

Mo-99 HEU

A Study on Nuclear Design Methodology for Fission Mo-99 HEU Target

Mo-99 . Fission Mo-99
 "MCNP-ORIGEN" "MCNP-"
 가 , Mo-99 1.6% 가 ,
 , , .
 , (recoil loss) 가 , 가
 20 μ m 가 . Cintichem HANARO
 , 50cm, UO₂ 11 μ m 가 , (Ni)
 , Cintichem 4

Abstract

In this study, a nuclear design for a fission Mo-99 HEU target was performed. A reliability of MCNP-ORIGEN code system used for target design was evaluated. Mo-99 production amount predicted by "MCNP-ORIGEN" was consistent with that by "MCNP-Analytic Eq." within 1.6% difference. A parametric study was done for the optimization of fuel thickness, Mo-99 recoil loss rate to the variation of thickness, target cladding materials, the thickness of irradiation guide tube, and barrier materials. The key parameters which affect the Mo-99 yield ratio and surface heat flux were fuel thickness, cladding materials, and recoil loss rate. The most effective fuel thickness was shown to be 20 μ m in case of no barrier tube. Cintichem target loaded in HANARO without modification could not satisfy the safety limit such as reactivity worth change limit and ONB temperature. The UO₂ electro-deposited target coated with 10 μ m Ni barrier material in a dimension of 50cm axial length and 11 μ m fuel thickness satisfied the all design constraints and produced radioactive waste 4 times less than that of original Cintichem target.

1.

Tc-99m 80% ,
 Mo-99 Tc-99m Tc-generator 가
 Tc-99m 3 \$(가)
 , 5% 가 가
 , Nordion 가 NRU(National Research Universal)
 80% , (U.S. Department of Energy)
 10 30% , 100%
 SNL(Sandia Nation Laboratory) ACRR(Annular Core Research Reactor)
 Mo-99 1996 .(1)(2)(3)
 1995 'Fission Moly'
 Mo-99 6 Ci
 , 1996 "Fission Mo"
 Mo-99 1997
 " " "Fission ⁹⁹Mo
 가 가 3가
 , MCNP-ORIGEN LEU
 Mo-99
 .(4)(5)
 MCNP-ORIGEN
 , 가 (electro- deposited)
 , Mo-99 (Ci ⁹⁹Mo/gU)

2. MCNP-ORIGEN2

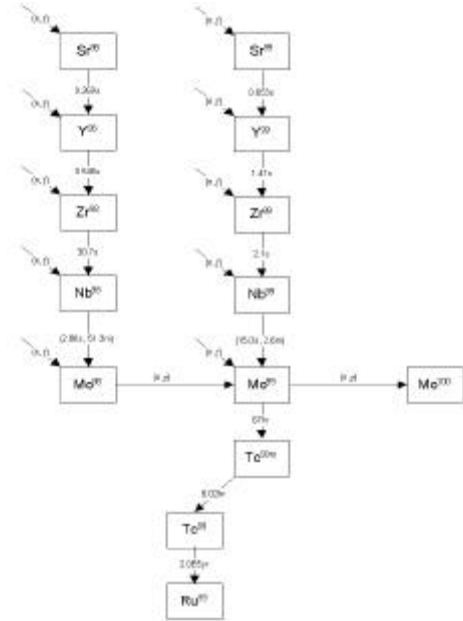
MCNP-ORIGEN MCNP-4B
 , ORIGEN-2
 Mo-99 HANARO MCNP-4B ORIGEN-2
 가
 Mo-99 가
 Mo-99 가
 MCNP-ORIGEN
 가 .

