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On Some Formulae for the Radioisotope Formation (I)

—When a Reactor is Operated Regularly at a Certain Time Intervals—

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Abstract

Some formulae have been derived for the handy calculation of the formation of radioisotope when a reactor is operated regularly on a usual on-off pattern. In particular, the case of isotope production with the present operation condition of the Korean reactor, which is in operation for 8.2 hours from Monday to Thursday and is not operated on Friday and Sunday but is back in operation on Saturday only for 3.2 hours, is discussed herein with special emphasis. Should there be no secondary nuclear reaction resulting in the transformation of produced nuclide, the formula for the calculation of its activity could be derived as follows:

$$A=\Phi N_0\sigma(1-e^{-8.2\lambda})\ e^{-\lambda(t-8.2)}\left[\frac{e^{24n\lambda}-1}{e^{24\lambda}-1}-\frac{e^{24m\lambda}}{e^{24\times7\lambda}-1}\{e^{-24\lambda}(e^{24\times7s\lambda}-1)\right.$$
$$\left.+(e^{24\times7r\lambda}-1)+e^{24\lambda}(e^{24\times7q\lambda}-1)\}\right]$$
$$+\Phi N_0\sigma\frac{(1-e^{-3.2\lambda})}{e^{24\times7\lambda}-1}e^{-\lambda(t-3.2-24m)}(e^{24\times7r\lambda}-1)$$

where A: activity (dps),

Φ: neutron flux (n cm^{-2} sec^{-1}),

N_0: number of atoms before the irradiation,

σ: activation cross section (cm^2),

λ: disintegration constant of the radioactiveisotope formed (hr^{-1}),

t: elapsed time of target in the reactor (hr),

n: number of elasped days of target in the reactor,

m: number of days from the first day of sample irradiation to Friday,

s, r, q: number of weekday of Friday, Saturday and Sunday, respectively.

Since the above formula consists of many invariables on the whole, the activity of each radioisotope to be produced can be easily and conveniently made available from the chart in advance which is made of the invariable terms calculated.

요 약

원자로를 여러가지 불연속적 방법으로 가동시킬 때 얻어지는 방사성동위원소의 생성량을 구하는 식을 안출하였다. 특히 현재 우리나라 원자로의 가동조건인 월요일부터 목요일까지 매일 평균 8.2시간을 가동하고 금요일과 일요일은 가동치않고 토요일은 평균 3.2시간으로 가동하는 조건을 중점적으로 다루었다. 이 경우의 activity를 구하는 식은 생성동위원소가 제2의 핵반응으로 소실되지 않을 경우에는

$$A=\Phi N_0\sigma\ (1-e^{-8.2\lambda})e^{-(t-8.2)}\left[\frac{e^{24n\lambda}-1}{e^{24\lambda}-1}-\frac{e^{24m\lambda}}{e^{24\times 7\lambda}-1}\{e^{-24\lambda}\ (e^{24\times 7s\lambda}-1)+(e^{24\times 7r\lambda}-1)+e^{24\lambda}(e^{24\times 7q\lambda}-1)\}\right]+\Phi N_0\sigma\frac{(1-e^{-3.2\lambda})}{e^{24\times 7\lambda}-1}e^{-\lambda(t-3.2-24m)}\ (e^{24\times 7r\lambda}-1)$$

가 된다. 여기서 A: activity (dps), Φ: 중성자속(n cm^{-2} sec^{-1}), N_0: 조사되기전 원자수, σ: 방사화단면적(cm^2), λ: 생성방사성동위원소의 붕괴상수(hr^{-1}), t: 조사하기 시작해서 끄집어낼 때까지의 시간(hr), n: 조사일수, m:금요일이 처음 나타날때까지의 조사일 수, s, r, q: 조사기간중 금요일, 토요일 및 일요일이 나타난수를 각각 뜻한다.

윗식은 거의 고정항들로 구성되있으므로 각 동위원소에 대해 이 고정항들을 계산하여 그 값을 구해 도표를 만들어 이 식이 보다 편리하게 이용되도록 하였다.

1. Introduction

It would be extremely convenient if the yield of radioisotope formation could be precisely calculated prior to its production. It is a common practise, for the radioisotope production, to simply refer to the yield table[1] or to carry out calculation by hand on case-by-case basis, when the reactor is put into continuous operation.

