Calculation Results for Enhancing Ability of I-131 Radioisotope Production Using Tellurium Dioxide Target on the Dalat Nuclear Research Reactor

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Abstract: The paper presents the calculation results in re-design of neutron trap of the Dalat Nuclear Research Reactor (DNRR) for I-131 radioisotope production using $TeO₂$ target. The new design permits for loading more $TeO₂$ capsules from 9 to 12, 15 and 18 in the neutron trap. The enhancement of radioisotope production is implemented by re-arrangement of the neutron trap without changing the dimension or geometry of irradiation capsules. By using neutronics computer code as MCNP6, calculation results of I-131 activity in 6 investigated cases were obtained to confirm about the new designs of neutron trap to carry out radioisotope production on the DNRR. The re-arrangement of the neutron trap can be used effectively for radioisotope production with thermal neutron flux in average rage from 5.3×10^{12} to 1×10^{13} n/cm².s and the total activity of I-131 isotope is increased from about 19.2% to 38.8% comparing with the original method using 9 capsules. The negative reactivity insertion of the capsules from 0.60 β_{eff} to 0.96 β_{eff} when loading capsules were also met the safety requirements of operational conditions of the DNRR.

Keyword: The DNRR, Radioisotope Production, TeO2, I-131, MCNP6 code.

1. Introduction

Research reactor is a very useful facility for radioisotope production by using neutron to irradiate the target under normal operation. The most convenience of this method is simple in preparation of target capsule and in irradiation at fixed positions inside or outside reactor core. I-131 is an important radioisotope that can be used for cancer diagnostics and treatment and it is also produced easily by using research reactor with neutron irradiation on $TeO₂$ target [1]. Even the DNRR has low power of 500 kWt and average thermal neutron flux is smaller than 5.0×10^{12} n/cm².s, radioisotope production is still applied as a main purpose to exploit the reactor during more than 35 years from 1984 until now [2]. The demand of I-131 isotope utilization in nuclear medicines is increasing so the effective utilization of the DNRR for radioisotope production needs to be considered to satisfy the domestic market [3]. Then, the calculation for enhancement of I-131 radioisotope production was carried out and the experiments will be implemented after having fully in safety analysis for modification of neutron trap structure on the DNRR.

By modifying the neutron trap structure, the enhancement of I-131 isotope production will be increased by loading more capsules from 9 to 12, 15 or 18 and keeping the same geometry and dimension of target capsules. So far, 9 TeO_2 capsules have been irradiated at neutron trap of the reactor with 150 hours continuously. Normally, the average activity of I-131 isotope can be reached to 35 Ci in each reactor operation cycle. Basically, the method to produce I-131 isotope with half-life of about 8.05 hours can be expressed as follows [1]:

$$
^{130}_{52}Te + {}_0n^1 \rightarrow {}^{131}_{52}Te^* + \gamma
$$

 ${}^{31}_{52}Te^* \rightarrow \beta^- + {}^{131}_{53}I$ and the half-life of Te-131 is only 25 minutes.

Fig. 1. The micro cross section of Te-130 isotope [4].

The capture cross section of Te-130 isotope (in **Fig. 1)** in the rage from 0.01 eV to 1000 eV is quite high compared with cross section at higher energy region. The linear of Te-130 neutron capture cross section is in thermal energy range under 0.625 eV. So the main calculation results for the I-131 isotope production are in the thermal neutron energy group. The average of neutron capture reaction of Te-130 is about 67 mili-barns.

I-131 isotope can be chemically separated from the $TeO₂$ target after irradiation by thermal neutron on nuclear reactor as research reactor. By applying this method, we can obtain high specific activity of target under irradiation by high thermal neutron flux of research reactor. Of course, another method can be used on the research reactor to produce I-131 as fission product by irradiation of the U-235 HEU or LEU but the facility chain is very complicated with solid waste treatment and separation of I-131 through fission products as well.

The DNRR is a pool type reactor with nominal power 500 kWt and its main purposes are radioisotope production, neutron activation analysis and training. Recently the demand of radioisotope market especially I-131 isotope increasing so the enhancement of I-131 production on the DNRR has been considered in calculation by using neutronics computer codes as MCNP6 and ORIGEN2 [5, 6]. The detailed calculation about the neutron fluxes inside $TeO₂$ target and reaction rate as well as neutron fluxes on the irradiation capsules were done by MCNP6 code.

