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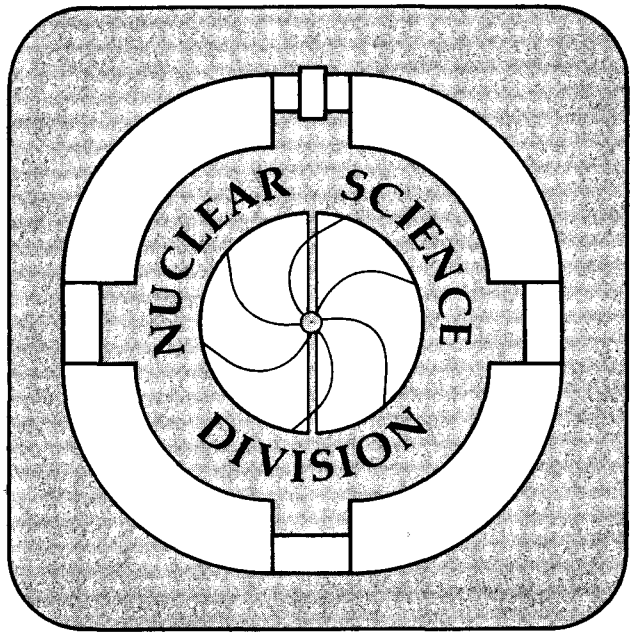

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Thick-target yields of iodine isotopes from proton interactions in Te, and the double-β decays of ^{128,130}Te

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Abstract

We measured thick-target yields of ^{126,128,130}I from bombardments of natural Te targets with 15-, 30-, 45- and 50-MeV protons, together with iodine production cross sections for 1.85- and 5.0-GeV protons. Using these data, we have estimated the relative cosmic-ray induced production of ^{126}Xe, ^{128}Xe and ^{130}Xe in Te ores. These quantities are significantly different from those used previously in a determination of the ratio of the double-β decay half-lives of ^{130}Te and ^{128}Te. A revised correction of cosmic-ray produced xenon can change the half-life ratio by as much as 8%. This quantity is of importance because it can be used to set a limit on the 0-ν double-β decay mode.

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I. INTRODUCTION

The first evidence of ^{130}Te double-β decay was obtained more than forty years ago by Inghram and Reynolds [1], who performed a geochemical experiment, showing that ^{130}Te decays to ^{130}Xe. The geochemical method is based on the measurement of the number of double-β decay daughter atoms that are trapped in the parent mineral. The accuracy of this technique depends on several factors. First, large differences in the chemical properties of the parent and daughter species facilitate the separation and counting of the relatively small number of daughter atoms. Second, contaminations of the parent mineral with the daughter species at the time of formation of the mineral can limit the technique's accuracy. Third, the mobility of the daughter species in the mineral and the diffusion times will effect the measurement. Besides simple diffusion of the atoms, seismic and thermal effects must be considered. Fourth, the ore must be precisely dated and be of sufficient age that an adequate concentration of the daughter species has accumulated to permit accurate measurement. Finally, the measured quantity of double-β decay candidate nuclei must be corrected for other processes that can also generate the daughter species. Processes such as fission, charged-particle capture and cosmic-ray spallation reactions must be considered. So far, the geochemical method has been successfully applied only to those double-β decay candidates which lead to noble-gas daughter species: $^{82}\mathrm{Se}\rightarrow{}^{82}\mathrm{Kr}$, and $^{128,130}\mathrm{Te}\rightarrow{}^{128,130}\mathrm{Xe}$. These results have been recently reviewed by Manuel [2]. The ^{82}Se case is of particular interest because double-β decay has been measured for this system both by the geochemical method and by direct counting. The agreement of the half-lives determined by these two very different techniques provides an excellent consistency check of both methods.

The double-β decay half-lives of ^{128,130}Te have been recently remeasured by Bernatowicz *et al.* [3], using the geochemical method. A recent review by Moe [4] points out a possible discrepancy between the absolute half-life values obtained by Bernatowicz *et al.* and those from previous results [2], where the half-life of ^{130}Te decay was measured relative to that of ^{82}Se, from ores containing minerals of both elements. However, the half-life ratio

$t_{\frac{1}{2}}(^{130}\mathrm{Te})/t_{\frac{1}{2}}(^{128}\mathrm{Te})$ is independent of the age of the minerals and of the retention of the daughter atoms. Bernatowicz *et al.* measured the number of atoms, and isotopic abundances of Xe present in samples collected from several tellurium ores. Deviations from the isotopic abundances of atmospheric Xe, in the form of excesses of some of the isotopes, were found. As can be seen from Fig. 1, several processes besides double-β decay can produce Xe isotopic excesses: (i) fission, (ii) neutron capture on Te, (iii) $^{127}\mathrm{I}(n,\gamma)$, (iv) α-particle-, (v) neutrino-, (vi) cosmic-ray muon- and secondary-proton reactions. After correcting the measured excesses for the mechanisms (i-v), there still remained a small excess of ^{126}Xe, which is not a double-β decay product, that Bernatowicz *et al.* attributed to muon- and proton-induced reactions.