2.1 Mo-99

1 Mo-99 ,
 , (n,) , (n, f)
 Mo-99 , U-235 (direct fission)
 , U-235 Y⁹⁹, Zr⁹⁹, Nb⁹⁹ -

, U-235
 -)⁹⁹Mo
 , U-235
 (direct fission yield)
 가

Y⁹⁸, Zr⁹⁸, Nb⁹⁸
 Mo-98 ⁹⁸Mo(n,
 Mo-99



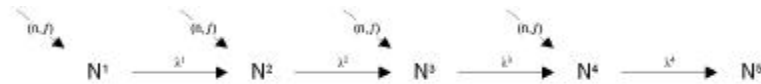
67

(half-life) 가
 143keV
 , ^{99m}Tc
⁹⁹Mo(n,)¹⁰⁰Mo

2.2 Mo-99 가
 Mo-99 가
 Mo-98 Mo-99가

1. Production-Destruction Scheme of ⁹⁹Mo in a Fission Moly Target

2 N¹, N², N³, N⁴, N⁵
 Sr-99, Y-99, Zr-99, Nb-99, Mo-99 Sr-98, Y-98, Zr-98, Nb-98, Mo-98



2. Mo-98 Mo-99 -

$$\frac{dN^1}{dt} = \gamma^1 \sigma_f \Phi N^{235} - \lambda^1 N^1 \quad (1)$$

$$\frac{dN^2}{dt} = \gamma^2 \sigma_f \Phi N^{235} + \lambda^1 N^1 - \lambda^2 N^2 \quad (2)$$

$$\frac{dN^3}{dt} = \gamma^3 \sigma_f \Phi N^{235} + \lambda^2 N^2 - \lambda^3 N^3 \quad (3)$$

$$\frac{dN^4}{dt} = \gamma^4 \sigma_f \Phi N^{235} + \lambda^3 N^3 - \lambda^4 N^4 \quad (4)$$

, γ^i : fission yield fraction of nuclide i ,
 σ_f : fission cross section of fissile nuclide,
 λ^i : decay constant of nuclide i ,
 Φ : neutron flux

$$N^{235} \text{ 가 } t \text{ } ^{235}\text{U} \text{ (} \sigma_a^{235} \text{)} \quad (5)$$

$$N^{235}(t) = N_0^{235} e^{-\mu t} \quad (5)$$

, N_0^{235} = number density of U^{235} at $t=0$, $\mu = \sigma_a^{235} \Phi$

$$N^i(t) = N^i(0) e^{-\lambda^i t} + \sum_{j=1}^{i-1} \frac{\gamma^j \sigma_f \Phi N_0^{235} \prod_{k=1}^{j-1} \lambda^k}{\mu - \sum_{k=1}^j \lambda^k} \prod_{k=1}^j (e^{-\lambda^k t} - e^{-(\mu - \sum_{l=1}^k \lambda^l)t}) \quad (1), (2), (3) \quad (4)$$

$$N^i = \frac{\gamma^i \sigma_f \Phi N_0^{235}}{\mu - \lambda^i} (e^{-\lambda^i t} - e^{-\mu t}) + \sum_{j=1}^{i-1} \frac{\gamma^j \sigma_f \Phi N_0^{235} \prod_{k=1}^{j-1} \lambda^k}{\mu - \sum_{k=1}^j \lambda^k} \prod_{k=1}^j (e^{-\lambda^k t} - e^{-(\mu - \sum_{l=1}^k \lambda^l)t}) \quad (6)$$

Mo-99 3
(7), (8)

$$\frac{dN^{99}}{dt} = \gamma^{99} N^{235} \sigma_f^{235} \Phi + \lambda^{b99} N^{b99} + \sigma_r^{98} N^{98} \Phi - \lambda^{99} N^{99} - \sigma_r^{99} N^{99} \Phi \quad (7)$$

$$\frac{dN^{98}}{dt} = \gamma^{98} N^{235} \sigma_f^{235} \Phi + \lambda^{b98} N^{b98} - \sigma_r^{98} N^{98} \Phi \quad (8)$$