However, that is practically not so easy a job in actual work and such is not applicable in our daily work owing to the fact that the reactor is operated discontinuously. So far as the present methodology of radioisotope production is concerned, formula is not yet available to calculate the activity of radioisotopes in advance when the reactor is frequently turned on and off periodically as in the operational cases of most reactor establishments.

The Korean research reactor having 250 KW thermal capacity is normally in operation for 8.2 hours per day on the average from Monday to Thursday, however, it is not operated on every Friday for the regular check-up and maintenance, while it is back in operation on Saturday but only for 3.2 hours. On every Sunday, the reactor usually remains idle. This time-schedule is repeated on weekly basis during spring, summer and autumn, except on special occasion, or when holiday comes on a weekday. During the winter months, the reactor is operated one hour shorter than the normal operation in a day.

The present work is aimed at deriving a convenient and logical calculation formula in the case of regular reactor operation at a certain time intervals.

The derived formula looks somewhat complicated because it has many terms. However, since it is comprised of invariable terms on the whole, a handy chart can be readily drawn up by taking all the calculated values of invariables into account.

For convenience' sake, some examples of the radioisotope formation by means of (n, γ) reaction are illustrated herewith. In the examples, selection is made of isotopes commonly produced with the neutron flux in the range of $10^{12}-10^{13}$ n cm^2 sec^{-1} for more than 8-hour irradiation time. For the activity calculation of radioisotopes to be produced for the irradiation time of shorter than 8 hours, the Whitson's Yield Table[1] is more than enough to be of use.

The decay scheme data referred to here in the calculation were quoted from the Chart of the Nuclides[2] published in July 1966 and Dzhelepov's Decay Schemes of Radioactive Nuclei[5].

2. Theory and Derivation of Formulae

The two schemes of radioisotope formation as shown in Fig. 1 are taken into practical consideration:

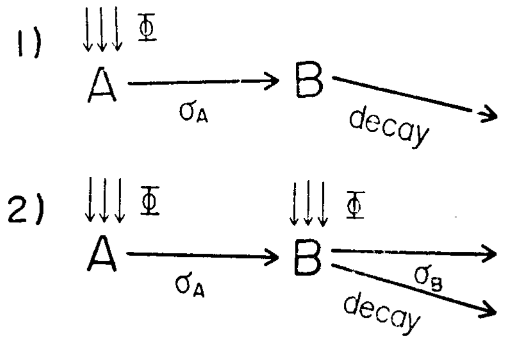

Fig. 1. Formation schemes of radioisotopes

In the first case, there should occur no secondary nuclear reaction of the produced radioisotope, or its secondary nuclear reaction, if any, could be taken negligible.

In the second case, the formed radioisotope is subject to the secondary nuclear reaction, thereby the transformation of the radioactive nuclide produced takes place in the reactor as irradiation goes on.

2-1. The case in which no secondary nuclear reaction of the produced radioisotope should occur.

In this case, if there is no significant burn-out of the parent isotope, its formation formula is given by:

$$A=\Phi N_0\sigma(1-e^{-\lambda t}) \quad \cdots\cdots\cdots\cdots(1)$$

where A: activity (dps),

Φ: neutron flux (n cm^{-2} sec^{-1}),

N_0: number of atoms before the irradiation,

σ: activation cross section of the target atom (cm^2),

λ: disintegration constant of the radioactive isotope formed (hr^{-1}),

t: rradiation time of target in the reactor(hr).

Hence, consideration is given to the various cases when the irradiation time is not continuous.

2-1-1. The case when the reactor is operated for *k* hours per day regularly for a period of *n* days.

Under the practical circumstances, the reactor is turned on and off in accordance with a certain timetable everyday, i. e., it starts operating from the scheduled preset time every morning and is put in for t hours in total.

