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Analysis of fission products during long-term shutdown of research reactor

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Introduction

- **Long-term shutdown of HANARO**

- HANARO has been in long-term shutdown since the 96th operation cycle in 2014. If the reactor is re-operated after a long-term shutdown, it is necessary to evaluate whether the criticality of the reactor is possible using the delayed neutrons in the core.
- During cold start-up, the reactor is in sub-critical state and the external neutron source should be considered.
- Single-group delayed neutron point-reactor kinetics equations

$$\frac{dn}{dt} = \frac{\rho(t) - \beta}{\Lambda} n(t) + \sum_i \lambda_i C_i(t) + S, \qquad \frac{dC_i}{dt} = \frac{\beta_i}{\Lambda} n(t) - \lambda_i C_i(t)$$

where, $n(t)$ is the total number of neutrons in the core, C_i the total number of precursors of delayed neutrons of group i, S the total neutron source strength, Λ the neutron generation time and ρ the reactivity

- If the neutrons in the core is insufficient, the criticality of the reactor should be reached by loading an external neutron source into the core. In this study, the trends of the fission products during long-term shutdown was analyzed based on the RPS signals. The photonuclear reaction rate of D_2O reflector was calculated to evaluate the neutron source term in the core.

Fig 1. Long-term shutdown of HANARO due to seismic reinforcement work

Methods and Results

- **Neutron power of the HANARO core**

- Neutron powers measured by Reactor Protection System (RPS) channels A, B and C are shown in figure 2. The x-axis is the time from 2014-07-01 to 2016-04-05 and, the y-axis denotes the percent full power (%FP).
- After the 96th operation cycle on 2014-07-01, the neutron power sharply decreased to about 4E-08 %FP on 2014-12-01.
- Half-life of the major contributing nuclides
  : 13 days until December 2014
  : 10 months after December 2014

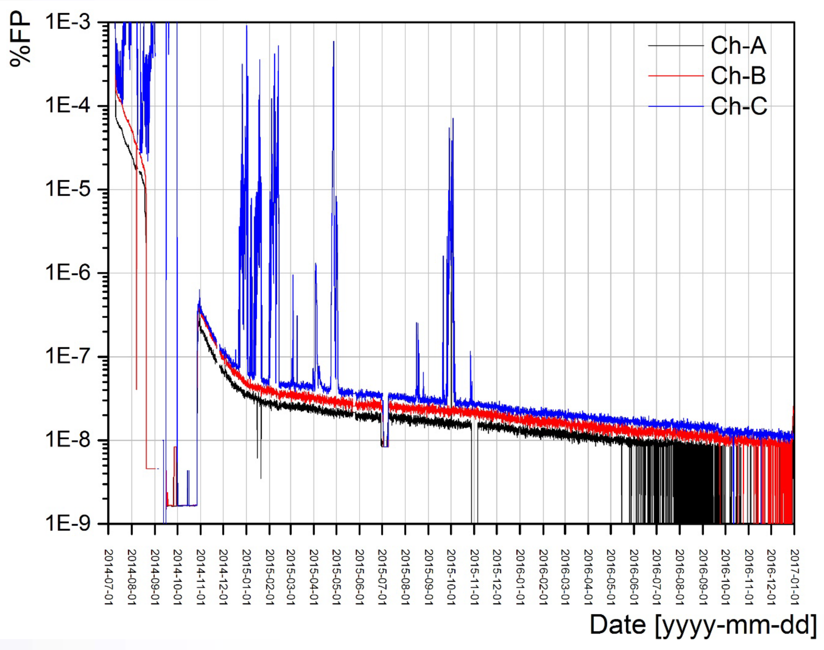

Fig 2. Neutron power signals during the long-term shutdown of HANARO measured by RPS channel A, B and C

- Contribution to the output signal of RPS during long-term shutdown
  1) Photo-neutrons generated by reaction of gamma rays with energy over 2.23 MeV emitted from fission products or activation products with heavy water
  2) Photo-neutrons generated by fission after gamma rays with an energy over 5 MeV emitted from fission products or activation products are absorbed by U-235, U-238 and etc.
  3) Neutrons emitted from a nuclide with a long half-life among delayed neutron precursors.