γ^i : fission yield fraction of nuclide i ,

σ_r^i : capture cross section of nuclide i ,

N^i : number density of nuclide i at time t ,

σ_f^i : microscopic fission cross section of nuclide i ,

λ^i : decay constant of nuclide i ,

Φ : neutron flux,

, $b98, 98, b99, 99, 235$ $^{98}\text{Nb}, ^{98}\text{Mo}, ^{99}\text{Nb}, ^{99}\text{Mo}, ^{235}\text{U}$ (6)
(7)

$$\begin{aligned} & \gamma^{99} N^{235} \sigma_f^{235} \Phi + \lambda^{b99} N^{b99} \\ &= \sigma_f \Phi N_0^{235} e^{-\mu t} \left[\gamma^{99} + \frac{\gamma^{b99} \gamma^i}{\mu - \lambda^i} (e^{-(\lambda^i - \mu)t} - 1) + \sum_{j=1}^{i-1} \frac{\gamma^{b99} \gamma^j e^{\mu t} \prod_{k=1}^{j-1} \lambda^k}{\mu - \sum_{k=1}^j \lambda^k} \prod_{k=1}^j (e^{-\lambda^k t} - e^{-(\mu - \sum_{l=1}^k \lambda^l)t}) \right] \quad (9) \end{aligned}$$

$$= \sigma_f \Phi N_0^{235} e^{-\mu t} \gamma_a^{99} \quad (10)$$

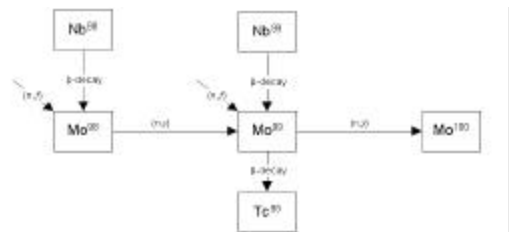
$$= \sigma_f \Phi N_0^{235} \gamma_a^{99} \quad (11)$$

Nb-99 γ_a^{99} , $i = 4$,

Mo-99가 (fission yield) -

(accumulated yield)

(saturation) Mo-98 가



3. Simplified Production-Destruction Scheme of ^{99}Mo in a Fission Moly Target

(accumulated yield fraction), γ_a^{98}

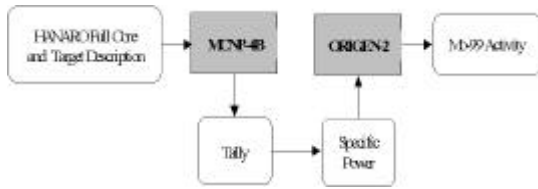
Mo-99

, ^{98}Mo ^{99}Mo 가 $t=0$ $N^{99}(t)$
(12)

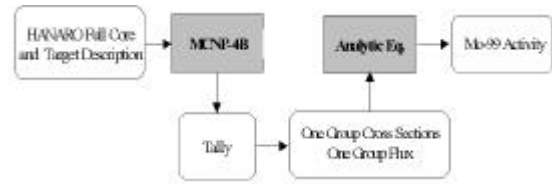
$$\begin{aligned}
N^{99}(t) = & \frac{\gamma_a^{99} N_0^{235} \sigma_f^{235} \Phi}{\lambda^{99} + \sigma_r^{99} \Phi - \sigma_a^{235} \Phi} (e^{-\sigma_a^{235} \Phi \cdot t} - e^{-(\lambda^{99} + \sigma_r^{99} \Phi) \cdot t}) \\
& + \frac{\gamma_a^{98} N_0^{235} \sigma_f^{235} \Phi \sigma_r^{98} \Phi}{(\sigma_r^{98} \Phi - \sigma_a^{235} \Phi) (\lambda^{99} + \sigma_r^{99} \Phi - \sigma_a^{235} \Phi)} (e^{-\sigma_a^{235} \Phi \cdot t} - e^{-(\lambda^{99} + \sigma_r^{99} \Phi) \cdot t}) \\
& - \frac{\gamma_a^{98} N_0^{235} \sigma_f^{235} \Phi \sigma_r^{98} \Phi}{(\sigma_r^{98} \Phi - \sigma_a^{235} \Phi) (\lambda^{99} + \sigma_r^{99} \Phi - \sigma_r^{98} \Phi)} (e^{-\sigma_r^{98} \Phi \cdot t} - e^{-(\lambda^{99} + \sigma_r^{99} \Phi) \cdot t})
\end{aligned} \tag{12}$$

