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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 TeO_2 target. The new design permits for loading more TeO_2 capsules from 9 to 12, 15 and 18 in the neutron trap. The enhancement of radioisotope production was implemented by re-arrangement of the neutron trap without changing the dimension or geometry of irradiation capsules. By using neutronics computer code as MCNP6, the obtained calculation results of I-131 activity in 6 investigated cases showed that the new design by the re-arrangement of the neutron trap can be used effectively for radioisotope production with thermal neutron flux in average range from 5.3×10^{12} to 1×10^{13} n/cm^2.s and the total activity of I-131 isotope was increased from about 19.2% to 38.8% comparing with the original design using 9 capsules. The negative reactivity insertion was from 0.60 β_{eff} to 0.96 β_{eff} when loading capsules that also met the safety requirements of operational conditions of the DNRR.

Keyword: *The DNRR, Radioisotope Production, TeO_2, I-131, MCNP6 code.*

I. 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 capsules and in their irradiation at fixed positions inside or outside reactor core. I-131 isotope 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 TeO_2 target [1]. Even the DNRR has low power of 500 kWt and average thermal neutron flux is smaller than 5.0×10^{12} n/cm^2.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 medicine is increasing so the effective utilization of the DNRR for radioisotope production needs to be considered to satisfy the domestic market [3]. Therefore, the design calculation for enhancement of I-131 radioisotope production was carried out.

By modifying the neutron trap structure, the activity of I-131 radioisotope was increased by loading up to18 target capsules but keeping their geometry and dimension. In original design, 9 TeO_2 capsules have been irradiated at neutron trap of the reactor core with 150 hours continuously, and 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 days can be expressed as follows [1]:

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

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

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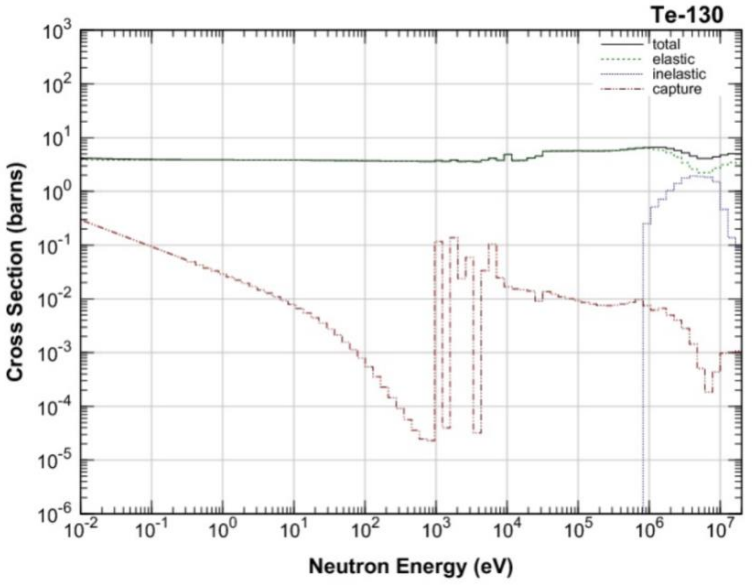

Fig. 1. The micro cross section of Te-130 isotope [4]

The neutron capture cross section of Te-130 isotope (see in Fig. 1) in the range from 0.01 eV to 1000 eV is quite high compared with cross section at higher energy region. The linear of Te-130 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 cross section of neutron capture reaction of Te-130 is about 67 mili-barns in this region.

I-131 isotope can be chemically separated from the TeO_2 target after irradiation by thermal neutron on research reactor. By this method, high specific activity of target can be obtained by irradiation with high thermal neutron flux. 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 of 500 kWt and its main purposes are radioisotope production, neutron activation analysis and training. Recently the demand of radioisotope market, especially I-131 isotope is increasing so the enhancement of I-131 production on the DNRR has been considered in calculation by using neutronics computer codes as MCNP6 [5] and ORIGEN2.1 [6]. The detailed calculation about the neutron fluxes inside TeO_2 target and reaction rate as well as neutron fluxes on the irradiation capsules were done by MCNP6 code.

The number of loaded TeO_2 capsules in the neutron trap after modifying is about 12, 15 and 18 capsules. The rearrangement of neutron trap with beryllium rods around can be created more new irradiation channels. The disadvantage of this rearrangement is mainly in reducing the neutron flux and adding more negative reactivity, however the loaded mass of TeO_2 increases and I-131 isotope activity increases from about 19.2% to 38.8%.

