EVALUATION OF RADIO-NUCLIDES ACCUMULATED IN A TARGET IRRADIATED IN A NUCLEAR REACTOR

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الملخص

تم استنباط وتطوير معادلات لحساب كمية ونشاط النظائر المشعة الناتجة مــن تــشعيع عينات بواسطة النيوترونات في المفاعلات النووية مثل إنتاج نظير اليود–131 فــي مفاعـل تاجوراء في ليبيا وذلك عند تشغيل المفاعل في فترات متقطعة أو مستمرة. أوضــحت النتــائج النظرية أن التشعيع المتقطع لعينات مثل التيليريوم لإنتاج اليود–131 لا يوصى بــه ويفــضل التشعيع المستمر في حالة إنتاج النظائر المشعة الطبية منها خصوصا لأنه في حالــة التــشعيع المتقطع يكون الناتج منخفض والملوثات من النويات غير المرغوب فيها مرتفعــة بالمقارنــة بالتشعيع المستمر.

ABSTRACT

Formulae have been derived and developed to evaluate amounts and activities of individual radio-nuclide being accumulated within a target irradiated in nuclear reactor (similar to Tajoura Nuclear Reactor (TNRR)). Two regimes of reactor operation were considered: (i) continuous reactor operation regime; (ii) interrupted reactor operation regime. The results indicated that the amount of ^{131}I produced from interrupted irradiation of Tellurium target for medical use was low compared with that obtained from continuous irradiation and the amounts of the unwanted containments (^{127m}Te, ^{129m}Te and ^{121}Te) were relatively high. As such, interrupted reactor operation regime is not recommended as it would lead to a contamination of the radio-nuclide of interest.

KEYWORDS: Radio nuclide; Irradiation; Activity; Targets; Medical Use; Nuclear Reactor.

INTRODUCTION

During the past twenty years many nuclear research reactors have become operational and several are in the process of being installed, many of them in developing countries. A research reactor is an essential facility for the production of radioisotopes that would cover the national demand.

One of the most important applications of nuclear reactors and accelerators is the large-scale production of radioisotopes. Short-lived radioisotopes have significant roles as tracers in the fields of chemistry, medicine, biology, industry, etc. Problems associated with scheduling reactor production and rapid transportation of short-lived radioisotopes is so serious that use of radioactivity in routine test procedures is precluded. Accelerators are still required for most neutron deficient isotopes. However, it is advantageous to use reactors instead of accelerators for the following reasons:

- In reactors it is easy to irradiate larger and thicker targets at higher fluxes for longer periods of time.
- Irradiation and arrangement of a target in a reactor is easier than in an accelerator.
- Cost of irradiation is usually considerably lower in a reactor than in an accelerator.

The evaluation of the amounts and activities of individual radio-nuclei and the total fission-product inventory in the reactor core during reactor operation or after shutdown is important from the nuclear engineering point of view.

Despite the numerous theoretical studies of radio-isotope production in reactors due to neutron irradiation, the mechanisms of interrupted irradiation remain far from being fully understood especially in the case of production of an individual radio-isotope in small reactors. Previous studies concentrated on the continuous irradiation [1,2], and using special codes through numerical methods to calculate fission-product inventory in power reactor. Arebi and Abdunabi [3], and Omran, N. [4] developed formulas for particular cases of interrupted irradiation. Great effort has to be devoted to seek and develop a general formula for evaluation of radio-isotopes produced in reactors for interrupted and continuous irradiation. This paper focuses only on aspects related to the production of radioisotopes within research reactors similar to Tajoura Nuclear Research Reactor (TNRR) in Libya.

THEORETICAL ANALYSIS

Most of the radioactive nuclides of importance in isotope techniques are produced artificially by changing a stable isotope of an element found in nature into a radioactive isotope by a nuclear reaction.

Problem Formulation

The rate of concentration of specific nuclei (N_i) during and after reactor operation may be expressed in the general form as;

$$dN_i / dt = \text{Formation rate - Depletion rate - Decay rate} \qquad (1)$$

The nuclide I (mass number A, atomic number Z) can be formed by fission, by neutron capture in the nuclide J (mass number A-1, atomic number Z), and by radioactive (usually beta) decay of the nuclide K (mass number A, atomic number Z-1) .