The number of loaded $TeO₂$ capsules in the neutron trap after modifying is about 12, 15 and 18 capsules. The re-arrangement of neutron trap with beryllium rods around can be created more new irradiation channels. The disadvantage of this re-arrangement is mainly in reducing the neutron flux and adding more negative reactivity, however the loaded mass of $TeO₂$ increases and I-131 isotope activity increases from about 19,2% to 38.8%.

2. Calculation model and method

From 2014, the neutron trap of the DNRR was modified for loading ability to 9 capsules at neutron trap and the total activity of I-131 after 150 hours reactor operating continuously approximates 35 Ci. **Fig. 2** shows the loading position of 9 TeO₂ capsules at neutron trap of the DNRR currently and the operation time is about 130 to 150 hours continuously.

Fig. 2. The irradiation positions of 9 TeO₂ capsules at neutron trap of the DNRR.

The TeO₂ target material used for I-131 isotope production is in powder with 99.9% purity and the mass of Te-130 isotope is about 27% in weight. The $TeO₂$ target is covered by aluminum container with the dimension of 2.6 cm in diameter with a thickness of 1.5 mm and 20 cm in height with 16 cm height for target. The total volume of target is about 65 cm³ that is equivalent with 200 to 230 gram of $TeO₂$.

By removing the whole neutron trap with beryllium blocks (about 6 half beryllium blocks), the center of the neutron trap was replaced by a beryllium rod to avoid the core has too high power density. Until now, the DNRR was operated about more than 300 full power days with the configuration of 92 LEU fuel assemblies so the power peaking factor is smaller than those at the beginning of operation cycle, and the fuel cladding temperature is lower than those of the fresh core [7]. This means that the operational conditions of the reactor meet the safety requirements.

After modifying the neutron trap, 6 available positions can be loaded by maximum 3 $TeO₂$ capsules in each hole. At the present work, the loading of 12 capsules with and without graphite reflector on the top and bottom, and the loading with 18 capsules was considered in calculation models. The MCNP6 calculation models are shown in **Fig. 3**.

Fig. 3..The arrangement of 12 and 18 TeO_2 capsules at neutron trap of the DNRR.

By re-arrangement of the beryllium rods around neutron trap, 2 new positions for loading 3 capsules each hole can be created. The total 15 capsules can be loaded in the new neutron trap including 9 capsules are still in the old positions. The calculation model by using MCNP6 code is depicted in **Fig. 4**. The re-arrangement of the neutron trap satisfies the safety condition as well as the thermal neutron flux for radioisotope production.

Fig. 4. Loaded positions of 15 TeO₂target capsules in the neutron trap after its re-arrangement with 4 options.

In the new method, it is very simple and easy for carrying out experiments to test and to confirm about the calculation results. Under this re-arrangement, the violation in thermal hydraulics limitation is avoided as calculation results were confirmed in the design of new LEU cores with second candidate core for reactor conversion from HEU to LEU fuel [7].

The calculations were carried out for 6 cases with different loading capsule numbers from 9 to 18. The explanation of loading scheme for detail calculations is described in the **Table. 1**. It is convenient that all case studies were used old capsule and only adding more capsules at neutron trap after its re-arrangement.

Number of	Detailed explanation					
loaded capsules						
	Original neutron trap using 9 capsules currently (Fig. 2) with 3 layers					
	and in each layer 3 capsules can be loaded.					
$12 - Normal$	All capsules located at second ring with beryllium rod at center (Fig. 3).					
	Optimizing axial direction of thermal neutron flux.					

Table 1. Calculation cases for enhancement of isotope production on the DNRR.