Cosmic rays observed at sea level consist mostly ($\approx$75%) of muons, with a vertical flux of about 1.3×10^2 m^{-2}s^{-1} [5]. The muon energy spectrum falls steeply with energy, $\propto E^{-2}$, and faster above a few TeV. Muons are highly penetrating particles; they are observed in several underground experiments, with a total flux decreasing approximately exponentially as a function of the depth [6,7].

Bernatowicz *et al.* considered muon-induced reactions, and also those caused by secondary protons (*i.e.* protons produced in the muon-induced nuclear spallation in the rock), showing that their contribution does not affect the value inferred for the ^{130}Te double-β decay half-life, because they account for only 10^{-5} of the ^{130}Xe measured excesses. On the other hand, the same calculations show an effect as large as 20% for the ^{128}Te half-life, as observed in one of Bernatowicz *et al.*'s samples. However, the correction to the ^{128}Xe excess for this mechanism depends critically on how one scales the ^{128}Xe excess from that of ^{126}Xe, the "cosmic-ray detector". Because these corrections are model-dependent, we decided to address them by measuring the yields of iodine from proton reactions on tellurium targets, to shed light on the contribution of the secondary protons underground, and estimate that of direct muon reactions.

II. EXPERIMENT

The Te targets were made of 99.99% purity metallic tellurium, purchased from Johnson Matthey. Tellurium pieces were crushed and pressed at 120 °C for 4 min, to form the targets. For the low-energy activations ($\leq$50 MeV) the beam was stopped in the targets, which were disks, 1.9 cm in diameter and 1.0 cm thick, with densities of about 6.01 g/cm^3 (96-97% of the crystalline tellurium density). The targets for the high-energy experiments were (i) a disk, 3.0 cm in diameter and 0.7 cm thick for the 1.85-GeV activation, with a density of 6.04 g/cm^3, and (ii) a square, 5.1 $\times$ 5.1 $\times$ 1.0 cm for the 5.0-GeV activation, with a density of 5.66 g/cm^3.

The low-energy proton activations were performed at the 88-Inch Cyclotron at Lawrence Berkeley Laboratory; those at high-energy were performed at the LBL's Bevatron accelerator. At the Bevatron, the targets were held in air, with the Te slabs assembled in a stack, together with polycast acrylic plastic sheets (polymethyl methacrylate, $[C_5O_2H_8]_n$). These plastic sheets served to monitor the integrated beam exposure, through the production of ^{11}C, from the C and O contents of the plastic [8,9]. The 15-, 30-, 45-, and 50-MeV proton activations were done with bombardment times of 10 min for the first two, and one hour for the last one, each having an integrated current of 10 μC. The 1.85- and 5.0-GeV proton experiments were done with bombardment times of approximately one hour, with integrated currents of 60 and 5 nC, respectively. To check for iodine losses during the activations, due to the heating of the target, one of the activated targets was heated to $\approx$100 °C on a hot plate, for a period corresponding to the bombardment time. The radioactivity lost was less than 1%.

Following the irradiations, the targets were γ-counted with a 100 cm^3 coaxial HPGe detector inside a 5-cm thick lead shielding. Due to the widely different half-lives of the iodine isotopes under study: ^{126}I (13 days), ^{128}I (25 min), ^{130m}I (9 min), and ^{130g}I (12 hours), three different sizes of time bins were used for counting: 5-min bins during the first hour, 1-hour bins during the next 24 hours, and then several 6-hour bins. Some of the targets were

measured two weeks later, to confirm the amount of radioactivity attributed to the 13-day component. All photopeaks in the spectra could be attributed to reactions on Te.

The data analysis was carried out by fitting the photopeaks of characteristic γ-rays of each isotope, with emission probabilities greater than 1% for all spectra, using the program GELIFIT [10]. The time-dependent yields of each γ-ray line were fit to determine initial activities. The decay of the 9-min isomer of ^{130}I has no pure, intense transitions (*i.e.* with an emission probability >1%) that are not produced in the decay of ^{130g}I. We then performed a two-component fit of the 536-keV photopeak time-dependent yields to deduce the separate contributions of ^{130m}I and ^{130g}I. All half-lives measured were in agreement with the values found in the literature, [11] indicating no loss of iodine from the targets during the measurements.

