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INTRODUCTION

This work describes a cyclotron technique for the production of high purity ^{123}I using the ^{121}Sb (α, 2n)^{123}I method. The production of this isotope has been prompted by its recent recognition as the most ideal radioisotope of iodine for low exposure in vivo measurements. (ref. 1). It has been calculated, for example, that the ^{123}I radiation dose to a patient in a human thyroid uptake test is only a few percent of that received with the more commonly used ^{125}I and ^{131}I (ref. 2). This reduction results from the fact that ^{123}I emits far less charged particle radiation than does ^{131}I and its physical half-life is only 0.9 percent and 6.9 percent of that of ^{125}I and ^{131}I, respectively. In addition, Wagner et al., who have introduced formulae to determine the ideal radiopharmaceutical as one with a physical half-life no longer than necessary to insure maximal activities in the organ studied at the optimal observation time, have found ^{123}I to be considerably better than ^{125}I and ^{131}I (ref. 3). Furthermore, the 159 keV gamma ray of ^{123}I approaches an optimal energy which is low enough to be efficiently counted with thin crystals and allow use of high resolution

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collimators, but high enough that very little tissue absorption occurs. With all of these desirable characteristics, it is obvious why this nuclide is in demand for nuclear medical applications. However, until recently, ^{123}I has been produced on only a few occasions in order to evaluate its efficacy as a radiopharmaceutical. Beaver and Halley (ref. 4) and Eldridge (ref. 5) have produced ^{123}I at the ORNL 86-inch cyclotron by the proton bombardment of enriched ^{123}Te according to the reaction ^{123}Te (p, n) ^{123}I while Myers and Anger (ref. 1) report its production at the 60-inch Crocker cyclotron by ^{4}He reactions on enriched ^{121}Sb. Both of these methods have resulted in yields of ^{123}I which were adequate for nuclear medical applications but which were contaminated with other iodine isotopes, notably ^{124}I, with half-lives longer than that of ^{123}I.

More recently, the Atomic Energy Commission has announced that they will produce and distribute ^{123}I on a limited basis for nuclear medical applications (ref. 6). This isotope, which will be made by the ^{123}Te (p, n) ^{123}I method, will contain about 3% iodine contamination at the time of processing (ref. 7). This contamination has been considerably reduced in the present work by using the ^{121}Sb (α, 2n) ^{123}I method which results in less than 0.38% ^{124}I contamination.

Also described in this work are the chemical procedures used to isolate the iodine products and those used to recover the enriched target material for subsequent use.

THEORY

Several nuclear reactions using stable xenon, tellurium and antimony as target nuclei lead to ^{123}I as a residual nucleus. In the majority of cases, however, the target isotope has very low natural abundance, and consequently, because of the effort and expense involved is not available in a highly enriched form. Target material free of isotopes with neutron number one to three higher than the target nucleus is necessary so that the same nuclear reaction does not produce longer lived ^{124}I, ^{125}I or ^{126}I with the desired ^{123}I. Iodine-124, which decays with a 4.2 day half-life and emits approximately 2.3 photons per disintegration ranging in energy from 0.028 to 2.26 MeV is an undesirable contaminant because it seriously masks the ^{123}I x-ray and gamma ray spectrum. Furthermore, ^{124}I decays 25% of the time with emission of 0.9 to 3.3 MeV positrons which contribute heavily to radiation dose. Iodine-125, which emits 27.5 to 31.7 keV x-rays and 35.4 keV gamma rays at a total rate of 1.4 photons per disintegration, does not interfere with the higher energy 159 keV gamma ray peak of ^{123}I, but because its x-rays are identical in energy to those of ^{123}I, their presence complicates calculation of ^{123}I x-ray to gamma ray ratios used for depth correction. (This technique for depth correction has been described in reference (8)). Iodine-126, which decays by emission of 0.4 to 1.25 MeV positron and electrons 96% of the time and emits 1.2 photons per disintegration with energies ranging from 20 to 747 keV interferes both with the ^{123}I photon spectrum and contributes heavily to radiation dose. Because of their shorter half-life (hence their higher decay rate), and their high emission rate of beta-like radiation, ^{124}I and ^{126}I are considered more serious contaminants, atom for atom, than ^{125}I.