Then, the activity produced on the first day, A_1, can be calculated by taking account of decay term in equation (1), and it turns out to be as follows:

$$A_1=\Phi N_0\sigma(1-e^{-k\lambda})e^{-\lambda(t-k)}$$

In the same manner, the activity on the second operation day, A_2, is:

$$A_2=p(1-e^{-k\lambda})e^{-\lambda(t-k-24)}$$

where p is $\Phi N_0\sigma$

And also those on the third consecutive day, the fourth consecutive day, ……, and the like, which are expressed as A_3, A_4, …, A_n, are derived as follows:

$$A_3=p(1-e^{-k\lambda})e^{-\lambda(t-k-24\times 2)}$$
$$A_4=p(1-e^{-k\lambda})e^{-\lambda(t-k-24\times 3)}$$
$$\vdots \qquad \vdots$$
$$A_n=p(1-e^{-k\lambda})e^{-\lambda\{t-k-24(n-1)\}}$$

As the consequence the accmulated total activity, A', becomes to be as below:

$$A'=\sum_{i=1}^{n}A_i=p(1-e^{-k\lambda})e^{-\lambda(t-k)}\,(1+e^{24\lambda}+e^{24\times 2\lambda}+\cdots\cdots+e^{24(n-1)\lambda})$$

$$A'=p(1-e^{-k\lambda})e^{-\lambda(t-k)}\left(\frac{e^{24n\lambda}-1}{e^{24\lambda}-1}\right) \quad \cdots\cdots\cdots (2)$$

2-1-2. The case when the reactor is operated for k hours per day regularly for a period of n days but on the m_{th} day from the first day of operation the reactor is turned off.

The activity in this case could be easily obtained by subtracting that produced on the m_{th} day from the equation (2). If the activity to be calculated is set forth as A″, t is the elapsed time of target in the reactor and the reactor starts operating from the same time in the morning everyday, it is then:

$$A''=p(1-e^{-k\lambda})e^{-\lambda(t-k)}\left(\frac{e^{24n\lambda}-1}{e^{24\lambda}-1}-e^{24(m-1)\lambda}\right) \quad \cdots\cdots(3)$$

2-1-3. The case when the reactor is operated for k hours per day regularly for a period of n days but on the f_{th} day the reactor is operated for k' hours.

On the f_{th} day, the reactor is not operated for k hours as in the normal operation pattern, but is put in operation for k' hours which would be either longer or shorter than the normal operation time. The activity in this case then is obtained by adding that produced during the operation time of k' hours to the equation (3). If its activity is defined as A‴, t is the elapsed time of target and the reactor starts operating from the same time in the morning everyday, then:

$$A'''=p(1-e^{-k\lambda})e^{-\lambda(t-k)}\left(\frac{e^{24n\lambda}-1}{e^{24\lambda}-1}-e^{24\lambda(f-1)}\right)+p(1-e^{-k'\lambda})e^{-\lambda\{t-k'-24(f-1)\}} \quad \cdots\cdots(4)$$

2-1-4. The case when the reactor is operated for k hours per day regularly for a period of n days, but on the m_{th} day from the first day of operation the reactor is turned off, and this turn-off day comes periodically at a time interval of every j days, regular turn off schedule is repeated for h consecutive rounds.

The activity to be built up in this particular case could be made available, which would be something like an expanded form of equation (3). If the activity is defined as A⁗, then its formula could be derived from equation (3) as follows:

$$A''''=p(1-e^{-k\lambda})e^{-\lambda(t-k)}\left\{\frac{e^{24n\lambda}-1}{e^{24\lambda}-1}-(e^{24(m-1)\lambda}+e^{24(m-1+j)\lambda}+e^{24(m-1+2j)\lambda}+\cdots\cdots+e^{24\{m-1+(h-1)j\}\lambda})\right\}$$

$$A''''=p(1-e^{-k\lambda})e^{-\lambda(t-k)}\left\{\frac{e^{24n\lambda}-1}{e^{24\lambda}-1}-e^{24(m-1)\lambda}\left(\frac{e^{24jh\lambda}-1}{e^{24j\lambda}-1}\right)\right\} \quad \cdots\cdots(5)$$

2-1-5. The case when the reactor, like the Korean research reactor, TRIGA Mark II, is operated for k hours per regularly for a period of n days, but on both Friday and Sunday the reactor is turned off, while it is put back in operaion on every Saturday for k' hours.