2.3 MCNP-ORIGEN 가

MCNP-ORIGEN Mo-99 가 MCNP-ORIGEN Mo-99 가 MCNP-ORIGEN Mo-99 가



4. MCNP-ORIGEN



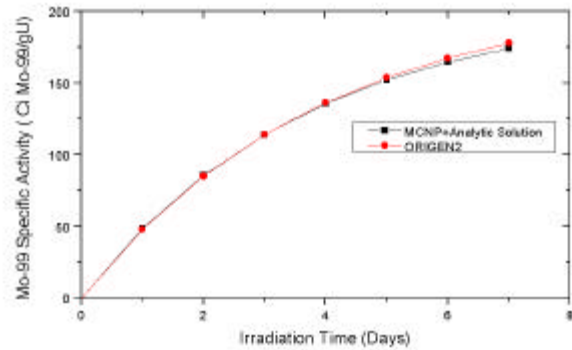
5. MCNP-Analytic Eq.

MCNP-ORIGEN-2 가 , (12) MCNP-4B Mo-99 가 , (biased)

(fission yield fraction) ORIGEN-2

6 MCNP-ORIGEN Mo-99 , MCNP-4B Mo-99 Mo-99 HANARO

ORIGEN-2 Mo-99 1.6% , MCNP-ORIGEN



6. MCNP-Analytic Eq. ORIGEN2 Mo-99 MCNP-

가

3.

가 가 , 가

Mo-99 가 , ,
가 .

1) 가(total reactivity worth) 1.25 %

2) 2.349 MW/m² 가 ,
146.57 가 .
가 .

3) Mo-99 4,000Ci/yr

4) Mo-99 Mo-99 (Ci ⁹⁹Mo/gU) 가 ,
HEU .

5) , ,
.

4. HEU

가
(Surface Heat Flux, SHF), (Recoil-Loss Rate) ,
Mo-99 가 .
가 가 가
가

Mo-99 가
가 가 가
(electro-deposited) 가 .

Cintichem (proven technology) 7

7 OR-3, 5 7.5cm ,
BOC(Bottom 25cm) 가
가 40cm , Mo-99

가 Mo-99가

Mo-99

4.1

가

1
가

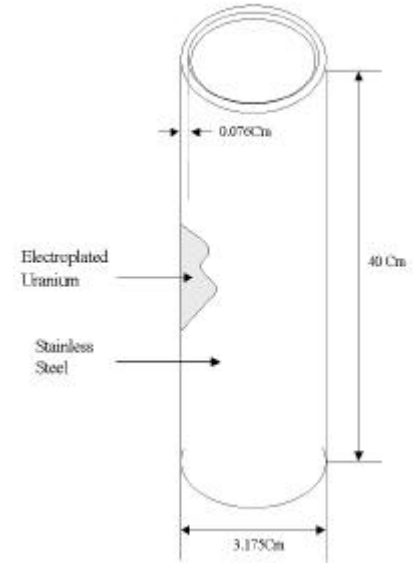
가

가

가

가

가

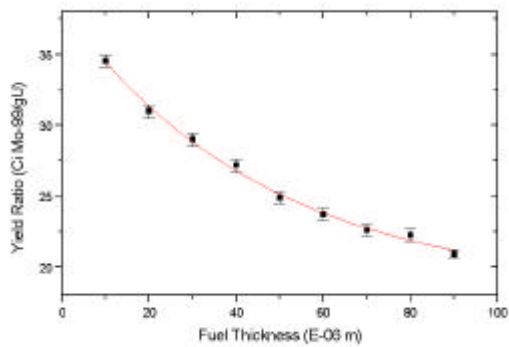


7. HEU Mo-99 Target

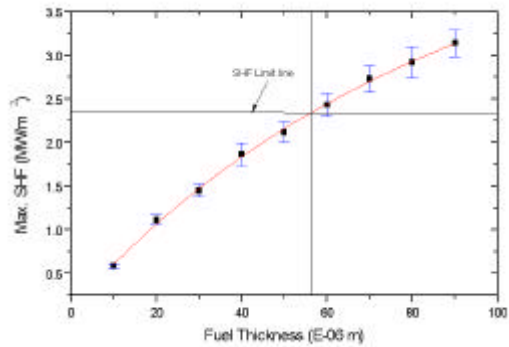
8 가 10 20 μ m 10
 μ m 10.3 \pm 1.83 % 가 가 70 80 μ m 1.6
 \pm 2.5 % 가
 9 가
 60 μ m 2.349 MW/m² 가
 60 μ m 가