Three possible target materials available in enriched isotopic form are ^{122}Te (90-95%), ^{123}Te (65-80%) and ^{121}Sb (95-99.5%)*. These target isotopes result in ^{123}I residual nuclei via ^{122} Te (d, n,), ^{123}Te (p, n) and ^{121}Sb (α, 2n) reactions. The (α, 2n) and (d, n) cross sections are somewhat larger than the (p, n) reaction. In addition, consideration of the impurities indicates the (α, 2n) reaction to be preferred. The distinct advantage of this method is that ^{121}Sb is available in the highest enriched form, with at worst, only a few per cent ^{123}Sb contamination which results primarily in ^{125}I. In contrast, ^{122}Te and ^{123}Te are contaminated with several other tellurium isotopes, notably ^{123}Te and ^{124}Te which upon (d, n) and (p, n) reactions respectively, produces the more serious contaminant, ^{124}I.

In using the ^{121}Sb (α, 2n) ^{123}I method for ^{123}I production, another avenue of ^{124}I contamination must be considered. If the incident energy of the alpha particle beam is not sufficiently high for emission of the second neutron, the (α, n) reaction with ^{121}Sb will produce ^{124}I. In

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this work, the correct alpha particle energy was estimated assuming that the (α, 2n) reaction is optimal at the threshold energy for the (α, 3n) reaction (ref. 9). The threshold energy was approximately calculated using mass excess values (ref. 10) for the tertiary reaction. This calculation shows that the cross section for ^{121}Sb (α, 2n) ^{123}I reaction is maximum using approximately 25 MeV alpha particles. In practice, the reaction energy can be slightly higher than 25 MeV since the tertiary reaction, ^{121}Sb (α, 3n), results in 3.5 minute ^{123}I which decays quickly and hence, is not considered a significant contaminant.

EXPERIMENTAL

Preparation of Target

The enriched ^{121}Sb (98.4%) was spread uniformly into a cavity milled into a water-cooled aluminum target plate and covered with tungsten foil. The area of the cavity was slightly larger than the alpha-particle beam area and its depth was just large enough to contain the powder so that when covered and in the vertical position the powder distribution remained uniform. The tungsten foil, which was 1 to 4 mil thick prevented escape of target material into the cyclotron's vacuum system as well as attenuated the beam energy to the desired value.

Irradiation

A typical irradiation of a 50 mg Sb target with a superficial density of 70 mg/cm^2 requires an incident beam energy of 30 MeV. Using Range-Energy tables (ref. 11), it is calculated that in traversing

70 mg/cm^2 of antimony this energy would attenuate to an average energy of 26 MeV which is just slightly greater than the calculated optimal energy for the ^{121}Sb (α , 2n) ^{123}I reaction. The target was irradiated for 3 to 4 hours at a beam strength of 4.0 μ amp without significant damage to the target holder.

Chemical separation of ^{123}I

The antimony powder (50 mg) was removed from the target holder under acetone, collected on a glass fiber filter and dissolved under reflux with 400 μg iodine carrier in 20 ml concentrated H_2SO_4. The solution was cooled and diluted to 120 ml with distilled water. The iodine then was distilled into a receiving vessel containing 50 ml. 0.5 N NaOH. The chemical yield of this procedure using ^{131}I tracer was 91.2 ± 1.2%. The yield of an actual ^{123}I production run was determined by gamma counting the relative amounts of ^{123}I (or any other iodine present as a contaminant) both in the distillation residue and distillate.

Chemical Recovery of ^{121}Sb

The acidic residue from the iodine distillation containing 50 mg ^{121}Sb was neutralized with NH_4OH and treated with 3 ml hydrazine hydrate ($N_2H_4H_2O$). The solution was adjusted in acidity to pH 1 with H_2SO_4 and saturated with gaseous H_2S at room temperature.

The Sb_2S_3 precipitate was collected by centrifugation and washed with 1 N H_2SO_4. The sulfide was then dissolved in 10 ml concentrated HCl, boiled to remove H_2S, diluted with 10 ml distilled water and filtered through a glass fiber filter. The clear solution was treated with chromous chloride to reduce the Sb as the metal which was then collected on a filter, washed with ethyl alcohol and dried at 100^o. The chemical yield determined with natural antimony carrier and ^{125}Sb tracer was 94.8 ± 1.5%. The recovered ^{121}Sb was analyzed for gamma emitting radionuclide contaminants. In a typical bombardment of 50 mg ^{121}Sb for a total charge of 0.04 coulomb, the main contaminants 60 days after the bombardment were approximately 4 nCi ^{123m}Te produced from (α, d) reactions with ^{121}Sb and approximately 0.6 nCi ^{185}Os produced probably from (α, n) reactions with ^{182}W in the tungsten covering foil. These slight contaminants did not interfere with subsequent reuse of the target material.