In this case, its activity, A, could be derived from the formulae, (1), (2), (3), (4) and (5), as follows:

$$\begin{aligned}A=&p(1-e^{k\lambda})e^{-\lambda(t-k)}\left\{\frac{e^{24n\lambda}-1}{e^{24\lambda}-1}-e^{24(m-1)\lambda}\left(\frac{e^{24\times7s\lambda}-1}{e^{24\times7\lambda}-1}\right)-e^{24m\lambda}\left(\frac{e^{24\times7r\lambda}-1}{e^{24\times7\lambda}-1}\right)\right.\\&\left.-e^{24(m+1)\lambda}\left(\frac{e^{24\times7q\lambda}-1}{e^{24\times7\lambda}-1}\right)\right\}\\&+p(1-e^{-k'\lambda})e^{-\lambda(t-k'-24m)}\\&+p(1-e^{-k'\lambda})e^{-\lambda\{t-k'-24(m+7)\}}\\&+p(1-e^{-k'\lambda})e^{-\lambda\{t-k'-24(m+7\times2)\}}\\&+p(1-e^{-k'\lambda})e^{-\lambda[t-k'-24\{m+7(r-1)\}]}\end{aligned}$$

where m: number of days from the first day of sample irradiation to Friday,
s,r,q: the numbers of weekdays of Friday, Saturday and Sunday, respectively, which occur during the sample irradiation.
Arranging the above,

$$\begin{aligned}A=&p(1-e^{-k\lambda})e^{-\lambda(t-k)}\left[\frac{e^{24n\lambda}-1}{e^{24\lambda}-1}-\frac{e^{24m\lambda}}{e^{24\times7\lambda}-1}\{e^{-24\lambda}(e^{24\times7s\lambda}-1)+(e^{24\times7r\lambda}-1)+e^{24\lambda}(e^{24\times7q\lambda}-1)\}\right]\\&+p\frac{(1-e^{-k'\lambda})}{e^{24\times7\lambda}-1}e^{-\lambda(t-k'-24m)}(e^{24\times7r\lambda}-1)\end{aligned} \quad \cdots\cdots(6)$$

In case when s, r and q in equation (6) would have the same value, s, the formula could then be simplified as follows:

$$A=p(1-e^{-k\lambda})e^{-\lambda(t-k)}\left\{\frac{e^{24n\lambda}-1}{e^{24\lambda}-1}-\frac{(e^{24\times 7s\lambda}-1)}{e^{24\times 7\lambda}-1}e^{24m\lambda}(1+e^{-24\lambda}+e^{24\lambda})\right\}+p\frac{(1-e^{-k'\lambda})}{e^{24\times 7\lambda}-1}e^{-\lambda(t-k'-24m)}(e^{24\times 7s\lambda}-1) \qquad \cdots\cdots(7)$$

Should there be some change in the conditions of 2-1-5, anothr modified, revised or altered formaul might be obtainable by means of 2-1-1, 2-1-2, 2-1-3 and 2-1-4.

Table I is the chart made of calculated values of the invariable terms in equation (6) and (7) for some examples of radioisotopes.

Table 1. The calculated values of the invariables in equation (6) and (7)