II. CALCULATION MODEL AND METHOD

A. Calculation model

Since 2014, the neutron trap of the DNRR was modified for loading up to 9 capsules and the total activity of I-131 after 150-hour reactor operating continuously approximated 35 Ci. Fig. 2 shows the loading positions of 9 TeO_2 capsules at neutron trap.

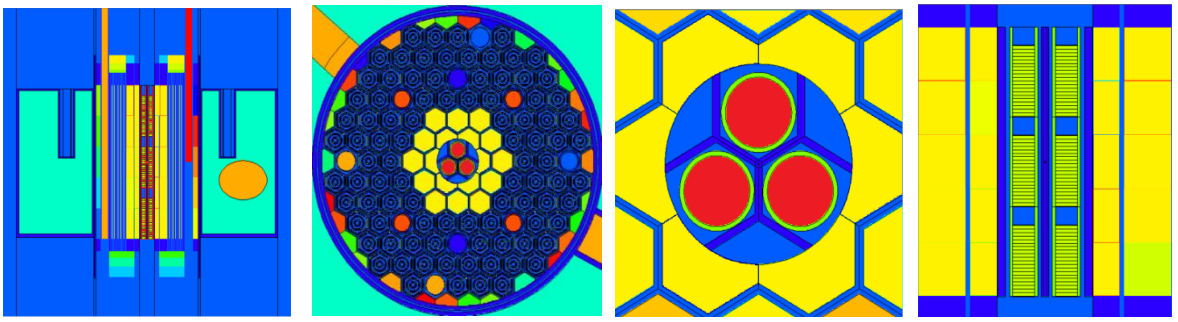

Fig. 2. The irradiation positions of 9 TeO_2 capsules at neutron trap of the DNRR.

The TeO_2 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 TeO_2 target of 16 cm height is covered by aluminum container with the dimension of 2.6 cm in diameter, 1.5 mm in thickness and 20 cm in height. The total volume of target is about 65 cm^3 that is equivalent with 200 to 230 gram of TeO_2.

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 safety requirements.

After modifying the neutron trap, 6 available positions can be loaded by maximum 3 TeO_2 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.

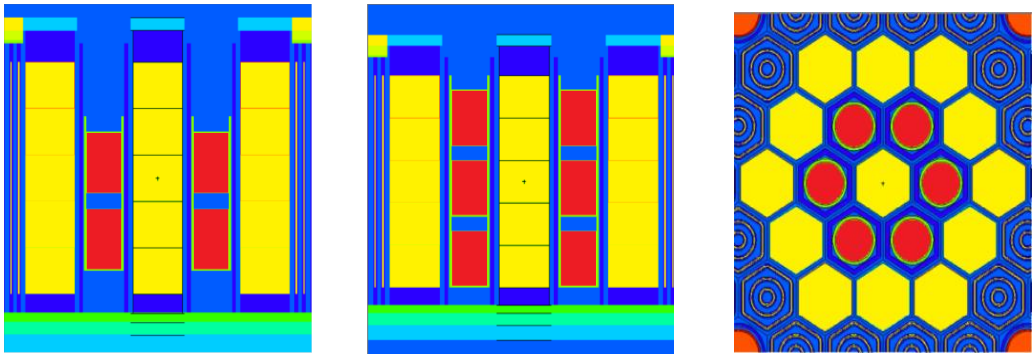

Fig. 3. The arrangement of 12 and 18 TeO_2 capsules at neutron trap of the DNRR

By rearrangement 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 rearrangement of the neutron trap satisfies the safety condition as well as the thermal neutron flux for radioisotope production.