3. MCNP code for neutron fluxes and reaction rate calculation

The MCNP6 code was used for neutron flux and reaction rate calculation for $TeO₂$ target in capsules. The calculation models of the DNRR together with target capsules were described as detail as possible in **Fig. 2**, **Fig. 3** and **Fig. 4**. The ENDF/B7.1, JEFF3.3.1 [8] libraries were used for calculation. The *kcode* option of MCNP6 code was chosen for the calculation with positions of control rods were set nearly critical status. The standard deviation of *keff* in the calculation was lower than 8.0×10^{-5} with 160 active cycles and 1 million particles in each calculation cycle. The errors of obtained neutron flux as well as reaction rates were smaller than 0.3%. The burn-up calculations of the DNRR were mainly at two points: after 300 full power days and 120 full power days.

Neutron fluxes and reaction rates of Te-130 reactions and multiplication factor were calculated by using the MCNP6 code and three group neutron fluxes ϕ in active cell are provided by tally *F4*:

$$
\phi_m = \int \phi_m(E) dE \tag{1}
$$

Neutron fluxes have to be normalized to get absolute value of neutron flux Φ under thermal power of reactor. The absolute neutron flux can be obtained as follows [14]:

$$
\Phi_m = \frac{P\overline{v}}{1.6022 \times 10^{-13} w_f} \frac{1}{k_{\text{eff}}} \phi_m \tag{2}
$$

where *P* is power of reactor (MW), \overline{v} is the average number of neutrons released per fission, w_f is effective energy released per fission (~ 193.7 MeV/fission to VVR-M2 LEU fuel type) and *keff* is the effective multiplication factor.

Actually, getting data including neutron flux, reaction rates and effective multiplication factor from MCTAL file is easier than reading from output file of MCNP6 code. Reaction rates *R* are received in tally as:

$$
R_{mnx} = \int \sigma_x^n(E)\phi_m(E)dE
$$
 (3)

where R_{mnx} is reaction rate type *x* of nuclide *n* in cell *m*, micro cross section has unit barns.

4. I-131 activity calculation after its irradiation and cooling

The number density of I-131 can be calculated by equation [9]:

$$
\frac{dN(^{131}_{52}Te)}{dt} = \sigma(^{130}_{52}Te)\varphi N(^{130}_{52}Te) - \lambda(^{131}_{52}Te)N(^{131}_{52}Te) \tag{4}
$$

$$
\frac{dN(^{131}_{53}I)}{dt} = \lambda(^{131}_{52}Te)N(^{131}_{52}Te) - \lambda(^{131}_{53}I)N(^{131}_{53}I) \tag{5}
$$

We can interfere that

$$
\frac{dN(^{131}_{53}I)}{dt} = \sigma(^{130}_{52}Te)\varphi N(^{130}_{52}Te)\left(1 - e^{-\lambda(^{131}_{52}Te)t}\right) - \lambda(^{131}_{53}I)N(^{131}_{53}I) \tag{6}
$$

with σ is micro cross section (barn); φ is neutron flux (n/cm².s); λ is decay constant (s⁻¹) and *N* is the number density of isotopes (atoms/cm³).

The equation can be solved by using numerical method or direct calculation after integrating the equation (6). Then, the number density of I-131 can be obtained as follow:

$$
N(^{131}_{53}I) = \sigma(^{130}_{52}Te)\varphi N(^{130}_{52}Te) \left(\frac{1 - e^{-\lambda(^{131}_{53}I)t_{irr}}}{\lambda(^{131}_{53}I)} + \frac{e^{-\lambda(^{131}_{52}Te)t_{irr}} - e^{-\lambda(^{131}_{53}I)t_{irr}}}{\lambda(^{131}_{52}Te) - \lambda(^{131}_{53}I)} \right) e^{-\lambda(^{131}_{53}I)t_{decay} + \frac{\left(1 - e^{-\lambda(^{131}_{52}Te)t_{irr}}\right)\left(e^{-\lambda(^{131}_{52}Te)t_{decay} - e^{-\lambda(^{131}_{53}I)t_{decay}}\right)}{\lambda(^{131}_{53}I) - \lambda(^{131}_{52}Te)}
$$
\n(7)

where *tirr* and *tdecay* are irradiation and decay time (hour) respectively.

