Nuclear Instruments and Methods in Physics Research B 269 (2011) 3125–3128

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Carrier-free production of ^{95}Tc at an electron accelerator

N.I. Aizatskiy, N.P. Dikiy, A.N. Dovbnya, Yu.V. Lyashko, V.I. Nikiforov, V.A. Shevchenko, A.Eh. Tenishev, V.L. Uvarov *

NSC KIPT, 1 Akademicheskaya Str., Kharkov 61108, Ukraine

ARTICLE INFO

Article history:
Available online 22 April 2011

Keywords:
Electron accelerator
Photonuclear reaction
Bremsstrahlung
Technetium-95
Ruthenium

ABSTRACT

We report on the results of studies of the ^{95}Tc isotope production conditions in a poorly investigated ^{96}Ru(γ,N)^{95}Tc reaction at a relatively inexpensive electron accelerator. Based on the analytical model it is demonstrated the possibility for estimating the effective and peak cross-sections and the isotope yield for a given reaction in a thick production target without preliminary determination of its excitation function. For this purpose we compared specific activities of two small samples that were activated under the same conditions using the reference reaction ^{68}Zn(γ,p)^{67}Cu and the one under investigation. The experiment on simultaneous photo-activation of natural zinc and ruthenium targets was performed followed by the investigation of their isotope composition. The specific activity of the radionuclide was measured and cross-sections for the ^{96}Ru(γ,N)^{95g,m}Tc reactions were determined. The yields of desired isomers and admixtures in the natural ruthenium targets of different size were estimated. It is shown, in particular, that the operating conditions of the NSC KIPT accelerator KUT-30 can provide ^{95g}Tc yields up to 120 mCi/h.

1. Introduction

In the modern nuclear medicine, ^{99m}Tc radionuclide is one of the most widely used unstable technetium isotopes utilized in more than 80% of the diagnostic procedures [1]. For this radionuclide, a great variety of pharmaceutical carriers have been developed, which might also incorporate, if needed, other technetium isotopes and particularly ^{95g}Tc isomer ($T_{1/2}$ = 20.0 h) which shows considerable importance. Its decay is accompanied by the emission of Auger-electrons of 2.27 and 14.8 keV energy (quantum yields 96.5% and 20.3%, respectively), by characteristic X-rays of 2.29 (3.82%), 17.37 (19.5%), 17.48 (37.0%), 19.59 (2.96%), 19.61 (5.75%), and 19.965 keV (1.26%) energy and by gamma-radiation of 765.79 keV energy (93.8%) [2].

As a rule the ^{95g}Tc is generated with an admixture of ^{95m}Tc isomer ($T_{1/2}$ = 61 d) [3]. The decay of the latter is also accompanied by the emission of Auger electrons of 2.27 (94.7%) and 14.8 keV (19.9%) energy, by characteristic X-rays of 2.29 (3.75%), 17.37 (19.1%), 17.479 (36.3%), 19.59 (2.91%), 19.607 (5.64%), and 19.965 keV (1.24%) energy and by gamma-radiation with the most intense lines: 204.12 (63.2%), 582.1 (30.0%), 786.2 (8.66%), 820.6 (4.71%) and 835.15 keV (26.6%). A trace amount (0.44%) of ^{95m}Tc isomer also decays by positron emission [2].

The presence of low-energy Auger electrons that have high ionizing power, combined with its acceptable half-life of ^{95g}Tc, makes this isomer applicable for cancer therapy.

The practical utilization of ^{95}Tc is impeded by the insufficient data on the reactions for its production or by limitations of the known production techniques. Thus, ^{95}Tc can be produced by the proton irradiation of natural molybdenum. In this case, a mixture of technetium isotopes, $^{95-99}$Tc, is generated in the target. The production of ~1 Ci ^{95}Tc per day requires 1 mA proton beam of 40 MeV energy. This isotope can also be obtained by irradiating molybdenum and niobium with α-particles. The reactions on the molybdenum also yield a mixture of technetium isotopes. The α-bombardment of niobium provides the carrier-free generation of ^{95}Tc, but its yield is rather low [3].

Recently, the cross-sections of the reactions ^{96}Ru(n,d*)^{95g}Tc and ^{96}Ru(γ,n)^{95}Ru for production of ^{95}Tc using ^{96}Ru as target nucleus have been experimentally measured [4,5]. The isotope ^{95}Ru decays to ^{95}Tc by electron capture ($T_{1/2}$ = 1.64 h). These experiments have been carried out in a limited range of bombarding particle energies. In particular, the last reaction was investigated in the electron energy (E_0) range 11–14 MeV. In other words, the end-point energy of Bremsstrahlung photons used to irradiate the target was in the neighborhood of the reaction threshold, 10.69 MeV. By fitting the cross-sections, measured in this energy range, with the Breit–Wigner model, the authors have estimated the excitation function parameters for the whole region of the giant dipole resonance.