On the other hand it is depleted by neutron capture and by beta particle emission. The various nuclear processes are represented in Figure (1). The terms on the right hand side of equation (1), at a given location or uniform region, are as follows;

- formation rate = $\gamma_i \, N_f \; \sigma_f \, \emptyset + \; N_j \, \sigma_j \, \emptyset + \; \lambda_k \, N_k$

  where: γ_i is the fission yield of the nuclide (i) expressed as a fraction of the total number of fission in which (i) is produced directly. N_f, N_j and N_k are the nuclear number densities of the fissile species and of the nuclides (J and K), respectively; σ_f is the fission cross section of the fissile material, σ_j is the capture cross section of J, λ_k is decay constant of nuclide K and $\emptyset$ is the neutron flux ($n/cm^2.sec$).

- Depletion rate = $N_i \, \sigma_i \, \emptyset$
- Decay rate = $\lambda_i \, N_i$
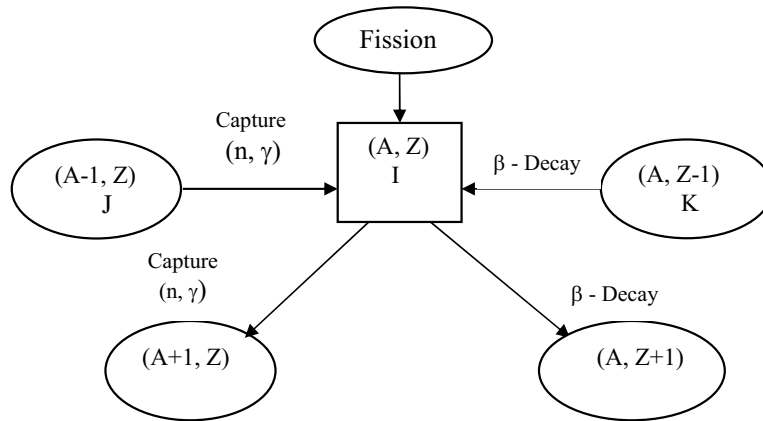
Figure 1: Nuclear processes of the formation and removal of specific nuclei

The significance of these terms is obvious, hence, upon combining a, b and c with equation (1), the result is:

$$dN_i/dt = \gamma_i\ N_f\ \sigma_f\ \emptyset + N_j\ \sigma_j\ \emptyset + \lambda_k\ N_k - N_i\ \sigma_i\ \emptyset - \lambda_i\ N_i \qquad (2)$$

Where: the γ, σ, and λ values may be regarded as known. The term on the left hand side of equation (2) represents the formation rate of (i). Upon rearrangement, this yields;

$$(dN_i/dt +\ N_i\ \sigma_i\ \emptyset + \lambda i\ N_i) = \gamma_i\ N_f\ \sigma_f\ \emptyset + N_j\ \sigma_j\ \emptyset +\ \lambda_k\ N_k \qquad (3)$$

This is a first order linear differential equation. If both sides are multiplied by the integrating factor $e^{(\sigma i\ \emptyset + \lambda i)\ t}$ the left side becomes a complete differential; thus:

$$[(dN_i/dt\ )e^{(\sigma i\ \emptyset +\ \lambda i)\ t} +\ N_i\ (\sigma_i\ \emptyset + \lambda i\ )\ e^{(\sigma i\ \emptyset +\ \lambda i)\ t}] =\ \gamma_i\ N_f\ \sigma_f\ \emptyset\ e^{(\sigma i\ \emptyset +\ \lambda i)\ t} +$$

$$N_j\sigma_j\ \emptyset\ e^{(\sigma i\ \emptyset +\ \lambda i)\ t} + \lambda_k\ N_k\ e^{(\sigma i\ \emptyset +\ \lambda i)\ t} \qquad (4)$$

So that

$$d/dt\ (N_i\ e^{(\sigma i\ \emptyset +\ \lambda i)\ t}) = \gamma_i\ N_f\ \sigma_f\ \emptyset\ e^{(\sigma i\ \emptyset +\ \lambda i)\ t} + N_j\ \sigma_j\ \emptyset\ e^{(\sigma i\ \emptyset +\ \lambda i)\ t} + \lambda_k\ N_k\ e^{(\sigma i\ \emptyset +\ \lambda i)\ t} \qquad (5)$$