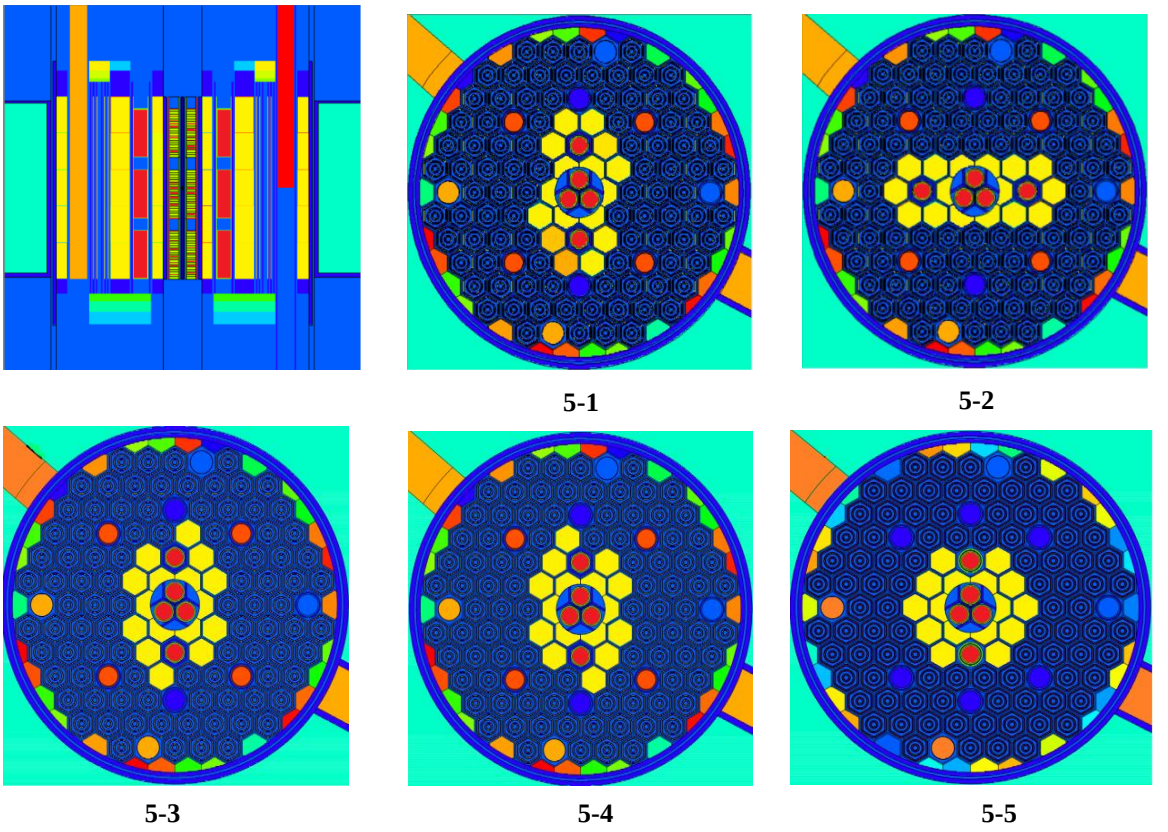

Fig. 4. Loaded positions of 15 TeO_2 target capsules in the neutron trap after its rearrangement with 5 different options.

In the new design, it is very simple and easy for carrying out experiments to test and to confirm the calculation results. Under this arrangement, the violation in thermal hydraulics limitation is avoided as calculation results were confirmed in the design of new LEU core as the 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 I. It is convenient that all cases studied were used old capsules and only adding more capsules at neutron trap after its rearrangement.

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

Number of loaded capsules	Detailed explanation
9	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 the second ring with beryllium rod at center (Fig. 3). Optimizing axial direction of thermal neutron flux. Top and bottom of all capsules covered by light water reflector.
12 – Graphite	As the first new case above but top and bottom of all capsules covered by graphite reflector.
15 – Long (5-1, 5-3, 5-4)	Creating two more irradiation channels next to neutron trap following axial direction in the reactor core (Fig. 4). It is an easy way for rearrangement of neutron trap with 3 options to select.
15 – Wide (5-2)	Creating two more irradiation channels next to neutron trap following radial direction in the reactor core (Fig. 4). It is also very easy way for rearrangement of neutron trap.
15 – Normal (5-5)	Creating two more irradiation channels next to neutron trap at cell 5-6 and 9-6 (Fig. 4).
18	As the first new case above but each irradiation channel can be loaded with 3 capsules in axial direction.

B. MCNP code for neutron fluxes and reaction rate calculation

The MCNP6 code was used for neutron flux and reaction rate calculation for TeO_2 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 libraries [8] 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 k_{eff} 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*:

$$\phi_m = \int \phi_m(E)dE \qquad (1)$$

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 [5]:

$$\Phi_m = \frac{P\bar{\nu}}{1.6022\times10^{-13}w_f}\frac{1}{k_{eff}}\phi_m \qquad (2)$$

where P is power of reactor (MW), $\bar{\nu}$ 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 k_{eff} 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:

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

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

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

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

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

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

We can interfere that

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

With σ is micro cross section (barn); φ is neutron flux (n/cm^2.s); λ is decay constant (s^{-1}) and N is the number density of isotopes (atoms/cm^3).