* Corresponding author. Tel.: +380 57 335 63 32; fax: +380 57 335 14 44.
E-mail addresses: aizatsky@kipt.kharkov.ua (N.I. Aizatskiy), ndikiy@kipt.kharkov.ua (N.P. Dikiy), dovbnya@kipt.kharkov.ua (A.N. Dovbnya), juvl@kipt.kharkov.ua (Yu.V. Lyashko), vinikiforov@kipt.kharkov.ua (V.I. Nikiforov), uvarov@kipt.kharkov.ua (V.A. Shevchenko), uvarov@kipt.kharkov.ua (A.Eh. Tenishev), uvarov@kipt.kharkov.ua (V.L. Uvarov).

0168-583X/$ - see front matter © 2011 Elsevier B.V. All rights reserved.
doi:10.1016/j.nimb.2011.04.098

However, direct cross-section measurements at electron energy $E_0 > 14$ MeV have not been performed. Similarly, there are practically no data on the ^{96}Ru(γ,p)^{95}Tc reaction that provides direct carrier-free production of the desired isotope.

In paper [6], the possibility of ^{95g}Tc generation through the activation of ruthenium by Bremsstrahlung photons has been demonstrated. However, the author also gave no data on the reaction cross-sections and the desired isomer yield.

2. Methods

The objective of the present work was to estimate the ^{95}Tc yield under photonuclear production conditions. When the excitation function is known, the estimation can be done using the Monte–Carlo simulation method for any target activation regime [7]. However, for the case in question, the necessary data are available. Besides that, the calculation of the photonuclear yield of isotopes by means of validated codes can frequently give, even for cases of known reaction cross-sections, results essentially deviating from experimental data [8,9]. Therefore, here we have proposed a method for estimating the reaction cross-section and the isotope yield in the target of arbitrary dimensions based on the comparison between the experimentally measured specific activities of two small samples activated under the same conditions by a reaction of known excitation function and in the channel under investigation. It is worthwhile to note that the specific activity measurement can be carried out with higher accuracy and considerably less effort than the determination of excitation function of the photonuclear reaction [5]. The method has been used to estimate the cross-sections of the ^{96}Ru(γ,N)^{95g,m}Tc reaction, and the yields of ^{95g}Tc and of the impurities for a real-size natural ruthenium target activated at the NSC KIPT electron linac KUT-30 [10].

It has been demonstrated in Ref. [11] that the photonuclear isotope yield $y_i(E_0)$, normalized to accelerated electron energy E_0, for the cylindrical target irradiated by the Bremsstrahlung photons with the Gaussian flux density distribution can be represented as

$$y_i(E_0) = \{1 - \exp[-R^2/2\delta_r^2(d + a + H/2)]\} \{1 - \exp[-\mu(k_i) \cdot H]\} \cdot y_i^{\infty}(E_0), \quad (1)$$

where $y_i^{\infty}(E_0)$ is the yield in a semi-infinite target described by the expression

$$y_i^{\infty}(E_0) = 2\eta(E_0, d) \cdot n_i \cdot \mu^{-1}(k_i) \cdot S_i(E_0) \cdot \sigma_i^{\max}, \quad (2)$$

R and H are the target radius and height respectively, d the converter thickness, a the target-to-converter distance, δ_r the rms deviation of photon flux density radial distribution, $\eta(E_0,d)$ the efficiency of electron energy conversion into Bremsstrahlung, n_i the volume density of target nuclei, $\mu(k)$ the linear attenuation coefficient of photons of energy k in the target material, $S_i(E_0)$ the convolution of the linearized photon spectrum and the excitation function of i-reaction in the Breit–Wigner formula, both properly normalized, and k_i the photon energy corresponding to the i-reaction cross-section maximum $\sigma_i^{\max}$.

In particular, the formula (1) enables one to derive the expression for the specific isotope yield in a thin target ($H << \mu^{-1}(k_i)$)

$$y_{i,m}(E_0) = dy_i/dm = \eta(E_0, d) \cdot v_i \cdot (N_A/\overline{A_i}) \cdot [\pi \cdot \delta_r^2(d + a)]^{-1} \cdot S_i(E_0) \cdot \sigma_i^{\max}, \quad (3)$$

where v_i is the relative concentration of target nuclei, $\overline{A_i}$ the average atomic number of the target material and N_A the Avogadro number.