The integration of equation (5) results in the following equation:

$$\int d(N_i\ e^{(\sigma i\ \emptyset +\ \lambda i)\ t}) = \int \gamma_i\ N_f\sigma_f\ \emptyset\ e^{(\sigma i\ \emptyset +\ \lambda i)\ t}\ dt + \int N_j\sigma_j\ \emptyset\ e^{(\sigma i\ \emptyset +\ \lambda i)\ t}\ dt + \int\ \lambda_k N_k e^{(\sigma i\ \emptyset +\ \lambda i)\ t}\ dt \qquad (6)$$

This integration can be performed when a certain scheme of radionuclide accumulation is known. The integration constant can be found using the initial conditions.

Continuous irradiation

Suppose the number of atoms of the target (element) decreases due to the neutron capture (n,γ) and β-decay from N_{01} to N_1. Then at any time (t_{irr}) during or immediately after end of irradiation, N_1 can be calculated as follow:

$$N_1 = N_{01} \times e^{-(\Lambda_1 t_{irr})} \tag{7}$$

Where:

$N_{01} = 6.023 * 10^{23} * w*f /A$, f is fraction of the element in mixture; w is weight of target and A is atomic weight of nuclei of the target. $\Lambda_1 = \lambda_1 + \emptyset \, \sigma_1$, λ_1 is decay constant of element sec^{-1}, $\emptyset$ is neutron flux cm^{-2} sec^{-1} and σ_1 is the microscopic absorption cross section of the element cm^2.

Production of radioactive isotopes

Different types of nuclear reactions are utilized for production of radioactive isotopes of a large number of elements. The majority of these reactions are induced by neutron irradiations, but for specific purposes other reactions are more suited or exclusive.

The first daughter product

In case, where the radio nuclides accumulation and decay are subjected to the following scheme;

$$N_1(n,\gamma) \xrightarrow{\sigma_1} N_2 \xrightarrow{\sigma_2}$$
$$\downarrow \lambda_1 \qquad\qquad \downarrow \lambda_2$$
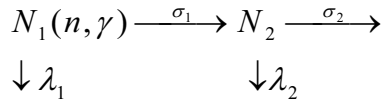
Where: N_2 is the atom number density of the daughter product, it is produced due to interaction of N_1 with neutron and depleted due to decay and interaction with neutrons, then N_2 can be calculated at any time during or immediately after the end of irradiation time (t_{irr}) using equation (6) which reduces to:

$$\int d(N_i e^{(\Lambda_i t)}) = \int N_j \, \sigma_j \, \emptyset \, e^{(\Lambda_i t)} \, dt \tag{8}$$

where i=2, j =1 and $N_i = N_2$, $N_j = N_1$, $N_{i\,0} = N_{02} = 0$, $N_{j0} = N_{01}$, $\Lambda_i = \lambda_i + \sigma_i \Phi$, $\Lambda_i = \lambda_2$ decay constant, $\sigma_i = \sigma_2$ microscopic cross section of daughter product.

Substitution of $N_j = N_{0j} \, e^{-(\Lambda_j t)}$ in equation (8), where $\Lambda_j = \lambda_j + \sigma_j\Phi$ and $N_{j0} = N_{01}$ at t = 0 equation (8) becomes:

$$N_2 = \frac{\Phi \, \sigma_1 N_{01}}{(\Lambda_2 - \Lambda_1)} \left[e^{-\Lambda_1 t_{irr}} - e^{-\Lambda_2 t_{irr}} \right] \tag{9}$$

Where t_{irr} is the irradiation time.
The radio nuclides activity in the target at any moment after irradiation in the reactor is,

$$A_2 = \lambda_2 . N_2$$

Time of maximum activity

The time of maximum activity (t_{max}) of the first daughter product occurs when $dN/dt_{irr} = 0$, from equation (9), one can get;

$$t_{max} = \ln (\Lambda_{2/} \Lambda_1) / (\Lambda_2 - \Lambda_1) \tag{10}$$

From equation(10), one can determine the time of irradiation which gives the maximum activity for a given neutron flux. Also for economic reasons, the time of irradiation must be less than or equal to t_{max}.