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 follows:

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

And t_{irr} and t_{decay} 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 follows:

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

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

The number density of Te-130 can be calculated by

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

With $N_A = 6.023.10^{23} mol^{-1}$ 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:

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

So the activity in each TeO_2 target capsule can be determined through the calculation by applying equations from (4) to (11).

I-131 activity can also be determined by using ORIGEN2.1 code with input data including irradiation time under specified neutron flux, cooling time and substituted capture cross section of Tellurium isotopes in activation material library. The thermal neutron flux and micro cross section of Tellurium isotope were estimated by MCNP6 code.

III. CALCULATION RESULTS AND DISCUSSIONS

A. Thermal neutron flux

In the axial direction, the thermal neutron flux distribution of 1 cm/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. However in case adding only 2 new irradiation channels as in Fig. 4, the reducing of thermal neutron flux at neutron trap is not much and only under 5%. Fig. 5 depicts the profile of thermal neutron trap without loading capsules for radioisotope production.

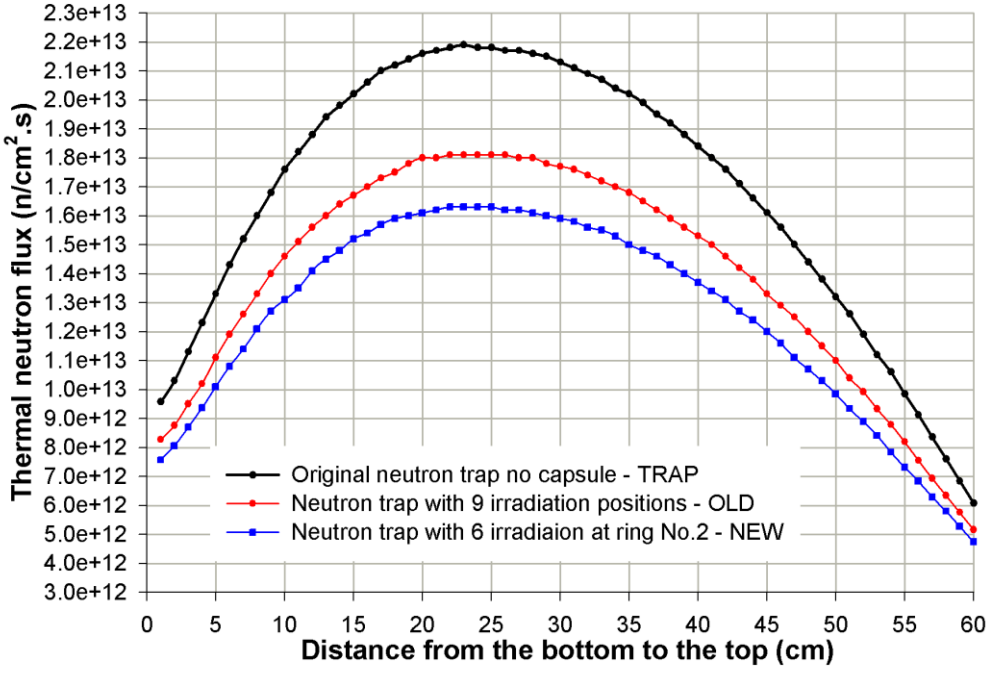

Fig. 5. Thermal neutron flux distribution at original neutron trap and modified for loading 9 capsules (OLD) and new irradiation positions (NEW)

Basing on the obtained calculation results, the best distance in axial direction that has the 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 the top and the bottom.

When loading TeO_2 capsules at different irradiation positions inside the reactor core as in Table I, the average thermal neutron flux inside TeO_2 target capsules is described in the Fig. 6. The maximum of average thermal neutron flux in the 9 capsules loaded is about 1.0×10^{13} n/cm^2.s while in other cases just of 8.8×10^{12}, 8.3×10^{12} and 8.0×10^{12} with 15, 18 and 12 capsules loaded, respectively. The micro cross section in barn unit of Te-130 isotope in thermal energy is about 1.53E-01 barns.