In the equation (7), we can separate as two parts with build-up process and decay time as follow:

$$
N(^{131}_{53}I) = \sigma(^{130}_{52}Te)\varphi N(^{130}_{52}Te) \left(\frac{1 - e^{-\lambda(^{131}_{53}I)t_{irr}}}{\lambda(^{131}_{53}I)} + \frac{e^{-\lambda(^{131}_{52}Te)t_{irr}} - e^{-\lambda(^{131}_{53}I)t_{irr}}}{\lambda(^{131}_{52}Te) - \lambda(^{131}_{53}I)} \right)
$$
(8)

$$
N(^{131}_{53}I) = N_0(^{131}_{53}I)e^{-\lambda(^{131}_{53}I)t_{decay}} + \frac{(1 - e^{-\lambda(^{131}_{52}Te)t_{irr}})(e^{-\lambda(^{131}_{52}Te)t_{decay}} - e^{-\lambda(^{131}_{53}I)t_{decay}})}{\lambda(^{131}_{53}I) - \lambda(^{131}_{52}Te)}
$$
(9)

The number density of Te-130 can be calculated by

$$
N(^{130}_{52}Te) = \frac{N_A \cdot m(^{130}_{52}Te)}{M(^{130}_{52}Te)}
$$
\n(10)

with $N_A = 6.023 \cdot 10^{23}$ mol⁻¹ Avogadro number;

 $m(^{130}_{52}Te)$ is mass of Tellurium in grams;

 $M(^{130}_{52}Te)$ is Atomic mass of Tellurium in grams.

The activity *A* by Curie unit of I-131 can be determined by formula:

$$
A(^{131}_{53}I) = \lambda(^{131}_{53}I)N(^{131}_{53}I) \tag{11}
$$

So the activity in each $TeO₂$ target capsule can be determined through the calculation by applying these equations from (4) to (11).

5. Calculation results and discussions

Thermal neutron flux

In the axial direction, the thermal neutron flux distribution of 1cm/node at the original neutron trap without loading target compares with modified for loading 9 capsules (OLD) and the case having 6 irradiation channels (NEW). The profiles of thermal neutron flux in three cases have different in maximum point of 10% and in the average between OLD and NEW type. It is a disadvantage of thermal neutron flux when creating new irradiation channels around neutron trap. The detailed of thermal neutron flux profiles are shown in **Fig. 5**.

Fig. 5. Thermal neutron flux distribution at original neutron trap and modified for loading 9 capsules (OLD) and new irradiation positions (NEW).

Basing on the obtained calculation results, the best distance following the axial direction having highest thermal neutron flux is from 8 to 48 cm at bottom to the top direction. The height is equivalent to the length of 2 capsules with light water or graphite reflector at top and bottom.

When loading $TeO₂$ capsules at different irradiation positions inside the reactor core as in **Table 1**, the average thermal neutron flux inside $TeO₂$ target capsules is described in the **Fig.** 6. The maximum of average thermal neutron flux in loaded 9 capsules is about 1.00×10^{13} n/cm².s while other cases just in 8.8×10^{12} , 8.3×10^{12} and 8.0×10^{12} with loaded 15, 18 and 12 capsules respectively. The micro cross section in barn unit of Te-130 isotope in thermal energy is about 1.49E-01 barn.

Fig. 6. The average thermal neutron flux in each capsule of 6 calculated cases.

The thermal neutron flux inside all capsules is high enough from 5.3×10^{12} n/cm².s to 1.0×10^{13} n/cm².s at top and middle position at irradiation channel. The detail axial distribution of each node 0.78 cm/node inside 3 TeO₂ target capsules from bottom to the top is depicted in the **Fig. 7**.

Fig. 7. Relative thermal neutron flux of 3 capsules at each node in 1 irradiation channel from bottom to the top.

Negative reactivity and safety

The negative reactivity insertion when loading capsules was also estimated by comparing with the core configuration without capsules. Detailed negative reactivity insertion for different loading number of capsules is depicted on the **Table 2** with the difference of $TeO₂$ mass in the capsules. The maximum of the negative reactivity insertion depends on the mass of loaded targets and it will be increased when increasing mass of Te-130.

Number of capsule	Negative reactivity (cents)		
	$(60.2 - 70.2)$		
12 normal	$(69.2 - 79.2)$		
12 with graphite	$(63.4 - 73.4)$		
15 long	$(83.8 - 93.8)$		
15 wide	$(81.4 - 91.4)$		
	$(86.5 - 96.5)$		

Table 3. Negative reactivity insertion with different mass of Te-130 from 54 to 62 gram in each $TeO₂$ target capsule.