1. Mo-99

| Thick. (μ m) | U Loading /target (g) | Yield Ratio (Ci ⁹⁹ Mo/gU) | Max. SHF (MW/m ²) | (%/ 10 μ m) | Max. SHF (%/ 10 μ m) |
|----------------------|-----------------------------|---|----------------------------------|-------------------|-----------------------------|
| 10 | 2.70 | 34.54 \pm 0.4387 | 0.587 \pm 0.0235 | | |
| 20 | 5.39 | 30.99 \pm 0.4525 | 1.113 \pm 0.0548 | - 10.3 \pm 1.83 | 89.6 \pm 10.8 |
| 30 | 8.09 | 29.00 \pm 0.4698 | 1.451 \pm 0.0730 | - 6.4 \pm 2.11 | 30.4 \pm 8.33 |
| 40 | 10.78 | 27.17 \pm 0.4510 | 1.867 \pm 0.1307 | - 6.3 \pm 2.25 | 28.7 \pm 10.4 |
| 50 | 13.47 | 24.88 \pm 0.4155 | 2.120 \pm 0.1168 | - 8.4 \pm 2.26 | 13.6 \pm 9.4 |
| 60 | 16.16 | 23.71 \pm 0.4102 | 2.432 \pm 0.1321 | - 4.2 \pm 2.35 | 14.7 \pm 8.4 |
| 70 | 18.84 | 22.61 \pm 0.3889 | 2.732 \pm 0.1553 | - 4.6 \pm 2.39 | 12.3 \pm 8.4 |
| 80 | 21.53 | 22.24 \pm 0.4114 | 2.920 \pm 0.1723 | - 1.6 \pm 2.50 | 7.0 \pm 8.5 |
| 90 | 24.21 | 20.91 \pm 0.3074 | 3.147 \pm 0.1504 | - 6.0 \pm 2.31 | 7.5 \pm 7.8 |



8. Mo-99



9. SHF

4.2

가 Mo-99
 (Ci ⁹⁹Mo/gU)
 (fission fragment)
 (Recoil) 가
 Mo-99 가
 (Recoil Loss Fraction) 가
 가
 i (recoil range) μ , i (rate of production of
 recoils of species i), p_i(x)가 x 가 ,
 (source term) , q_i(x) (removal)
 ,

$$q_i(x) = \frac{1}{2} \left(1 + \frac{x}{\mu}\right) p_i \quad (\text{for } 0 \leq x \leq \mu) \tag{13}$$

$$= p_i \quad (\text{for } x > \mu) \tag{14}$$
 가 .
 (fission yield fraction) y_i i
 (balance) . λ_iC_i
 (λ_i=decay constant, C_i= concentration of product i)
 (fission fragment)
 (source term) (13) (14) , p_i
 y_iF (F=fission reaction rate), μ μ_{ff} (range of fission fragment)

$$\frac{dC_i(x, t)}{C_i} = \frac{\left(1 + \frac{x}{\mu_{ff}}\right)}{2} \quad (\text{for } 0 \leq x \leq \mu_{ff}) \tag{15}$$

$$= 1 \quad (\text{for } x > \mu_{ff}) \tag{16}$$

$$C_i = \frac{y_i F}{\lambda_i} (1 - e^{-\lambda_i t})$$

(target fuel outer surface)

$$\text{Recoil Loss Fraction} = \frac{S_o}{4V} \left(\mu - \frac{1}{12} r_o \mu^2 \right) \quad (17)$$

where, S_o : target fuel outer surface area

r_o : target fuel outer radius

μ : fission fragment recoil range

가

(17)