The thermal neutron flux inside all capsules is high enough from 5.3×10^{12} n/cm^2.s to 1.0×10^{13} n/cm^2.s at top and middle position at irradiation channel. The detail axial distribution of each node of 0.78 cm/node inside 3 TeO_2 target capsules from bottom to the top is depicted in the Fig. 7.

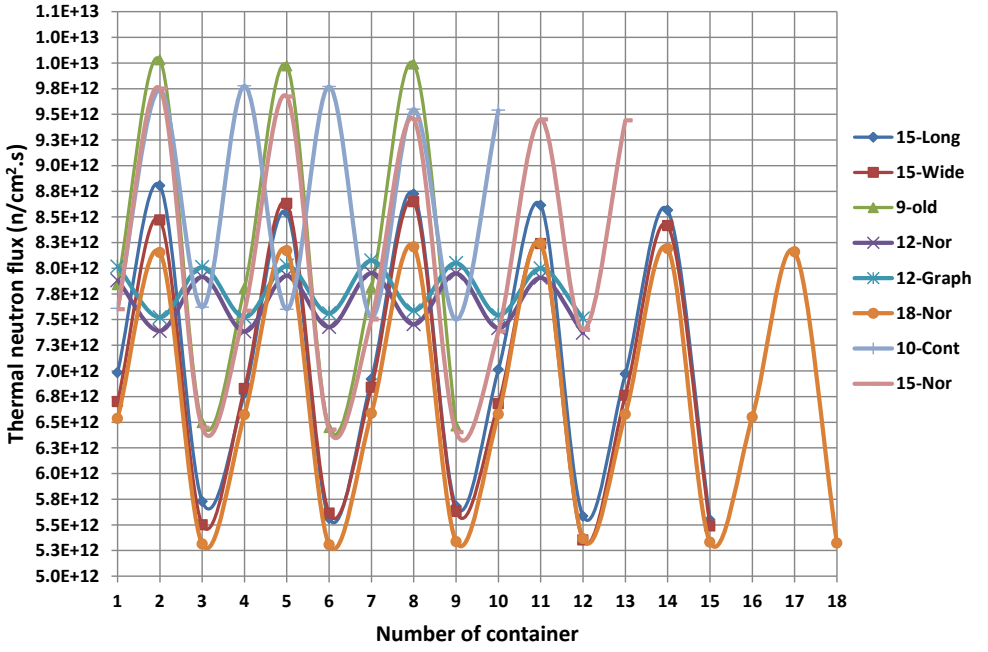

Fig. 6. The average thermal neutron flux in each capsule of 6 calculated cases

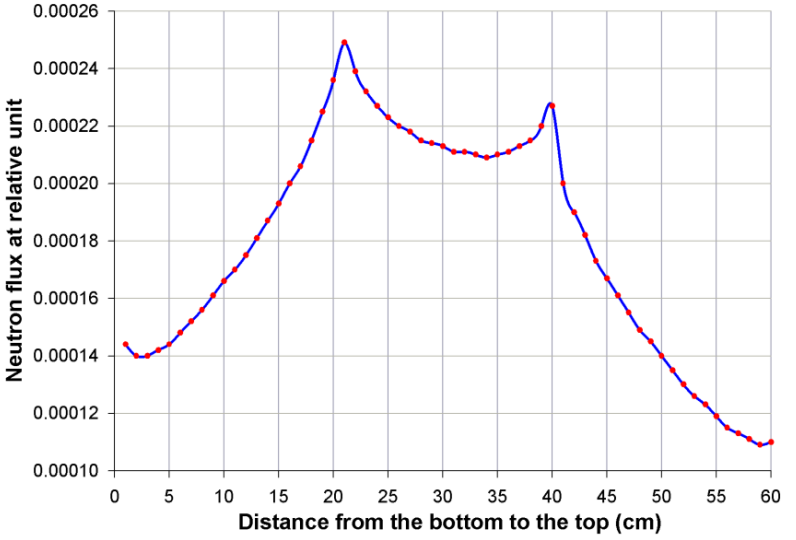

Fig. 7. Relative thermal neutron flux of 3 capsules at each node in an irradiation channel from bottom to the top

B. 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 II with the difference of TeO_2 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.