The range of negative reactivity is met requirements for the reactor safety in normal operation. In thermal hydraulics, fuel cladding temperature is a parameter needed to be evaluated to assure about the nuclear safety. As calculation results in the design of new LEU cores of the DNRR, the maximum temperature of the hottest channels in 2 core configurations is lower than permission temperature of 103 °C . So the re-arrangement of neutron trap in 5 investigated cases will also satisfy the limit of fuel cladding temperature after operating 8 years.

Activity of I-131 isotope in 6 investigated cases

The main effects of irradiation for radioisotope production include thermal neutron flux, mass of target and irradiation time or decay time. With different neutron flux range from

5.3E+12 to 1.0E+13 n/cm².s, irradiation time from 24 to 150 hours and the same target mass of 62 gram Te-130, calculation results are shown in **Table 2** and **Fig. 8**. The increasing of neutron flux is proportional with I-131 activity in the scale 1 by 1. So the improvement of thermal neutron flux is a way to enhancement for radioisotope production. In case of the DNRR, the method to increase neutron flux is difficult because of its low power and design especially using fuel with low power density. If upgrading power of the DNRR from 0.5 MW to 0.6 or 0.8 MW the expected thermal neutron flux can be increased 16 to 35% respectively. Because the irradiation positions in axial direction of 3 capsules in 1 channel have different thermal neutron flux, the archived activity of I-131 after 150-hour irradiation of current 9 capsules have the ratio as 1.00:1.20:1.54 as top, bottom and middle positions of target.

Irradiation	Activity of I-131 (Ci) with different thermal neutron flux $(n/cm2.s)$							
time (hr.)	$F=5.3E+12$	$F=5.5E+12$	$F=6.5E+12$	$F=6.8E+12$	$F=7.8E+12$	$F = 8.5E + 12$	$F=1.0E+13$	
24	0.51	0.53	0.62	0.65	0.75	0.81	0.96	
48	0.99	1.02	1.21	1.26	1.45	1.58	1.86	
72	1.42	1.48	1.75	1.83	2.10	2.28	2.69	
96	1.83	1.90	2.24	2.34	2.69	2.93	3.45	
120	2.20	2.28	2.69	2.82	3.23	3.52	4.14	
144	2.54	2.63	3.11	3.25	3.73	4.07	4.78	
150	2.62	2.71	3.21	3.36	3.85	4.19	4.93	

Table 2. Calculation results of I-131 activity (Ci) at different thermal neutron flux and the mass of Te-130 is about 62 gram in a capsule.

Fig. 8. Activity of I-131 using about 62 gram TeO₂ target capsule with different thermal neutron flux.

The calculation results of I-131 activity with different target mass in 6 investigated cases are described in **Table 3** and **Fig. 9**. The irradiation condition includes 150-hour irradiation time, target mass of from 54 to 62 gram of Te-130. The obtained results show that the total activity of I-131 in new methods comparing with old method using 9 capsules is increased from 19.24 to

38.77%. It is satisfied the requirements for enhancement of radioisotope production on the DNRR.

$\boldsymbol{9}$				12-graphite		
After 150	Decay 18	After 150	Decay 18	After 150	Decay 18	
hrs	hrs	hrs	hrs	hrs	hrs.	
31.23	29.27	38.67	36.25	39.47	37.00	
32.79	30.74	40.60	38.06	41.45	38.85	
34.35	32.20	42.53	39.87	43.42	40.70	
35.91	33.66	44.47	41.69	45.39	42.56	
		19.24		20.89		
15 -Long		15-Wide		18		
After 150	Decay 18	After 150	Decay 18	After 150	Decay 18	
hrs.	hrs.	hrs.	hrs.	hrs.	hrs.	
44.95	42.14	43.86	41.12	51.00	47.81	
47.20	44.24	46.05	43.17	53.55	50.20	
49.44	46.35	48.25	45.23	56.10	52.59	
51.69	48.46	50.44	47.29	58.65	54.98	
		28.81		38.77		
		30.53		12-normal		

Table 3. Calculation results of I-131 activity of 6 cases with different mass of Te-130.