The secondary daughter product

In case the daughter radio nuclides (N_3) are formed, and decay according to the following scheme;

$$N_1 (n, \gamma) \xrightarrow{\sigma_1} N_2 \xrightarrow{\sigma_2}$$
$$\downarrow \lambda_1 \qquad\qquad \downarrow \lambda_2$$
$$N_3$$
$$\downarrow \lambda_3$$
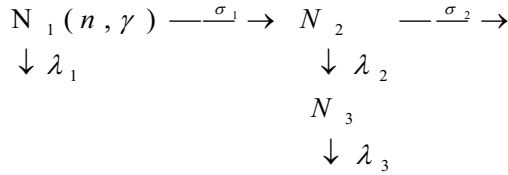
The number of atoms of this secondary product at any time during or immediately after the end of irradiation can be calculated using equations (6) and by substitution $N_i = N_3$, $N_k = N_2$; equation (6) becomes:

$$N_3 = \Lambda_1 \Lambda_2 N_{01} \left[\frac{e^{-\Lambda_1 t_{irr}}}{(\Lambda_2 - \Lambda_1)(\Lambda_3 - \Lambda_1)} + \frac{e^{-\Lambda_2 t_{irr}}}{(\Lambda_1 - \Lambda_2)(\Lambda_3 - \Lambda_2)} + \frac{e^{-\Lambda_3 t_{irr}}}{(\Lambda_1 - \Lambda_3)(\Lambda_2 - \Lambda_3)} \right] \tag{11}$$

Where; $\Lambda_1 = \sigma_1 \emptyset + \lambda_1$, $\Lambda_2 = \sigma_2 \emptyset + \lambda_2$, $\Lambda_3 = \sigma_3 \emptyset + \lambda_3$ and t_{irr} is the irradiation time.

The general case for continuous irradiation

The number of atoms of any product (N_n) at any time (t_{irr}) during or immediately after the end of irradiation can be calculated using the following formula which is comparable with equation (11) as:

$$N_n = \Lambda_1 \Lambda_2^* \dots\dots\dots\dots \Lambda_{n-1}^* N_{01} \sum_1^n C_i e^{-\Lambda_i t} \tag{12}$$

$$C_i = \Pi_{j=1}(\Lambda_j - \Lambda_i)^{-1} \qquad (j=1, j \neq i)$$

Where; $\Lambda = \sigma \emptyset + \lambda$, $\Lambda^* = \sigma^* \emptyset + \lambda^*$, Λ_i refers to disappearance constant of nuclei i, Λ^* is partial formation constant, σ^*, λ^* refers to partial reaction cross section and decay constant of nuclide (i+1).

Interrupted irradiation

In the case of interrupted irradiation as when the reactor is stopped at night and over the weekend. The authors have developed general formulas to calculate the activity of the first daughter product (N_2) and the second daughter product (N_3) for any

scheme of interrupted irradiation. In general, the formulas consider all the intervals of irradiation and decay (cooling) time. Starting from the first irradiation, the fraction of the initial number of nuclei (or activity) remaining after the end of cooling time is calculated, then a summation is made for the total number of nuclei (or activities) accumulated after the end of interrupted irradiation and cooling time.

(1) the activity of first daughter product (N_2) may be calculated as:

$$A_2 = \phi \sigma_1 N_{01} (1 - e^{-\lambda_2 t_{irr}}) \left[\sum_{n=1}^{n=m_1} e^{-\lambda_2 (t_1 - (n-1)t_2)} + \sum_{n=m_1+1}^{n=m_2} e^{-\lambda_2 (t_1 - (n+2)t_2)} + \sum_{n=m_2+1}^{n=m_3} e^{-\lambda_2 (t_1 - (n+5)t_2)} + \sum_{n=m_3+1}^{n=m_4} e^{-\lambda_2 (t_1 - (n+8)t_2)} \right] \tag{13}$$

where:
$m_1 = k_1$; $m_2 = k_1 + k_2$; $m_3 = k_1 + k_2 + k_3$; $m_4 = k_1 + k_2 + k_3 + k_4$.

    k_1 is the number of irradiation days for the first week,
    k_2 is the number of irradiation days for the second week,
    k_3 is the number of irradiation days for the third week,
    k_4 is the number of irradiation days for the fourth week,
    t_1 is the total time after the end of first irradiation of target including cooling and irradiation time (hr), t_2 is the time of one day ,including a night (24 hr).