- **Calculation of activities of fission products**

- In order to identify radionuclides with a half-life of about 10 months after December 2014, fission products and activation daughters were calculated during the long-term shutdown of HANARO
- Calculation code : ORIGEN2.1
- Burn-up of HANARO fuel after 96th operation cycle
  1) Actual : about 100,000 MWD/MTU
  2) Calculation : 60,000 MWD/MTU and burned for 200 days.

Table 1. Activity of the top 15 nuclides after 200 days of burnup.

Nuclide	0	50	100	500	550	600	Half-life [days]
Nb-95	7.19E3	6.43E3	4.58E3	7.91E1	4.61E1	2.69E1	64.17
Zr-95	8.08E3	4.70E3	2.74E3	3.59E1	2.09E1	1.22E1	64.01
Y-91	7.41E3	4.13E3	2.28E3	2.00E1	1.10E1	6.11E0	58.50
Sr-89	6.24E3	3.14E3	1.58E3	6.52E0	3.28E0	1.65E0	50.49
Ce-141	8.14E3	2.82E3	9.70E2	1.92E1	6.61E-2	2.28E-2	32.51
Pr-144	3.10E3	2.68E3	2.37E3	8.94E2	7.01E2	7.01E+02	284.35
Ce-144	3.03E3	2.68E3	2.37E3	8.94E2	7.00E2	7.00E2	284.02
Ru-103	4.59E3	1.90E3	7.87E2	6.77E1	1.16E-1	1.16E-1	39.28
Rh-103m	4.14E3	1.71E3	7.09E2	6.10E1	1.05E-1	1.05E-1	39.28
Pr-143	7.75E3	6.70E2	5.20E1	6.91E8	4.17E-10	4.17E-10	13.56
La-140	8.49E3	6.43E2	4.28E1	1.65E8	7.29E-11	7.29E-11	12.79
Ba-140	8.40E3	5.59E2	3.72E1	1.43E8	6.34E-11	6.34E-11	12.79
Pm-147	2.91E2	3.14E2	3.04E2	2.28E2	2.12E2	2.12E2	957.86
Ru-106	2.57E2	2.34E2	2.13E2	1.00E2	8.29E1	8.29E1	368.42
Rh-106	3.93E2	2.34E2	2.13E2	1.00E2	8.29E1	8.29E1	368.42

- Half-life is calculated based on the activity of 550 and 600 days (≠ actual half-life of each nuclide)
- Candidate nuclides
  1) Ba-140 (parent) and La-140 (daughter)
     : half-life of about 13 days
     : emission of gamma rays over 2 MeV
  2) Ce-144 (parent) and Pr-144 (daughter)
     : half-life of about 10 months
     : emission of gamma rays over 2 MeV
  3) Ru-106 (parent) and Rh-106 (daughter)
     : contribution of neutron power after 2015

Table 2. Energy and intensity of gamma rays from Rh-106, La-140 and Pr-144

Nuclide	Energy [keV]	Intensity [%]	Nuclide	Energy [keV]	Intensity [%]
Rh-106	2242.4	0.00206	Rh-106	2865	1.40E-05
	2271.9	0.00137		2902.5	6.50E-05
	2309	0.00563		2917.9	9.20E-04
	2316.4	0.00636		3037.3	0.00102
	2366.04	0.0233		3055	3.50E-04
	2390.6	0.00651		3164.7	2.90E-05
	2405.96	0.0145		3249.8	1.00E-04
	2439.1	0.00459		3273.4	5.10E-05
	2456.8	2.90E-04		3375.9	1.12E-05
	2484.6	9.00E-04		3401.8	1.24E-05
	2525.2	2.00E-04	La-140	2347.88	0.85
	2542.7	0.00296		2464.1	0.0114
	2571.1	0.00145		2521.4	3.46
	2651.4	6.50E-04		2547.34	0.101
	2705.3	0.00251		2899.61	0.0668
	2709.5	0.00373		3118.51	0.0248
	2740.1	2.40E-04		3320.4	0.0038
	2787.3	8.00E-05	Pr-144	2368.3	0.000054
	2809	6.90E-04		2654.0	0.00015
	2821.1	0.0012			

- **Photonuclear reaction rate by gamma rays from fission products**

- Photonuclear reaction rate at decay time, i is given by

$$R_i = A_i \times \sum_{j=1}^{n} I_j \sigma_j$$

where, j is the index of the gamma-ray, A_i is the activity of at decay time, I_i is the emission rate of jth gamma-ray, and σ_j is the photonuclear cross-section corresponding the energy of jth gamma-ray

- Photonuclear cross-section was obtained from ENDF/B-VII.1. To calculate the photo nuclear reaction rate by emission gamma rays from Rh-106, La-140 and Pr-144, cross-section data were fitted in polynomial curve.