The γ-ray detection efficiency curves for the thick-target yields were determined as follows. We measured the detection efficiency for standard γ-ray calibration sources placed in front and behind our targets, for several locations on the target surface. These scans of the target surface were then averaged for the front and back measurements separately, to approximate the extended-source geometry. We then corrected the efficiency data for coincident γ-ray summing effects using the program KORSUM [12], and fit the front and back efficiencies as a function of the photon energy. Because the high-energy measurements were done with target thicknesses much smaller than the corresponding proton range, we used the geometric average of the previous two efficiency curves as our effective efficiency. This corresponds to a first-order correction of the self-attenuation and solid-angle difference between the front- and back efficiencies. For proton energies of 50 MeV and below, the targets completely stopped the beam, so we had to do a different correction for the self-attenuation. For this, we fit the total photon-absorption cross-section for tellurium in the energy range of interest, with data taken from the work of Hubbell [13]. Then we took the efficiency curve determined for the front of the target and folded it with an energy-dependent term of the type $exp[-\mu(E) \times \frac{R}{2}]$, where $\mu(E)$ is the energy-dependent photon-absorption coefficient, and R is the range of the protons. We also performed a solid-angle correction to obtain

the effective detection efficiency. We verified that these effective efficiencies reproduced the correct relative γ-ray intensities for all decays studied. Fig. 2 shows a spectrum obtained from the 50-MeV activation, with the principal γ-rays identified.

III. RESULTS AND DISCUSSION

To make a statement about the proton-induced xenon excesses, Bernatowicz *et al.* used a simple model. They assumed that only (p,n) reactions take place, and that they all have the same cross section. Then, the relative yields of the resulting Xe isotopes would depend only on the Te isotopic abundances in the ores and on the β-decay branching ratios of the produced iodine to xenon. While for sufficiently low-energy protons it is true that only (p,n) reactions are possible, it can be seen from Table I that the Q-values for the ^{128,130}Te$(p,3n)^{126,128}$I and ^{130}Te$(p,5n)^{126}$I reactions are not terribly high. The energy spectrum of protons underground is not well characterized. Thus it is reasonable to expect that these additional reactions could also contribute to the observed ^{126,128}Xe excesses. The low-energy bombardments we did provide information on the separate contributions of the (p,n), $(p,3n)$ and $(p,5n)$ reactions. One can generalize the model of Bernatowicz *et al.* to include these reactions by assuming that, where they are energetically allowed, they have the same cross sections as those of the (p,n) reactions. The relative yields predicted by this model are shown in Table II.

Table III shows our results for the iodine thick-target yields, and the production cross sections for the high-energy experiments. The integrated beam current of the 50-MeV activation suffered a systematic error, later identified as being due to the high conductivity of the water used to cool the target and collimator assemblies. For this activation we only quote relative yields. Table IV contains the relative Xe yields inferred from the data in Table III. We also present data calculated from thick-target iodine yields of Roughton *et al.* [14].

Based on their model, Bernatowicz *et al.* inferred $N(^{128}\text{Xe})/N(^{126}\text{Xe})$=3.42 (this is 3.47

for the mass ratio). If we assume that (p, xn) reactions are the sole source of iodine and xenon in the ore, we would expect the relative Xe yields to be roughly given by the experimental ratios in Table IV. However, the appropriate value to use is obviously dependent on the proton energy spectrum. Table IV shows that even for energies between 3 and 15 MeV, the 3.42 ratio is only achieved in a very narrow energy range, a more appropriate value lying between $\approx$1 and $\approx$3.

The muon reaction $\mu^{\pm} + {}^{A}\mathrm{Te} \rightarrow \mu^{\pm} + \pi^{-} + {}^{A}\mathrm{I}$ is the equivalent of a (p, n) reaction. Bernatowicz *et al.* estimated that proton- and muon-induced reactions contribute roughly equal amounts of Xe. We can take into account these direct muon reactions, and keep as 3.42 the ^{128}Xe to ^{126}Xe ratio from their contribution. Using 1.0 for the xenon ratio coming from secondary proton reactions, yields a total ratio of 1.75, assuming equal reaction rates for muons and protons. In this case, the half-life ratio becomes $(3.77 \pm 0.13) \times 10^{-4}$, which is consistent with the result obtained by Bernatowicz *et al.* for the deeply buried Kalgoorlie krennerite sample.

