Applied Radiation and Isotopes 70 (2012) 76–80

Contents lists available at SciVerse ScienceDirect

Applied Radiation and Isotopes

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

Feasibility study for production of I-131 radioisotope using MNSR research reactor

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ARTICLE INFO

Article history:
Received 10 March 2011
Received in revised form
11 August 2011
Accepted 12 August 2011
Available online 23 August 2011

Keywords:
Radioisotopes
Radioactivity
High purity target
Cyclic irradiation
FORTRAN program

ABSTRACT

A feasibility study for ^{131}I production using a Low Power Research Reactor was conducted to predict the yield of ^{131}I by cyclic activation technique. A maximum activity of 5.1 GBq was achieved through simulation using FORTRAN 90, for an irradiation of 6 h. But experimentally only 4 h irradiation could be done, which resulted in an activity of 4.0×10^5 Bq. The discrepancy in the activities was due to the fact that beta decays released during the process could not be considered.

1. Introduction

^{131}I is a radioisotope produced in nuclear reactors by neutron irradiation of tellurium dioxide and during the fissioning of uranium. Its physical half-life is 8.02 days and emits β-particles of various energies, with the maximal β-energy being 606 keV and the mean energy being 191 keV. After the emission of a ß-particle, the ^{131}I atom undergoes further transformation with emission of γ-rays. The major γ-radiation is at 364 keV. These β- and γ-radiations account for 90% of the radiation from ^{131}I. As a result, ^{131}I is a major line of treatment for hyperthyroidism and thyroid cancer.

^{131}I is also used to diagnose abnormal liver function, renal (kidney) blood flow and urinary tract obstruction (*Fact Sheet #33, Division of Environmental Health Office of Radiation Protection, January 2003*).

Radioisotopes are atoms with unstable nuclei, which are the nuclei characterized by excess energy that is available to be imparted either to a newly-created radiation particle within the nucleus, or else to an atomic electron. They undergo radioactive decay, and emit gamma ray(s) and/or subatomic particles (Lilley, 2002, and Bhubaneswar, 1996). The amount of radioactivity produced is proportional to the quantity of the activable element (target), the flux of the bombarding particles and the probability of the occurrence of the desired nuclear reaction (Akaho et al., 2000). Their production involves several interrelated activities such as target fabrication, irradiation in reactor or accelerator, transportation of irradiated target to radioactive laboratory, radiochemical processing or encapsulation in sealed source, quality control and transportation to end users (IAEA-TECDOC-1340, 2003).

Radioisotopes are used in two major ways: for their chemical properties and as sources of radiation. They play important roles in food preservation, agriculture, environment, industry and medicine (Sahoo and Sahoo, 2006). In medicine they are used in imaging because the body treats them in the same way that it treats the nonreactive element, and in therapy to destroy cancer by teletherapy, brachytherapy and radiopharmaceutical therapy (Fromm, 1997). Most diagnostic radionuclides emit gamma rays, while the cell-damaging properties of beta particles are used in therapeutic applications.

At present, radioisotopes are mainly prepared with high flux nuclear reactors, (IAEA, 2000) and accelerators. But low power research reactors can also be used only that the yield is low due to their low neutron flux. For example, the Miniature Neutron Source Reactor (MNSR), which is commissioned in 1995 by Ghana Atomic Energy Commission (GHARR-1), Kwabenya, is a low power research reactor designed for neutron activation analysis,

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doi:10.1016/j.apradiso.2011.08.011

personnel training and production of short and medium-lived radioisotopes for application in industry (Yongchun et al., 1992).

Due to the limited core excess reactivities of GHARR-1 research reactor, it cannot sustain long periods of uninterrupted operations. It operates at maximum thermal flux of 1×10^{12} n cm^{-2} s^{-1} comparably to reactors used for radioisotope production. Therefore, it is mainly used for neutron activation analysis and training. The core is extended by the addition of some thickness of Be plates to compensate for fuel depletion and samarium poisoning and thereby increase the core excess reactivity for the next operation. However, after a certain maximum thickness of the Be shims, when the core excess reactivity is well below 3 mk and the reactor cannot be operated at its maximum flux level for a long period, then there is the need for core replacement. To circumvent or alleviate this problem, the application of cyclic activation analysis with repeated irradiation has been used to enhance the activities for NAA analysis and especially in the production of some medium half lives radioisotopes.

The principles of cyclic activation analysis are well described by Spyrou and Kerr (1979). In cyclic activation, a sample is irradiated for a period of time and then transferred to a detector for counting and the entire process is repeated for a number of cycles until the desired activity is achieved. Tout and Chatt (1981) have discussed the practical application of cyclic activation analysis for short-lived radionuclides.

A theoretical approach for the prediction of thermal neutron fluxes needed for repeated irradiation in order to achieve the same activity used for single irradiation at high flux has been determined by Akaho et al. (2000). Their predictions are used to present theoretical activities obtained by varying neutron flux, irradiation time, decay time and mass of the target used; and the number of cycles (n) to attain maximum activity for the production of ^{131}I. All these variations have been done using a fortran program. The theoretical results obtained are then validated by practical experiments.