(2) For secondary daughter product (N_3), the activity may be calculated as:

$$A_3 = \sum_{n=1}^{m_1} \left(A_{01}^d e^{-\lambda_3 (t_1 - (n-1)t_2)} + B e^{-\lambda_2 (t_1 - (n-1)t_2)} \left[e^{-\lambda_2 (t_1 - (n-1)t_2)} - e^{-\lambda_3 (t_1 - (n-1)t_2)} \right] \right)$$

$$+ \sum_{n=m_1+1}^{m_2} \left(A_{01}^d e^{-\lambda_3 (t_1 - (n+2)t_2)} + B e^{-\lambda_2 (t_1 - (n+2)t_2)} \left[e^{-\lambda_2 (t_1 - (n+2)t_2)} - e^{-\lambda_3 (t_1 - (n+2)t_2)} \right] \right)$$

$$+ \sum_{n=m_2+1}^{m_3} \left(A_{01}^d e^{-\lambda_3 (t_1 - (n+5)t_2)} + B e^{-\lambda_2 (t_1 - (n+5)t_2)} \left[e^{-\lambda_2 (t_1 - (n+5)t_2)} - e^{-\lambda_3 (t_1 - (n+5)t_2)} \right] \right) \tag{14}$$

$$+ \sum_{n=m_3+1}^{m_4} \left(A_{01}^d e^{-\lambda_3 (t_1 - (n+8)t_2)} + B e^{-\lambda_2 (t_1 - (n+8)t_2)} \left[e^{-\lambda_2 (t_1 - (n+8)t_2)} - e^{-\lambda_3 (t_1 - (n+8)t_2)} \right] \right)$$

where

$$A_{01}^d = \phi \sigma_1 N_{01} \left(1 - \frac{\lambda_3 e^{-\lambda_2 t_{irr}} - \lambda_2 e^{-\lambda_3 t_{irr}}}{(\lambda_3 - \lambda_2)} \right); \quad B = \frac{\phi \sigma_1 N_{01} (1 - e^{-\lambda_2 t_{irr}})}{(\frac{\lambda_3}{\lambda_2} - 1)}$$

and m's value are the same as in equation 13 above.

RESULTS AND DISCUSSIONS

Computer programs have been written to implement the equations discussed above. The computer programs calculate the following;
(1) The activity of first product. (2) The activity of secondary product. (3) The irradiation time to get maximum amount of product for a given neutron flux. (4) The accumulated radio-nuclides in the target after irradiation in the reactor.

Production of iodine-131 (^{131}I)

The accumulation of the radio nuclides in a target irradiated in reactor for continuous and interrupted irradiation regime have been calculated for tellurium target (1gm of TeO_2) in order to get ^{131}I, which is used for pharmaceutical purposes. The tellurium target isotope contents and its radio-nuclides products due to neutron irradiation are presented in Table (1) [4].

On the basis of equation (9) to (14), the calculation of the accumulations of ^{131}I in irradiated tellurium oxide target has been evaluated for an uninterrupted reactor operation and for long reactor operation period and its interruption schedule. The results of the calculations are shown in Figures (2) to (5). The mass of the target (tellurium oxide (TeO_2) is 1gm, flux of the reactor ($\emptyset = 10^{13}$ n/cm^2.sec), and the reactor was operating four days a week and 10 hours a day in case of interrupted operation.