The value of $y_{i,m}(E_0)$ can also be determined experimentally from the specific activity $A_{i,m}(E_0)$ of the sample induced by its irradiation for time period τ at an average beam current I with the electron energy E_0, using the formula

$$y_{i,m}(E_0) = eA_{i,m}(E_0)/I \cdot [1 - \exp(-\lambda_i \cdot \tau)], \quad (4)$$

where e is the electron charge and λ_i is the i-isotope decay constant.

This approach enables one to determine $S_x(E_0) \cdot \sigma_x^{\max}$ for any x-reaction with unknown excitation function. For this purpose, it is necessary to perform simultaneous photo-activation of two thin samples of different materials. One of the samples produces an isotope by a well-investigated (reference) i-type reaction, while the other one is used for the investigation of the x-reaction. Then, from the ratio of specific activities of the two samples, $A_{x,m}(E_0)$ and $A_{i,m}(E_0)$, on the basis of formulas (3) and (4), we obtain

$$S_x(E_0) \cdot \sigma_x^{\max} = v_i \cdot \overline{A_x} \cdot [1 - \exp(-\lambda_i \cdot \tau)] \cdot A_{x,m}(E_0) \cdot \{v_x \cdot \overline{A_i} \cdot [1 - \exp(-\lambda_x \cdot \tau)] \cdot A_{i,m}(E_0)\}^{-1} \cdot S_i(E_0) \cdot \sigma_i^{\max}, \quad (5)$$

where v_x and $\overline{A_x}$ are, respectively, the relative nuclei concentration and the average atomic weight of the target material to obtain the reaction under study.

Having determined $S_x(E_0) \cdot \sigma_x^{\max}$ and using formula (1), we can estimate the x-reaction yield in the target of arbitrary dimensions.

3. Results

In order to investigate the conditions of ^{95}Tc photonuclear production, we have used the ^{68}Zn(γ,p)^{67}Cu reaction as a reference one [12]. A natural zinc sample (mass 1327.6 mg) was prepared in the form of the flat plate of dimension 16 × 12 mm^2 and 1 mm thickness. The other sample consisted of ruthenium powder of natural composition (mass 64.6 mg) packed in an Al foil envelope of the same dimensions mentioned above. The samples were simultaneously irradiated at the accelerator LU-40 [13] for 6.25 h with 6 μA electrons of 40 MeV energy. The gamma-activity of samples was measured for 4 days using a 50 cm^3 Ge(Li) detector with 3.25 keV energy resolution at the 1332 keV ^{60}Co line. Figs. 1 and 2 show the spectra of the samples 24 h after the irradiation. The statistical error in determining the activity of ^{67}Cu (184.6 keV) and ^{95g}Tc (765.8 keV) does not exceed 2.5%. The total uncertainty for specific activities of the radionuclides under consideration (see Table 1) is no more than 7%.

From the specific activity data obtained for ^{67}Cu and ^{95g,m}Tc, using formula (5), one can calculate $S(E_0) \cdot \sigma^{\max}$ for the reactions ^{96}Ru(γ,N)^{95g,m}Tc:

$$S_g(40\ MeV) \cdot \sigma_g^{\max} = 48.4\ mb,$$
$$S_m(40\ MeV) \cdot \sigma_m^{\max} = 11.2\ mb$$

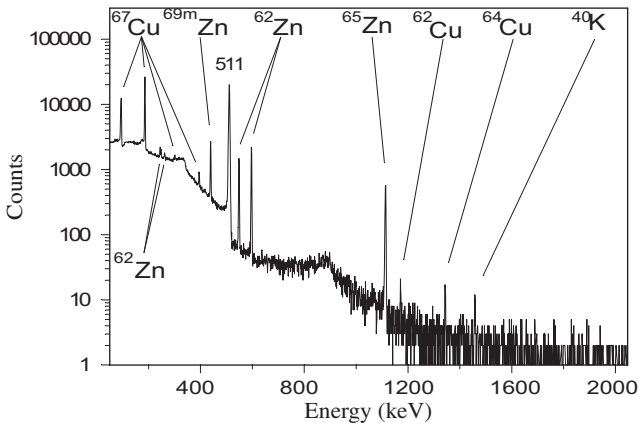

Fig. 1. Gamma-ray spectrum of irradiated zinc (^{40}K-line is a background).