가

Mo-99

가

Mo-99

UO₂

가 6 10 μ m

Mo-99

7 μ m

Mo-99

8 μ m 가

10

가

가

가

가

가 10 μ m

20%

4.3

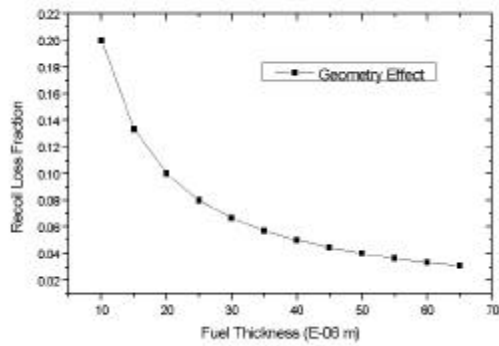
가

Mo-99

가

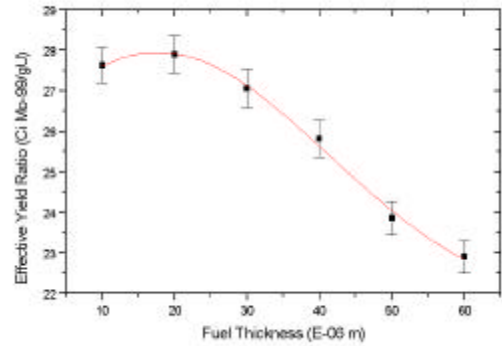
(effective yield)

(Ci ⁹⁹Mo/gU)



10.

Mo-99



11.

11

가

가

가

가

Mo-99

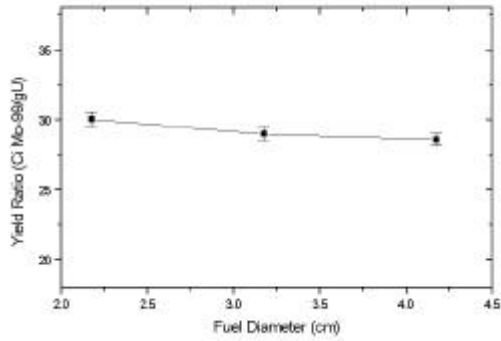
(effective yield ratio)

, 가

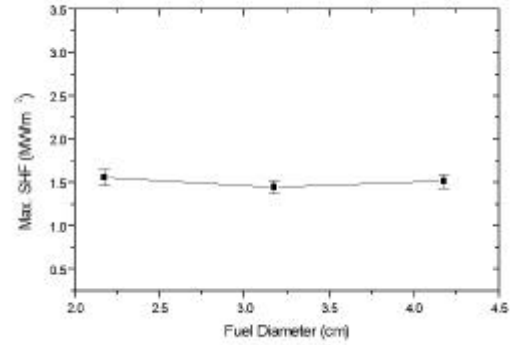
20 μ m

4.4

30 μ m
 가 , Mo-99 SHF 3.175cm \pm 1cm
 , 12 SHF 가 1cm
 2.51 \pm 1.6% SHF
 가 13 Mo-99
 SHF



12.



13.

SHF

4.5

, 2 가
 14.6 \pm 4.2(%)

Cintichem

2. Mo-99

| Thick. (μ m) | Clad. Mat. | Yield ratio \pm (Ci ^{99s} Mo/gU) | (%) |
|-------------------|------------|--|----------------|
| 15 | | 31.95 \pm 0.4100 | 14.6 \pm 2.3 |
| | | 37.40 \pm 0.6171 | |
| 30 | | 29.00 \pm 0.4698 | 13.5 \pm 2.5 |
| | | 33.53 \pm 0.5465 | |
| 60 | | 23.71 \pm 0.3153 | 15.7 \pm 2.4 |
| | | 28.13 \pm 0.4700 | |

4.6

7.3m/sec

OR
 (guide tube)

OR

가
 가 , 3 가
 가 $0.7 \pm 4.0\%$

3.

| Thick.(mm) | Yield ratio \pm (Ci ^{99s} Mo/gU) | (%) | (%) |
|------------|--|------------------|------------------|
| 0.0 | 29.00 ± 0.4698 | | -0.73 ± 1.32 |
| 2.0 | 28.83 ± 0.4613 | -0.59 ± 2.28 | |
| 4.0 | 28.34 ± 0.4563 | -1.73 ± 2.29 | |
| 6.0 | 28.37 ± 0.4709 | -0.11 ± 2.31 | |