RESULTS

Some typical decay spectra of an iodine fraction which was chemically separated by distillation are shown in Figures 1 and 2. Figures 1a and b show photon decay curves from 0 to 800 keV at 20 hours and 13.5 days, respectively, after irradiation. No major activity was found above 800 keV. In Figure 2a, b and c are gamma and x-ray spectra in the energy region from 7 to 50 keV accumulated

7.5 days, 64.6 days and 106.5 days, respectively, after the irradiation time. All spectra in Figures 1 and 2 were measured with a pulse height spectrometer using a 6 cm^2 x 7 mm lithium drifted semiconductor detector. The photon peaks were identified by energy and half-life decay analysis. The yields of various products were determined by integration analysis of the counts under their respective photon peaks. The yield of radioiodine products isolated from a 50 mg ^{121}Sb target bombarded for a total charge of 0.37 coulombs is shown in Table 1. All yields have been decay-corrected back to the end of the bombardment time. Also shown for each product are its half-life, the energy of the gamma ray peak used in the spectral analysis and the abundance value of that gamma ray used in the yield calculations (ref. 12). All errors in the yield values are 2 σ counting errors.

DISCUSSION

The spectrum in Figure 1a shows that the major activity present in the distilled iodine fraction 20 hours after bombardment are the 27.5 to 31.7 keV x-rays and the 159 keV gamma ray of ^{123}I. Also evident is the 530 keV gamma ray of ^{123}I which is only 2% abundant relative to the 159 keV gamma ray. Other peaks are caused by the 514 keV annihilation photon of ^{124}I (plus a little contribution from

TABLE I

Radioiodine Yield from 50 mg Enriched ^{121}Sb Target

Nuclide	$t^{1/2}$	E MeV	% Abundance	Yield (mCi)	Yield %
^{123}I	13.3 h	0.159	84	5.75 ± 0.05	99.6
^{124}I	4.2 d	0.603	66	(2.18 ± 0.06) x 10^{-2}	0.38
^{125}I	60 d	35.4	7	3.1 x 10^{-4}	5.3 x 10^{-3}
^{126}I	12.8 d	0.386	34	(1.26 ± 0.12) x 10^{-4}	2.2 x 10^{-3}

^{126}I), the 604 and 722 keV gamma rays of ^{124}I and the 665 keV gamma ray of ^{126}I. Other low abundance peaks in Figure 1 at 440-, 508-, and 540 keV decay with a 21 ± 2 hour half-life and are not positively identified. The 508 keV peak could possibly be due to ^{182}Os produced from (α, 2n) reactions with the ^{180}W in the tungsten foil covering the target assembly. The volatility of Osmium as the octavalent oxide (ref. 13) would explain its presence in the iodine fraction. The 440- and 540 keV photon peaks which also decay with a half-life similar to that of ^{182}Os are not listed for that isotope in the literature (ref. 12). Figure 1b accumulated 13.5 days after the irradiation shows no evidence of ^{123}I and only the longer-lived ^{125}I and ^{126}I contaminants remain. In Figure 2 are the decay spectra of the 27.5 to 31.7 keV x-rays of ^{123}I, ^{124}I, ^{125}I, and ^{126}I. In Figure 2a the x-ray spectrum completely masks the lower abundant 35 keV gamma photon peak of ^{125}I. However, after 64.6 days decay (Figure 2b) the ^{125}I peak becomes apparent, and after 106.5 days decay (Figure 2c) essentially only ^{125}I remains.

The results in Table 1 show that the isotopic purity of ^{123}I is better than 99.6% with the major iodine contaminant being ^{124}I which is only 0.38% of the total iodine activity. The combined concentration of ^{125}I and ^{126}I is less than 10^{-2}% of the total iodine yield.

The radioiodine contamination in the ^{123}I produced in this work using the ^{121}Sb (α, 2n) method is about 8 times less than that specified from the ^{123}Te (p, n) method (ref. 7). The yields in Table 1 also compare favorably with those of Eldridge et al. (ref. 14), who reports per cent yields of ^{124}I, ^{125}I and ^{126}I from the ^{123}Te (p, n) method as 0.85, 0.08 and 0.22%, respectively.

The maximum yield of ^{123}I using the present method for a 3 hour bombardment with a beam strength of 30 μ Amp on a 200 mg ^{121}Sb target would be 150 mCi.

