

DISCUSSION

The short-lived radionuclide Ta-178 has physical decay characteristics suitable for use in diagnostic nuclear medicine. Furthermore, the half-life of the parent W-178 (21.7 d) is long enough so that this nuclide could form the basis of a generator system for its short-lived product.

This study demonstrates that W-178 can be produced readily, using simple target techniques, in both the quantity and purity that would be demanded in diagnostic nuclear medicine. The optimal average proton energy in the target was 34 MeV, at which energy the (p,5n) product W-177 is negligible. This corresponds to a 38-MeV incident proton energy for a practical target thickness of 8 MeV.

The major limitation to diagnostic imaging with Ta-178 is its low photon energy. The half-value layers and transmission percentage through 1 cm of tissue are shown in Table 1 for Ta-178 (Hf-178 characteristic x-rays) and for 80- and 140-keV photons. The transmission percentage through muscle is only 3% less for Ta-178 than for 80-keV photons, but 11% less through bone.

The most likely application for Ta-178 is for cardiac imaging. Accurate assessment of ventricular function requires high count rates when first pass techniques are used. High doses of Ta-178 could be administered if the multiwire proportional counter were used as the detector, thereby obtaining a more accurate assessment of regional wall-motion abnormalities and regional cardiac hemodynamics. Thus, multiple studies could be performed at reasonable intervals under physiologically or pharmacologically altered conditions because of the short half-life of the radiotracer. Since the heart is not a deep-seated organ and can be closely approached by an external detector, and since the multiwire proportional camera is efficient for lower photon energies, tissue absorption may be sufficiently offset to warrant further investigation of this use.

FOOTNOTE

* Bio-Rad AG1-X8.

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