4.7

가
 Mo-99 (recoil loss)
 (recoil barrier material)
 $10\mu\text{m}$
 (recoil range)가 $6 \sim 10\mu\text{m}$
 Mo-99 Mo-99
 0% (6)
 가 가
 (Cu), (Fe), (Ni), (Zn)

4 (barrier material)

| Thickness.(mm) | Yield ratio \pm (Ci ^{99s} Mo/gU) | (%) |
|---------------------|--|------------------|
| No barrier material | 29.00 ± 0.4221 | - |
| Zn | No Calculation | - |
| Cu | 28.52 ± 0.4613 | -1.66 ± 2.21 |
| Fe | 28.94 ± 0.3473 | -0.21 ± 2.02 |
| Ni | 28.70 ± 0.4162 | -1.03 ± 2.19 |

4 가 Mo-99
 가 (Zn) 420
 (Cu) 가 1.675 MeV 2.642 MeV
 (Ni) (Fe)

5. HEU

5.1 Cintichem

HEU

가
Cintichem

Cintichem

가

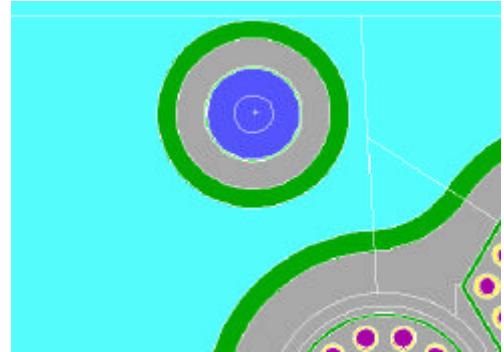
,
304
93w/o U₃O₈

3.175cm,

0.076cm

4.14

HANARO



OR-3 Cintichem

14 Cintichem

5.1.1

5

가
Cintichem

가가

가가 0.2473 ± 0.1334 %

(transient)

가 가

On-power Target Loading

가

5. Cintichem

가

| Axial Length (cm) | Fuel O.D. (cm) | Fuel Thickness (μm) | U Loading (g/target) | OR3, 5 Total U Loading (g) | Reactivity Worth (%) |
|-------------------|----------------|---------------------|----------------------|----------------------------|----------------------|
| 50 | 3.023 | 65 | 21.5 | 43 | 0.2473 ± 0.1334 |

OR hole

(T)

2.6

가

37.6 가

가

Bulk Boiling

ONB(Onset of Nucleate

Boiling) DNB(Departure form Nucleate Boiling)가

Nusselt

(7)

$$N_u = 0.036 R_e^{0.8} P_r^{\frac{1}{3}} (D/L)^{0.055}$$

(18)

6

SHF가

2.349MW/m²

377 ONB

146.57

Cintichem

HANARO OR

6. Cintichem Target SHF

| T (T _{in} - T _{out}) () | Max .SHF (MW/m ²) | Max. Temp. () | | |
|---|----------------------------------|----------------|---------------|---------------|
| | | Clad. | Fuel | |
| | | Outer Surface | Outer Surface | Inner Surface |
| 2.6 | 2.625 ± 0.0735 | 377.12 ± 9.6 | 497.39 ± 10.0 | 508.51 ± 10.0 |

5.1.2

가

Cintichem

가

Cintichem

7 Cintichem

Mo-99

5

6 day reference

가

. OR 2

30,000 Ci

8

0.5 , SUS

가

6kg

0.75 ,

7. Cintichem Target

Mo-99

| Production Yield Ratio ± 1 (Ci ⁹⁹ Mo/gU) | | Annual Production (Ci ⁹⁹ Mo/yr) | |
|--|------------------|---|--------------------|
| Total Production | With Recoil Loss | With 1 OR Hole | With 2 OR Holes |
| 22.35 ± 0.16 | 21.66 ± 0.16 | 14,793 | 29,586 |