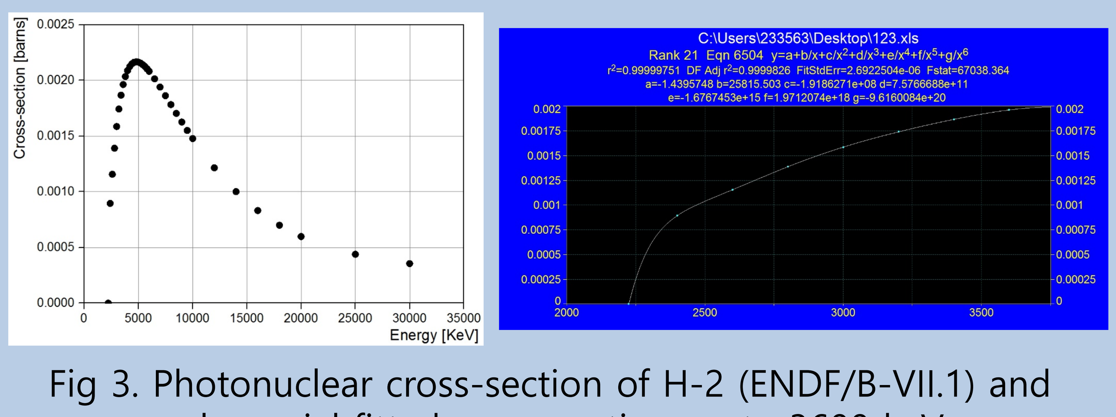

Fig 3. Photonuclear cross-section of H-2 (ENDF/B-VII.1) and polynomial fitted cross-section up to 3600 keV

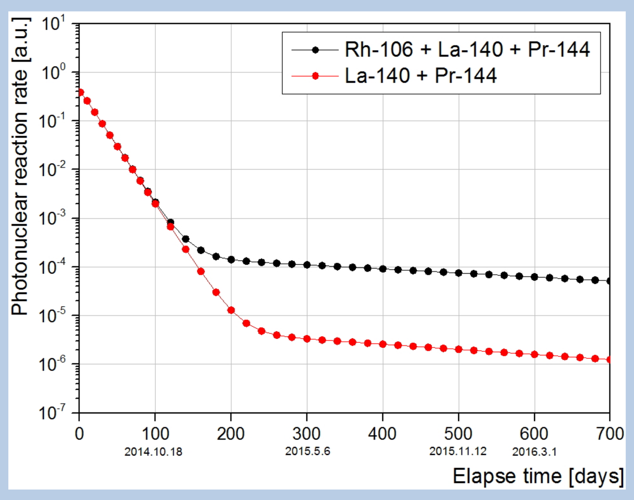

Fig 4. Calculated photonuclear reaction rate of H-2 by emitted gamma rays of Rh-106, La-140 and Pr-144

- Photonuclear reaction rate as a function of decay time was calculated.
- Ratio of the photonuclear reaction rate on 700 days to the 10 days of decay time is 1.13E-04 when all three nuclides are considered.
- Ratio of the photonuclear reaction rate on 700 days to the 10 days of decay time is 2.63E-06 when La-140 and Pr-144 are considered.

Conclusions

The neutron power of HANARO was reduced to 1E-4% FP and 1E-8% FP at 10 days and 600 days after shutdown, respectively. From the calculation results of the photonuclear reaction rate, it was confirmed that the trend of the photonuclear reaction rate by Rh-106, La-140 and Pr-144 was in good agreement with the trend of neutron power. However, when the decay time is longer than 600 days, the photonuclear reaction rate by the three nuclides is relatively higher than the neutron power. This is because other nuclides not considered in this study except La-140, Pr-144, and Rh-106 cause photonuclear reactions.