Figure (2) shows the activity of ^{131}I versus time for interrupted and uninterrupted reactor operation. It can be seen, that the amount of activities produced in case of uninterrupted irradiation is much higher than that for the case of interrupted regime, Moreover, in case of the interrupted operation, the activity stays low with respect to irradiation time and may never reach the desired amount of activity. As such, the interrupted regime of operation is not recommended for production of ^{131}I

Figure 2: The activity of ^{131}I as a function of irradiation time (1gm of TeO$_2$ irradiated in nuclear reactor)
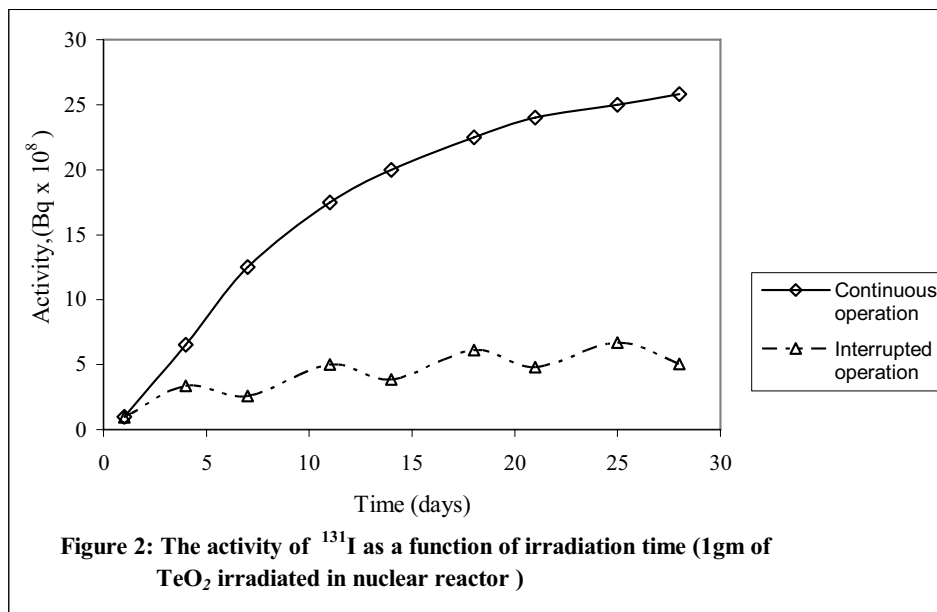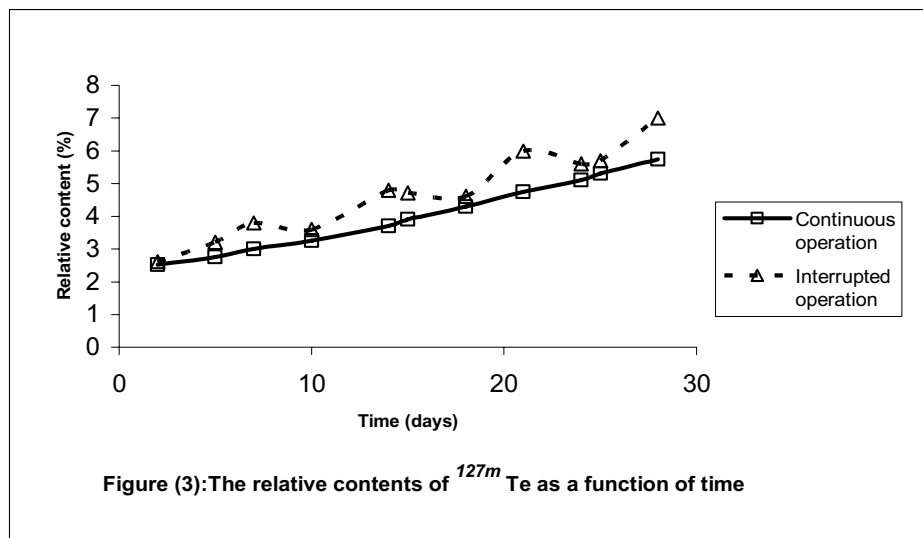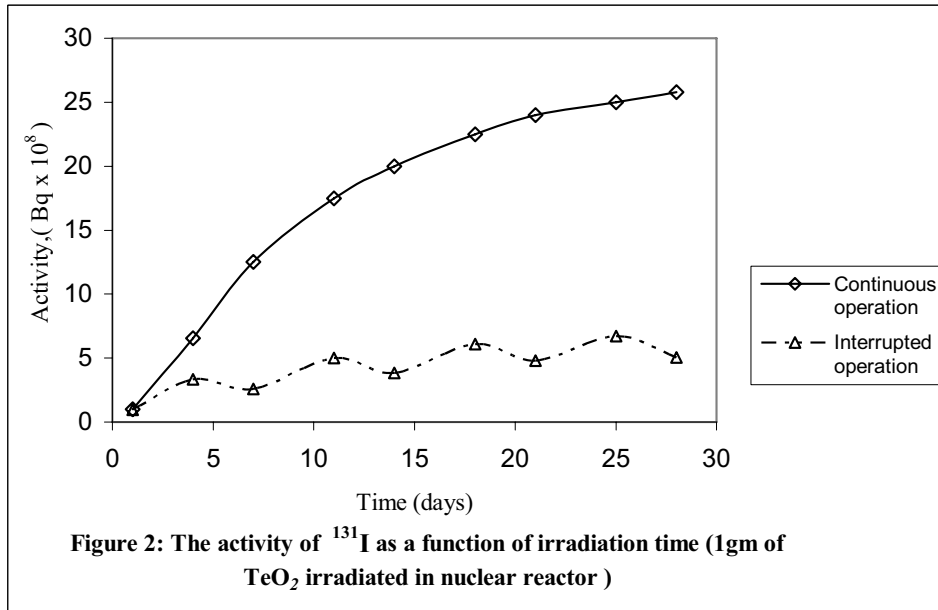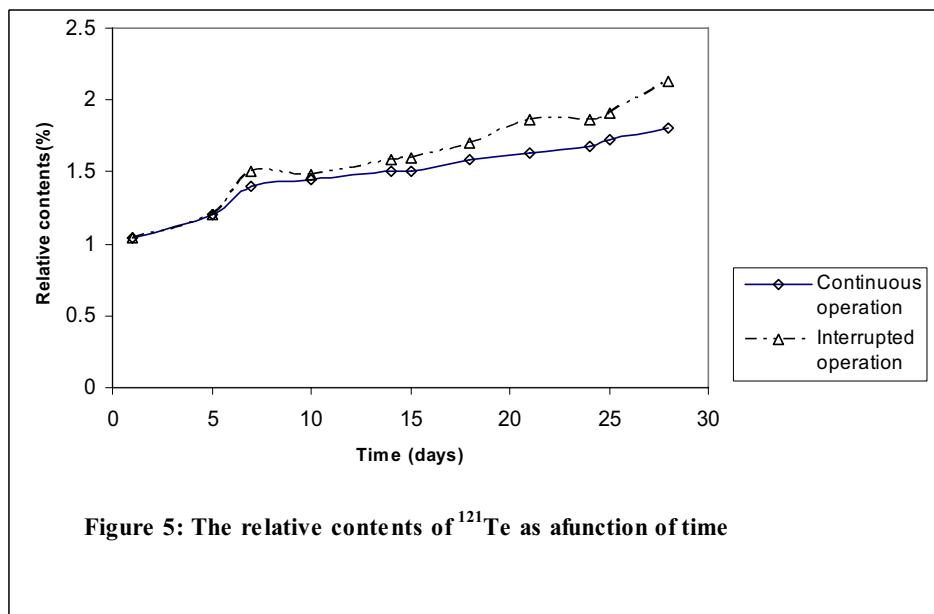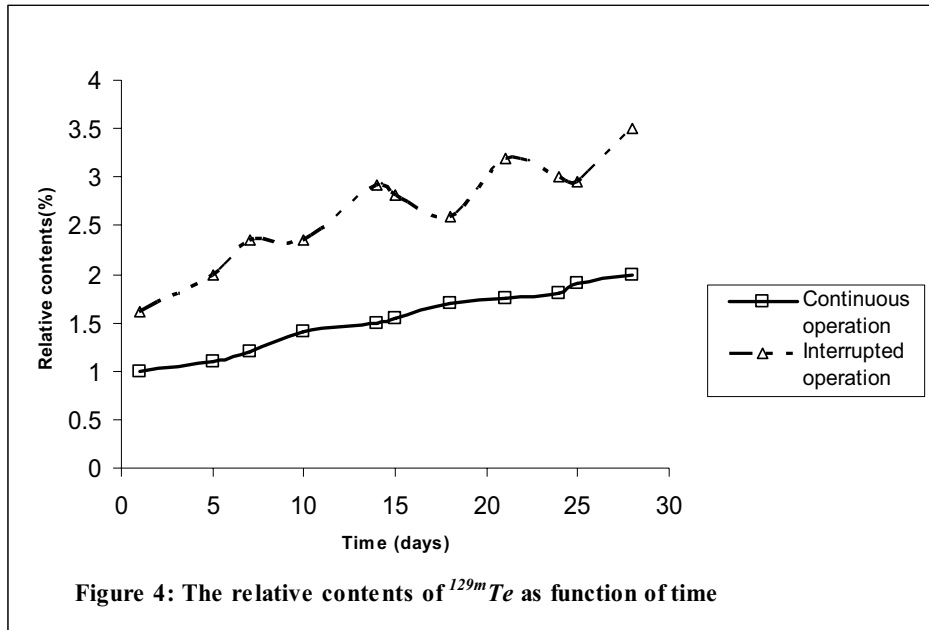
Table 1: isotopes and radio nuclides present in neutron irradiation of tellurium targets