Fig. 9. Total activity of I-131 with different Te-130 mass and under 150-hour irradiation.

In 5 new considered cases, the simplest way is re-arrangement of neutron trap with 3 options to load more 6 capsules in long sharp of the reactor core. These cases are satisfied all conditions

related to reactor safety in operation limit condition. The carrying out to arrange of 12 beryllium rods around neutron trap together with adding 2 irradiation channels can be easily conducted. The new core configuration still keeps a neutron trap and using 92 LEU fuel assemblies and 12 beryllium rods. Basically, in the future all the core configurations for loading 15 capsules will be considered in detail of neutronics and thermal hydraulics as well as safety analysis to solve the problem for enhancement in radioisotope production on the DNRR.

6. Conclusions

Based on the calculation results, we can have a conclusion that:

- All 5 cases for plan to enhance I-131 radioisotope production on the DNRR can be carried out easily and meet the safety requirements in operation of the DNRR. However, the new methods by re-arrangement of beryllium rods around neutron trap to create 2 irradiation channels are highly considered because of safety, easy implementation and effective in radioisotope production of I-131.
- The calculation is a very important step before carrying out testing one operation cycle with new design for changing neutron trap to satisfy safety of the normal operation condition of the DNRR.
- Increasing I-131 product and effective operation time as well as using economical fuel are the main purposes of the DNRR utilization and application.
- The calculation results can be used for the new research reactor for radioisotope production especially of I-131 isotope.

Future tasks need to be done:

- Carrying out experiments to measure the neutron flux distribution in the neutron trap.
- Testing one operation cycle for radioisotope production with new arrangement of neutron trap.
- Preparing safety analysis report to submit VARANS for approval of new methods to enhancement of radioisotope production on the DNRR.

Acknowledgement

The authors would like to express special thanks for the helpful discussions and willing carrying out experiments on the DNRR of the staff in Reactor Physics and Engineering Department, Reactor Center. We also appreciate about finance support, valuable helping and administrative process of Ministry of Science and Technology and VINATOM for implementation of the research project "*Study on effective, economic in using fuel and enhancement of radioisotope production on the Dalat Nuclear Research Reactor*".

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Kết quả tính toán nâng cao khả năng sản xuất đồng vị I-131 từ bia TeO² trên Lò phản ứng hạt nhân Đà Lạt

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Tóm tắt: Bài báo trình bày kết quả tính toán thiết kế lại bẫy neutron của Lò phản ứng hạt nhân Đà Lạt để sản xuất đồng vị I-131 bằng bia TeO₂. Thiết kế mới của bẫy neutron cho phép nạp tải số lượng capsule TeO₂ từ 9 lên 12, 15 và 18. Việc tăng cường sản xuất đồng vị phóng xạ được thực hiện bằng cách sắp xếp lại bẫy neutron để tăng nạp tải số lượng capsule mà không cần thay đổi về kích thước hình học của chúng. Bằng việc sử dụng chương trình tính toán như MCNP6, các kết quả tính toán hoạt độ đồng vị I-131 trong cả 6 trường hợp được xác định để khẳng định về những thiết kế mới của bẫy neutron cho việc thực hiện sản xuất đồng vị hiệu quả trên Lò Đà Lạt. Việc sắp xếp lại bẫy neutron được sử dụng hiệu quả cho sản xuất đồng vị phóng xạ với thông lượng neutron nhiệt trung bình từ 5.3×10^{12} đến 1.0×10^{13} n/cm².s và hoạt độ tổng của I-131 tăng từ khoảng 19,2% đến 38,8% so với phương pháp cũ dùng 9 capsule. Độ phản ứng âm đưa vào của các capsule có giá trị từ 0,60 β_{eff} đến 0,96 β_{eff} khi nạp tải các capsule chiếu mẫu là đáp ứng yêu cầu các điều kiện vận hành an toàn của Lò Đà Lạt.

Từ khóa: LPƯHNĐL, Sản xuất đồng vị phóng xạ, TeO2, I-131, chương trình MCNP6.