In conclusion, we find that the ratio of cosmic-ray produced ^{128}Xe/^{126}Xe is energy dependent. The simple model developed by Bernatowicz *et al.*, and generalized by us, fails to reproduce the measured ratios at any energy. Combining these results with the fact that the energy spectrum of underground protons is not well known, leads to an additional systematic uncertainty of 10% in the ratio of the double-β decay half-lives of ^{128,130}Te. With our half-life ratio of 3.77×10^{-4} and the data from Bernatowicz *et al.* for ^{130}Te half-life, we calculate a half-life of $(7.2 \pm 0.4) \times 10^{24}$ years for ^{128}Te. The values for the upper limits of the non-standard parameters are no more than 5% different from those obtained by Bernatowicz *et al.*, *i.e.*, 2.4 eV for the effective neutrino mass, $\langle m_\nu \rangle$, 6.0×10^{-8} for the mixing of left- and right-handed weak currents, $\langle \eta \rangle$, and 4.9×10^{-6} for the pure right-handed weak-current term $\langle \lambda \rangle$, as calculated in the manner of Suhonen *et al.* [15]. If we calculate the neutrino mass limit by taking both, $\langle \eta \rangle$ and $\langle \lambda \rangle$ as zero, we then get $\langle m_\nu \rangle \leq 1.5$ eV.

IV. ACKNOWLEDGMENTS

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TABLES

TABLE I. Q-values, in MeV, for the (p, xn) reactions of interest.

Target	(p, n)	$(p, 3n)$	$(p, 5n)$
^{126}Te	–2.9		
^{128}Te	–2.0	–18.0	
^{130}Te	–1.2	–16.5	–32.5

TABLE II. Relative production of Xe in a simple model in which it is assumed that, where they are energetically allowed, all (p, xn) reactions have the same cross section.

	$x = 1$	$x = 1, 3$	$x = 1, 3, 5$
$N(^{128}\mathrm{Xe})/N(^{126}\mathrm{Xe})$	3.42	2.65	1.59
$N(^{130}\mathrm{Xe})/N(^{126}\mathrm{Xe})$	3.89	1.45	0.87

TABLE III. Our results for the ^{126,128,130}I thick-target yields, together with the (p, xn) cross sections of the high-energy activations.

E_p	^{126}I	^{128}I	^{130}I
(MeV)	(10^{-4} nuclei/proton or mb) [a]		
15	1.31(2)	1.08(1)	0.42(1) g [b]
			0.70(1) m
30	13.5(1)	11.0(1)	1.04(3) g
			0.98(2) m
45	23.6(5)	13.8(1)	1.1(1) g
			0.90(6) m
1850	6.9(23)	5.7(4)	3.0(1) [c]
5000	15(6)	10(2)	4.6(9) [c]

[a] The values quoted for the 1850- and 5000-MeV activations are cross sections, in mb, for the (p, xn) reactions on natural Te.

[b] "g" and "m" identify the separate yields for the ground- and meta-stable states of ^{130}I

[c] The decay of the 9-min isomer was not observed in the high-energy experiments. These cross sections were calculated from the observed activities of the ground state only, being a combination of the value for the ground-state, plus 83% of that for the isomer, corresponding to the branching ratio for the isomeric transition.

TABLE IV. Relative Xe production from the thick-target I yields of Ref. 14, (upper part) together with our data (lower part), ^{126}Xe≡1. The numbers in parentheses are the uncertainties in our measurements.

E_p (MeV)	^{128}Xe	^{130}Xe
3.029	0.75	2.15
3.346	3.70	4.43
3.934	2.34	3.61
4.292	2.93	4.07
4.645	2.00	2.92
5.049	1.75	2.80
5.687	2.42	3.86
6.082	2.81	3.17
6.360	1.87	3.51
15	1.76(9)	1.95(10)
30	1.74(8)	0.34(2)
45	1.25(6)	0.19(2)
50	0.94(22)	0.17(5)
1850	1.4(5)	0.39(14)
5000	1.1(5)	0.28(13)

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FIGURES

FIG. 1. Region of the chart of the nuclides around Te-I-Xe. The circles contain the mass number of the isotopes in the corresponding diagonal, the filled squares represent stable isotopes, and the dashed ones are the double-β decay parent (^{128,130}Te) and daughter (^{128,130}Xe) nuclei. The radioactive iodine isotopes relevant for the problem have their half-lives and decay modes indicated.

FIG. 2. Partial γ-ray spectrum observed from the 50-MeV proton activation.

126
128
130
132
129
127
125
Xe
I
Te
β⁻, 44%
13d
β⁻, 93%
25m
β⁻
12h
9m
ε, 56%
ε, 7%

Thick–target yields of iodine isotopes...

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Figure 1

Counts per 0.447 keV
10^2
10^3
10^4
10^5
10^6
0
200
400
600
800
Energy (keV)
Xe x rays
Pb x rays
159.0 123I
212.5 121I
388.6 126I
442.9 128I
536.1 130g,mI
666.4 126I
668.6 130gI
739.5 130gI
753.9 126I

Thick–target yields of iodine isotopes...

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Figure 2

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