Target		^{44}Ca	^{46}Ca	^{50}Cr	^{75}As	^{121}Sb	^{130}Ba
Natural abundance (%)		2.06	0.0033	4.31	100	57.3	0.101
σ (n, γ) (barn)		0.7	0.3	17.0	4.5	6.06	8.8
Isotope to be produced		^{45}Ca	^{47}Ca	^{51}Cr	^{76}As	^{122}Sb	^{131}Ba
$t_{\frac{1}{2}}$		163 d	4.5 d	27.8 d	26.5 hr	2.8 d	11.6 d
$P=\Phi\cdot\frac{W^*}{A^{**}}\cdot f^{***}\cdot Av^{****}\cdot\sigma$	$\Phi=1.0\times10^{12}$	2.17×10^8	1.49×10^5	8.48×10^9	3.62×10^{10}	1.72×10^{10}	3.90×10^7
	$\Phi=1.5\times10^{12}$	3.25×10^8	2.23×10^5	1.27×10^{10}	5.42×10^{10}	2.57×10^{10}	5.84×10^7
	$\Phi=2.0\times10^{12}$	4.33×10^8	2.97×10^5	1.70×10^{10}	7.23×10^{10}	3.43×10^{10}	7.79×10^7
	$\Phi=3.0\times10^{12}$	6.50×10^8	4.46×10^5	2.55×10^{10}	1.08×10^{11}	5.15×10^{10}	1.17×10^8
	$\Phi=1.0\times10^{13}$	2.17×10^9	1.49×10^6	8.48×10^{10}	3.62×10^{11}	1.72×10^{11}	3.90×10^8
λ (hr^{-1})		1.77×10^{-4}	6.42×10^{-3}	1.04×10^{-3}	2.62×10^{-2}	1.03×10^{-2}	2.49×10^{-3}
$1-e^{-k\lambda}$	$k=7.2$	1.27×10^{-3}	4.49×10^{-2}	7.49×10^{-3}	1.72×10^{-1}	7.08×10^{-2}	1.78×10^{-2}
	$k=8.2$	1.45×10^{-3}	5.12×10^{-2}	8.53×10^{-3}	1.93×10^{-1}	8.08×10^{-2}	2.06×10^{-2}
$p(1-e^{-k\lambda})$	$\Phi=1.5\times10^{12}$, $k=7.2$	4.14×10^5	1.00×10^4	9.51×10^7	9.32×10^9	1.82×10^9	1.04×10^6
	$\Phi=1.5\times10^{12}$, $k=8.2$	4.73×10^5	1.14×10^4	1.08×10^8	1.05×10^{10}	2.08×10^9	1.20×10^6
	$\Phi=1.0\times10^{13}$, $k=7.2$	2.76×10^6	6.69×10^4	6.35×10^8	6.23×10^{10}	1.22×10^{10}	6.94×10^6
	$\Phi=1.0\times10^{13}$, $k=8.2$	3.15×10^6	7.63×10^4	7.23×10^8	6.99×10^{10}	1.39×10^{10}	8.03×10^6
$e^{24\lambda}$		1.004	1.17	1.03	1.88	1.28	1.06
$e^{-24\lambda}$		9.96×10^{-1}	8.85×10^{-1}	9.71×10^{-1}	5.32×10^{-1}	7.81×10^{-1}	9.43×10^{-1}
$e^{24\lambda}-1$		4.25×10^{-3}	1.67×10^{-1}	2.54×10^{-2}	8.75×10^{-1}	2.80×10^{-1}	6.16×10^{-2}
$e^{24\times7\lambda}$		1.03	2.94	1.19	8.18×10	5.64	1.52
$e^{24\times7\lambda}-1$		3.02×10^{-2}	1.94	1.91×10^{-1}	8.08×10	4.64	5.21×10^{-1}
$1+e^{-24\lambda}+e^{24\lambda}$		3.00	3.02	3.00	3.41	3.06	3.00
$1-e^{-k'\lambda}$	$k'=2.2$	3.89×10^{-4}	1.39×10^{-2}	2.28×10^{-3}	5.59×10^{-2}	2.24×10^{-2}	5.49×10^{-3}
	$k'=3.2$	5.66×10^{-4}	2.73×10^{-2}	3.32×10^{-3}	9.01×10^{-2}	3.29×10^{-2}	7.97×10^{-3}
$p(1-e^{-k'\lambda})$	$\Phi=1.5\times10^{12}$, $k'=2.2$	1.27×10^5	3.11×10^3	2.89×10^7	3.04×10^8	5.75×10^8	3.20×10^5
	$\Phi=1.5\times10^{12}$, $k'=3.2$	1.85×10^5	6.09×10^3	4.22×10^7	4.88×10^8	8.45×10^8	4.61×10^5
	$\Phi=1.0\times10^{13}$, $k'=2.2$	4.94×10^5	2.07×10^4	1.93×10^8	2.02×10^9	3.85×10^9	2.14×10^6
	$\Phi=1.0\times10^{13}$, $k'=3.2$	7.19×10^5	4.07×10^4	2.82×10^8	3.26×10^9	5.66×10^9	3.08×10^6

*Weight of target=1 gr
**Atomic weight
***Natural abundance
****Avogadro's number

Using Table 1 the formation curve for ^{122}Sb is drawn as in Fig. 2. This curve is made up under the condition that the sample was placed on Monday in the rotary specimen rack where the neutron flux is $1.5\,n\,cm^{-2}\,sec^{-1}$.