It has been reported that in vivo measurements of the human thyroid can be made with less than 100 μCi ^{123}I administered orally (ref. 14). Therefore, from a yield of 150 mCi of ^{123}I, approximately 100 to 200 thyroid uptake measurements could be made within the following 2-3 days after production before encountering significant interference from the ^{124}I contamination.

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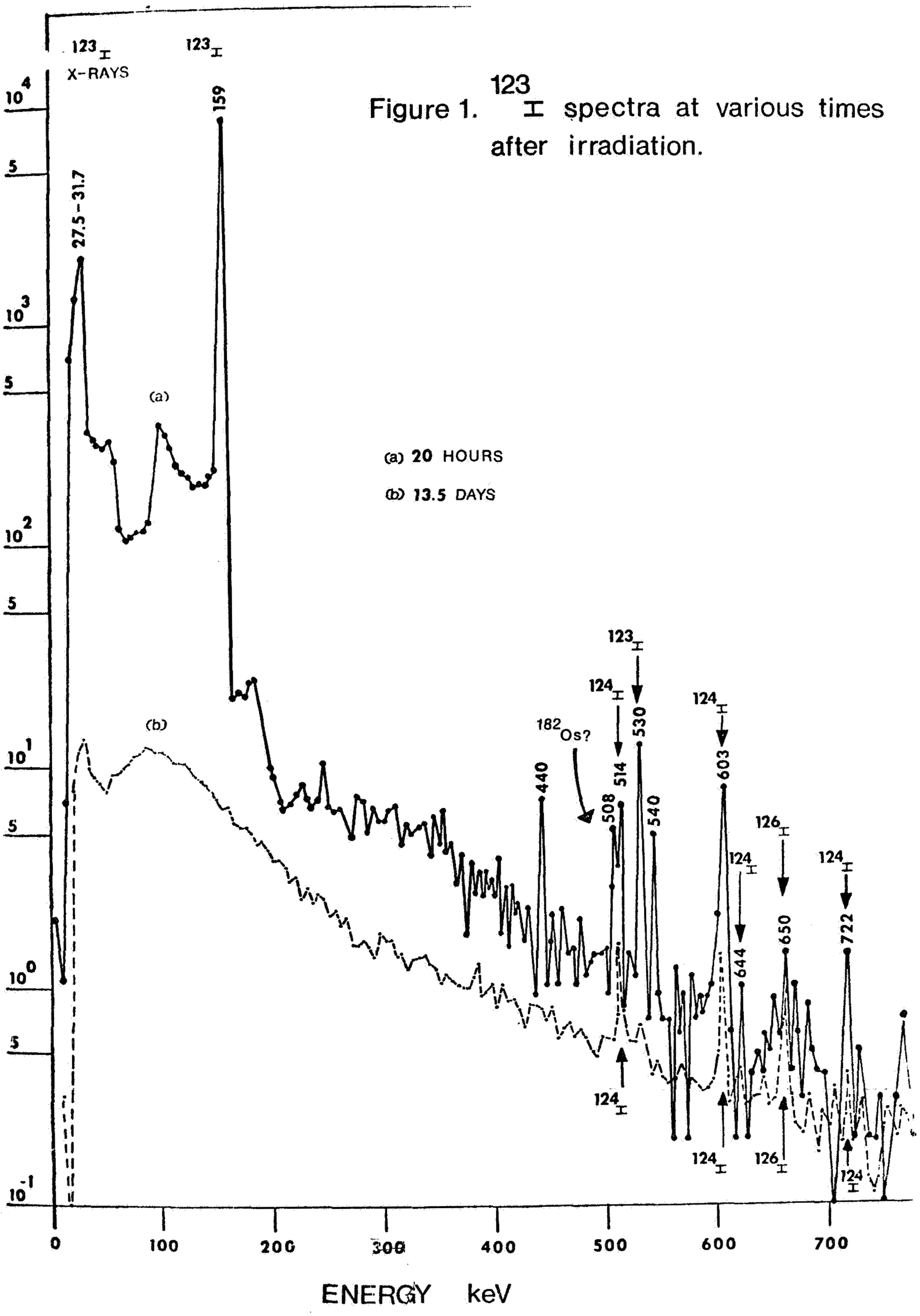

Figure 1. ^{123}I spectra at various times after irradiation.

Figure 2. Low energy spectra of iodine products at various times after irradiation.