Table III. Negative reactivity insertion with different mass of Te-130 from 54 to 62 gram in each TeO_2 target capsule

Number of capsule	Negative reactivity (cents)
9	(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)
18	(86.5 – 96.5)

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 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 103^0C. So the rearrangement of neutron trap in 5 investigated cases will be also satisfied the limit of fuel cladding temperature after operating of 8 years.

C. 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/cm^2.s, irradiation time from 24 to 150 hours and the same target mass of 62 gram Te-130, calculation results are shown in Table II 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 each channel have different thermal neutron flux, the archived activity of I-131 after 150-hour irradiation of current 9 capsules has the ratio as 1.00:1.20:1.54 as top, bottom and middle positions of target.

Table II. 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

Irradiation time (hr.)	Activity of I-131 (Ci) with different thermal neutron flux (n/cm^2.s)						
	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
160	2.75	2.85	3.37	3.52	4.04	4.40	5.18
170	2.87	2.98	3.52	3.68	4.23	4.60	5.42
180	2.99	3.10	3.67	3.84	4.40	4.80	5.65

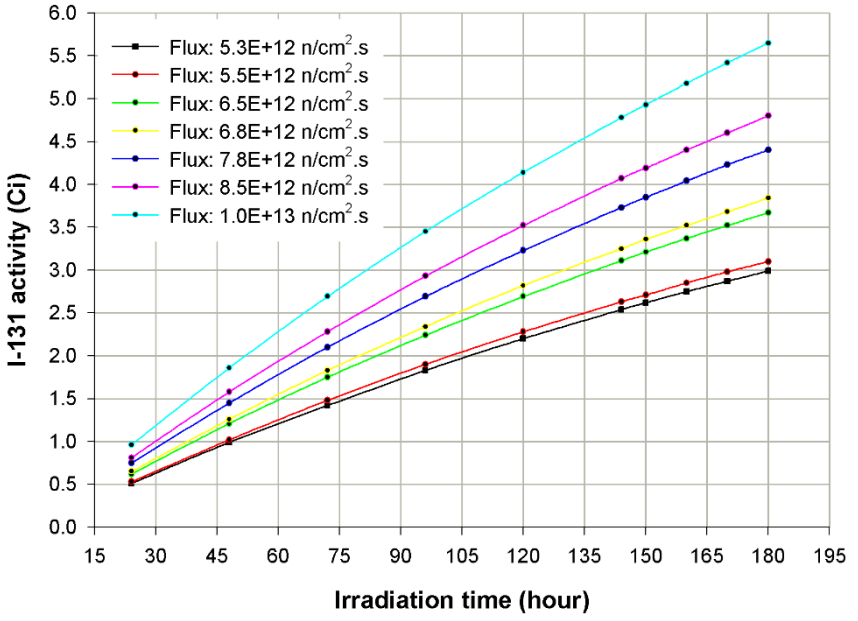

Fig. 8. Activity of I-131 using about 62 gram TeO_2 target capsule with different thermal neutron flux

The calculation results of I-131 activity with different target mass in 6 investigated cases are described in Table III and Fig. 9. The irradiation condition includes 150-hour irradiation time, mass of target from 54 to 62 gram of Te-130. The obtained results showed that the total activity of I-131 in new design comparing with old design using 9 capsules is increased from 19.24 to 38.77%. It is satisfied the requirements for enhancement of radioisotope production on the DNRR.

Table III. Calculation results of I-131 activity of 6 cases with different mass of TeO_2 target

Mass of TeO_2 target (gram)	**9**		**12-normal**		**12-graphite**	
	After 150 hrs	**Decay 18 hrs**	**After 150 hrs**	**Decay 18 hrs**	**After 150 hrs**	**Decay 18 hrs.**
200	31.23	29.27	38.67	36.25	39.47	37.00
210	32.79	30.74	40.60	38.06	41.45	38.85
220	34.35	32.20	42.53	39.87	43.42	40.70
230	35.91	33.66	44.47	41.69	45.39	42.56
Increasing % (comparing with 9 capsules)			19.24		20.89	
Mass of TeO_2 target	**15-Long (5-1, 5-3, 5-4)**		**15-Wide (5-2)**		**18**	
	After 150	**Decay 18**	**After 150**	**Decay 18**	**After 150**	**Decay 18**