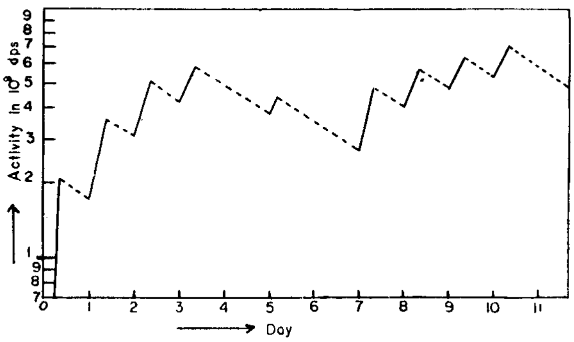

Fig. 2. Formation curve of ^{122}Sb being operated under the condition of 2-1-5.

2-2. The case when the transformation of the produced radioisotope occurs due to the secondary nuclear reaction.

Under the condition of continuous irradiation, the rate of formation of radioisotope, B, which is to be obtained from 2) of Fig. 1 is given by

$$\frac{dN_B}{dt}=\Phi N_A\sigma_A-N_B(\lambda+\Phi\sigma_B) \quad \cdots\cdots\cdots\cdots(8)$$

On the other hand, the rate of reduction of the target nuclide, A, is expressed as:

$$-\frac{dN_A}{dt}=\Phi N_A\sigma_A$$

From the above formula,

$$N_A=N_0e^{-\Phi\sigma_At} \quad \cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots(9)$$

Putting equation (9) to (8), we get:

$$\frac{dN_B}{dt}=\Phi\sigma_AN_0e^{-\Phi\sigma_At}-N_B(\lambda+\Phi\sigma_B) \quad \cdots(10)$$

The solution[3] of this differential equation is

$$\lambda N_B=\frac{\Phi N_0\sigma_A\lambda}{\lambda+\Phi\sigma_B-\Phi\sigma_A}(e^{-\Phi\sigma_At}-e^{-(\lambda+\Phi\sigma_B)t}) \quad \cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots(11)$$

Since it is obvious that the result of application for the case of regular reactor operation at a certain time intervals is almost identical to the case of 2-1, a formula which employs the typical case of 2-1-5 is given below as the representative example.

$$A=\frac{\Phi N_0\sigma_A\lambda}{\lambda+\Phi\sigma_B}(1-e^{-(\lambda+\frac{\Phi\sigma_B}{3600})k})e^{-\lambda(t-k)}[\frac{e^{24n\lambda}-1}{e^{24\lambda}-1}-\frac{e^{24m\lambda}}{e^{24\times7\lambda}-1}\{e^{-24\lambda}(e^{24\times7s\lambda}-1)+(e^{24\times7r\lambda}-1)+e^{24\lambda}(e^{24\times7q\lambda}-1)\}]+\frac{\Phi N_0\sigma_A\lambda}{\lambda+\Phi\sigma_B}(1-e^{-(\lambda+\frac{\Phi\sigma_B}{3600})k'})e^{-\lambda(t-k'-24m)}(\frac{e^{24\times7r\lambda}-1}{e^{24\times7\lambda}-1})$$

Here, it is assumed that the burn-out of the parent isotope is negligible and the cross section of B atom, σ_B, is much bigger than that of A atom, σ_A, and $\Phi\sigma_Ak$ and $\Phi\sigma_Ak'$ are sufficiently small.

3. Discussion

It is believed that the numerical solution of the invariables of the formulae could lead us to drawing up a convenient chart and table for each case of isotope production so that anyone who is interested in or engaged in the activity build-up of specific nuclides by means of neutron irradiation of TRIGA Mark II may be informed in advance of the activity level of the isotopes from the chart without tediously calculating the complicated formula.

As the equations discussed here have been derived under the supposition that the target be placed in the form of thin layer, the values calculated from the derived equations should give the maximum plausible yield. If the target is sufficiently thick[6] so as to enable to block an appreciable fraction of the particles in the incurrent beam, it is preferred that the flux of the equations discussed here must be replaced by the following equation:

$$\Phi=\Phi_o\ (1-e^{-\sigma Nx})$$

where x is the thickness of the target traversed, and N is the number of nuclei per unit volume. Here the cross section, σ, includes all the processes such as scattering, absorption and others including the shadow effect toward the incoming neutron beam.

The values of theoretical calculations by means of the derived formulae for the cases of commonly produced radioisotopes will be compared with the experimental data to be followed up in the later stage. In addition, schedule is being made to draw up calculation table to back up the formation of the case 2) of Fig. 1 as well as the formation of radioisotopes produced by the threshold reactions.

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