2. Materials and methods

2.1. Materials

FORTRAN 90 program is the main tool used for the variation of the various parameters of GHARR-1 reactor. The implicit None Syntax was used to ensure that every variable used in the programming process was declared before it was used. A brief message was used to inform the user of the specific method under consideration. And the parameters that are required from the user are displayed on the screen for the user to enter their values. The Do-End-Do Syntax was used to provide the solution required. The results obtained through the iteration presented were directed to external files using the Open–Close Syntax.

2.2. Method

^{131}I is produced by (n,γ) reaction followed by β^- decay (Lieser, 2001). The irradiation yield is described below.

$$\boldsymbol{Te}^{130}(\boldsymbol{n},\boldsymbol{\gamma})^{131}\xrightarrow{\beta^-}\boldsymbol{I}^{131}$$

In this case the net production rate of nuclide Z (^{131}**I**) is given by:

$$\frac{dN_Z}{dt}=N_X(1-e^{-\lambda_Y t})\phi\sigma_X-N_Z\lambda_Z \tag{1}$$

Where, N_X is the number of radioactive atoms X (^{130}Te), N_Z is the number of radioactive atoms Z formed (^{131}I), t is the irradiation time (s), ϕ is the neutron flux (cm^{-2} s^{-1}), σ_X is the neutron capture cross section of X (cm^{-2}), λ_Y is the decay constant of Y (s^{-1}), λ_Z is the decay constant of Z (s^{-1}).

If the number of target atoms, N_X, can be assumed to remain constant (no considerable target burn-up) and $N_Y=N_Z=0$ at $t=0$ (start of irradiation), the following equation is obtained by the integration of Eq. 1:

$$N_Z=N_x\phi\sigma_x\left[\left(\frac{1-e^{-\lambda_Z^t}}{\lambda_Z}+\frac{e^{-\lambda_Y^t}-e^{-\lambda_Z^t}}{\lambda_Y-\lambda_Z}\right)-e^{-\lambda_Z t_d}+\frac{(1-e^{-\lambda_Y t})(e^{-\lambda_Y t_d}-e^{-\lambda_Z t_d})}{\lambda_Z-\lambda_Y}\right] \tag{2}$$

where t is the time of irradiation and t_d is the decay time after the end of irradiation.

Eq. (2) is the basic equation used for the calculation of yield of the (n,γ) reaction followed by β^- decay. It is obvious that this equation should be modified accordingly in order to account for various effects like e.g. target burn-up, self-shielding, or non-1/v behavior.

As far as there are no significant target burn-up and self shielding, and also the reaction obeys 1/v behavior, the above equation has then been applied with some slight modification due to the reactor used. As a result, the specific activation due to a single radionuclide can be written as

$$\boldsymbol{A}=\boldsymbol{B}\times\boldsymbol{C} \tag{3}$$

where

$$B=I\frac{N_X}{W}m\lambda_Z\frac{\phi\sigma_X}{(\lambda_Z+\phi\sigma_Z)}\times\frac{1}{[(\lambda_Z+\phi\sigma_Z)-(\lambda_Y+\phi\sigma_Y)]}$$

$$C=(\lambda_Z+\phi\sigma_Z)[1-e^{-(\lambda_Y+\phi\sigma_Y)t}]-(\lambda_Y+\phi\sigma_Y)[1-e^{-(\lambda_Z+\phi\sigma_Z)t}]$$

A is the initial activity (Bq), W is the atomic weight of target element (g/mol), t is the irradiation time (s) and m is the amount weighed (g).

After a decay time t_d, the activity becomes

$$A_1=A\times e^{-\lambda_Z t_d} \tag{4}$$

λ_Z is the decay constant of ^{131}I (s^{-1}), t_d is the decay time (s), A_1 is the decay activity (Bq).

We shall define a period as

$$T=t_i+t_d$$

For the second cycle period T, the new activity A_2 is related to the first activity A_1 as follows:

$$A_1+A_1(e^{-\lambda_Z T})=A_1(1+e^{-\lambda_Z T}) \tag{5}$$

The activity after n^{th} counting becomes

$$A_n=A_1\frac{1+e^{-n\lambda_Z T}}{1-e^{-\lambda_Z T}}. \tag{6}$$

3. Results and discussion

3.1. Theoretical results

The data obtained from the simulation using irradiation times of 4 and 6 h and their respective decay times after 4 cycles (4days) at neutron fluxes of 5×10^{11} and 1×10^{12} n cm^{-2} s^{-1} are shown in Figs. 1–5. Figs. 1 and 2 portray the variation of ^{131}I activity of various masses at neutron fluxes of 5×10^{11} and 1×10^{12} n cm^{-2} s^{-1} for an irradiation time of 4 h during each cycle. It is seen that for both fluxes, the activity increases with the amount of the sample weighed and the increment is of factor of two. For example the activity obtained after the irradiation of 1 g of Tellurium dioxide is almost twice of that obtained from 0.5 g of the same sample. The differences in activity obtained at both neutron fluxes for the 4 h irradiation were clearly depicted in Fig. 3.