(gram)	hrs.	hrs.	hrs.	hrs.	hrs.	hrs.
200	44.95	42.14	43.86	41.12	51.00	47.81
210	47.20	44.24	46.05	43.17	53.55	50.20
220	49.44	46.35	48.25	45.23	56.10	52.59
230	51.69	48.46	50.44	47.29	58.65	54.98
Increasing % (comparing with 9 capsules)	30.53		28.81		38.77	
Mass of TeO_2 target (gram)	**10 (5-5)**		**13 (5-5)**		**15 (5-5)**	
	After 150 hrs.	**Decay 18 hrs.**	**After 150 hrs.**	**Decay 18 hrs.**	**After 150 hrs.**	**Decay 18 hrs.**
200	37,18	34,85	41,26	38,68	45,32	42,48
210	39,03	36,59	43,32	40,61	47,58	44,61
220	40,89	38,33	45,39	42,55	49,85	46,73
230	42,75	40,08	47,45	44,48	52,11	48,85
Increasing% (comparing with 9 capsules)	16.00		24.32		31.10	

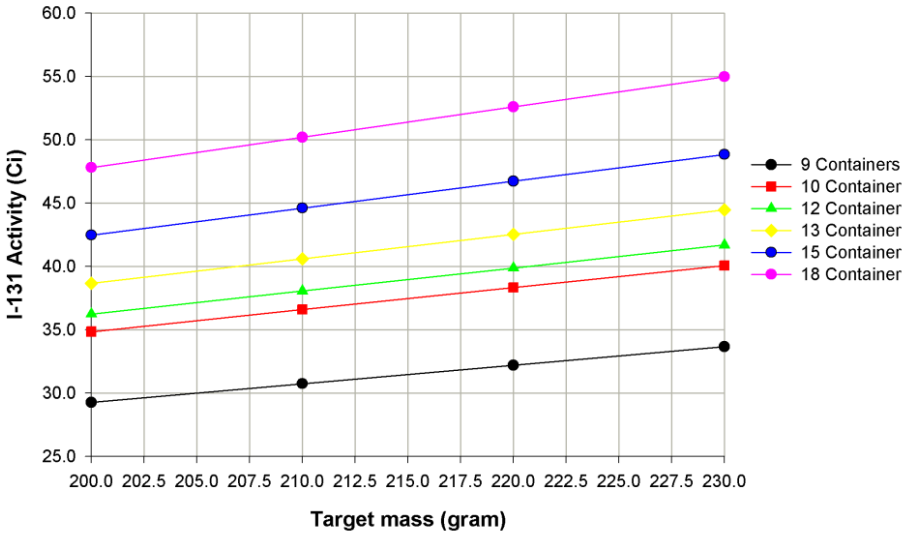

Fig. 9. Total activity of I-131 with different TeO_2 mass and under 150-hour irradiation

In 6 new cases considered, the simplest way is rearrangement of neutron trap with an option to create 2 irradiation channels by replacing the beryllium rods at cell 5-6 and 9-6 to load 6 capsules more. The new core configuration still keeps the neutron trap, 92 LEU fuel assemblies and 10 beryllium rods. And this core configuration was confirmed the safety by evaluating the maximum fuel cladding temperature by PLTEMP4.2 code [10] at hottest fuel assembly located at cell 4-5. Following the calculation results, under 27^{o}C of water at core inlet, the maximum temperature is still lower than 88^{o}C and meets requirements for operating limit condition of the VVR-M2 LEU fuel type of the DNRR.

IV. CONCLUSIONS AND REMARKS

The calculation about the enhancement radioisotope production for I-131 by rearrangement of neutron trap to loading more target capsules using MCNP6 code were conducted. In general, 6 cases for enhancement of I-131 radioisotope production on the DNRR can be carried out easily and meet the safety requirements in reactor operation. Comparing with old design using 9 containers, the total activity of I-131 isotope was increased from about 19.2% to 38.8% with loading TeO_2 target mass increasing from 25% to 50%. The rearrangement of beryllium rods around neutron trap at cell 5-6 and 9-6 to create 2 new irradiation channels is highly considered because of safety, easy implementation and effectiveness in production of I-131.

The design calculation is a very important step before carrying out testing a reactor operating cycle with new design for changing neutron trap to satisfy safety of the normal operation condition of the DNRR. The increasing I-131 product and operation time as well as using economical fuel are the main purposes in effective utilizations and applications of the DNRR. The calculation results can also be used as reference for the new research reactor in design of radioisotope production especially of I-131 isotope.

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