8. Cintichem Target

| Uranium (g) | HLW (/yr) | LLW (/yr) | Clad. Amount (g) |
|----------------|---------------|---------------|---------------------|
| 860 | 152.5 | 104.0 | 5,836 |

5.2 HEU

가

9

가

Cintichem

SUS

, 5-1 5-3

UO₂

가

. 5-4

5-5

(Ni)

UO₂

9

, 10 11

5-1 5-3

OR

가

, 50cm

10μm

11μm

UO₂

5-5

가가 0.2911 ± 0.2667 %

137.70 ± 5.3

9. 가

| Case # | Axial Length (cm) | Fuel Thickness (μm) | U Loading (g/target) | OR3, 5 Total U Loading (g) | Reactivity Worth (%) |
|--------|-------------------|----------------------------------|----------------------|----------------------------|---------------------------------------|
| 5-1 | 50 | 10 | 4.58 | 9.15 | 0.0733 ± 0.2578 |
| 5-2 | | 11 | 5.03 | 10.07 | 0.2524 ± 0.2667 |
| 5-3 | | 12 | 5.50 | 10.99 | 0.3344 ± 0.2827 |
| 5-4 | | 10 | 4.58 | 9.15 | -0.0291 ± 0.2558 |
| 5-5 | | 11 | 5.03 | 10.07 | 0.2911 ± 0.2667 |

10. SHF

| Case # | Max. SHF (MW/m^2) | Max. Temp. () | | |
|--------|-------------------------------------|------------------------------------|------------------------------------|------------------------------------|
| | | Clad. | Fuel | |
| | | O.S | O.S | I.S. |
| 5-1 | 0.788 ± 0.039 | 139.05 ± 5.2 | 175.62 ± 5.6 | 176.12 ± 5.6 |
| 5-2 | 0.830 ± 0.043 | 144.67 ± 5.7 | 183.22 ± 6.0 | 183.81 ± 6.0 |
| 5-3 | 0.859 ± 0.050 | 148.54 ± 5.7 | 188.45 ± 6.1 | 189.11 ± 6.1 |
| 5-4 | 0.796 ± 0.040 | 138.74 ± 5.2 | 175.21 ± 5.5 | 175.32 ± 5.5 |
| 5-5 | 0.788 ± 0.041 | 137.70 ± 5.3 | 173.80 ± 5.6 | 173.91 ± 5.6 |

11.

| Case # | 5 days Irradiation | | | |
|--------|---|------------------------------------|--|----------------|
| | Yield Ratio ($\text{Ci } ^{99}\text{Mo}/\text{gU}$) | | Production ($\text{Ci } ^{99}\text{Mo}/\text{yr}$) | |
| | produced | effective | wiht 1 OR hole | with 2 OR hole |
| 5-1 | 30.54 ± 0.47 | 23.956 ± 0.47 | 3,519 | 7,022 |
| 5-2 | 29.93 ± 0.46 | 24.025 ± 0.46 | 3,872 | 7,745 |
| 5-3 | 29.47 ± 0.46 | 24.099 ± 0.46 | 4,237 | 8,475 |
| 5-4 | 30.66 ± 0.47 | 30.66 ± 0.47 | 4,421 | 8,842 |
| 5-5 | 30.48 ± 0.47 | 30.48 ± 0.47 | 4,834 | 9,669 |

12 5-4 5-5 가
 32.5 , 22.2 , 6kg
 가

12.

| Case # | Uranium (g) | HLW (/yr) | LLW (/yr) | Clad. Amount (g) |
|--------|-------------|------------|------------|------------------|
| 5-4 | 183.2 | 32.5 | 22.2 | 5,836 |
| 5-5 | 202.4 | 35.9 | 24.5 | 5,836 |

5.3 가

가 , Cintichem 1.4 ,
 Cintichem 4 .

6.
 MCNP-ORIGEN MCNP- 가 , Mo-99
 1.6% 가 .

가 ,
 가 ,
 20 μ m 가 .

Cintichem HANARO
 가 , (Ni) , 50cm, UO₂
 11 μ m , Cintichem
 4 .

Acknowledgements

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