No	Isotope Compositions of target	Abundance (%)	Reaction
1	^{120}Te	0.091	$^{120}Te\ (n,\gamma)^{121}Te \xrightarrow{T_{1/2}\ =\ 16.2d} {}^{121}I$ $\sigma\ =\ 2.0b$ $^{120}Te(n,\gamma)^{121m}Te \xrightarrow{T_{1/2}\ =\ 154d} {}^{121}I$ $\sigma\ =\ 0.3b$
2	^{126}Te	18.7	$^{126}Te(n,\gamma)^{127}Te \xrightarrow{T_{1/2}\ =\ 9.35h} {}^{127}I$ $\sigma\ =\ 0.9\ b$ $^{126}Te(n,\gamma)^{127m}Te \xrightarrow{T_{1/2}\ =\ 109d} {}^{127}I$ $\sigma\ =\ 0.17\ b$
3	^{128}Te	31.7	$^{128}Te(n,\gamma)^{129m}Te \xrightarrow{T_{1/2}\ =\ 335d} I^{129}$ $\sigma\ =\ 0.016\ b$
4	^{130}Te	34.5	$^{130}Te(n,\gamma)^{131}Te \xrightarrow{\beta^-\ T_{1/2}\ =\ 25m}$ $\sigma\ =\ 0.2\ b$ $^{131}I \xrightarrow{T_{1/2}\ =\ 8.04d}$ $^{130}Te(n,\gamma)^{131m}Te \xrightarrow{\beta^-\ T_{1/2}\ =\ 30h}$ $\sigma\ =\ 0.03\ b$ $^{131}I \xrightarrow{T_{1/2}\ =\ 8.04d}$

Figures (3), (4) and (5) present the activity of ^{127m}Te, ^{129m}Te and ^{121}Te radio nuclides accumulated in the target. These radio nuclides emerging in the target are not desired because they may contaminate the radio pharmaceutical preparation in the course of that production. It can also be seen that impurities are higher in case of interrupted irradiation than that for uninterrupted irradiation. This is a second reason for not recommending the interrupted irradiation regime to be the regime of choice for the production of ^{131}I.

Figure 2: The activity of ^{131}I as a function of irradiation time (1gm of TeO$_2$ irradiated in nuclear reactor)

Figure (3):The relative contents of ^{127m}Te as a function of time

Figure 4: The relative contents of ^{129m}Te as function of time

Figure 5: The relative contents of ^{121}Te as afunction of time

For continuous irradiation, the time (t_{max}) required to get maximum activity of ^{131}I accumulated in the target for different neutron fluxes are presented in Table(2).

Table 2: Time for maximum activity vs flux.

Flux (n/cm^2.sec)	10^{12}	10^{13}	10^{14}	10^{15}
Time of maximum activity (days)	178	151	125	98

CONCLUSIONS

It may be concluded that the equations developed in this paper give the possibility of evaluating the accumulation of radio-isotopes in irradiated target for interrupted and uninterrupted reactor operation regimes. The theoretical results show that the production of ^{131}I by interrupted irradiation of Tellurium target is not recommended. Therefore, only the continuous reactor operation regime is recommended for production of radio-pharmaceuticals.

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