

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
 가 , (Ni)
 , 50cm, UO₂ 11 μ m
 , 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)

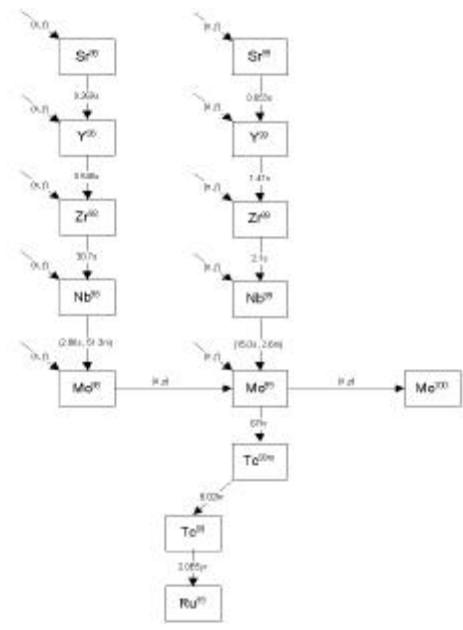
2. MCNP-ORIGEN2

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 Y⁹⁸, Zr⁹⁸, Nb⁹⁸
 - Mo-98 ⁹⁸Mo(n,
)⁹⁹Mo . Mo-99
 , U-235
 (direct fission yield)
 가
 (half-life) 가
 143keV ^{99m}Tc
 , ⁹⁹Mo(n,)¹⁰⁰Mo

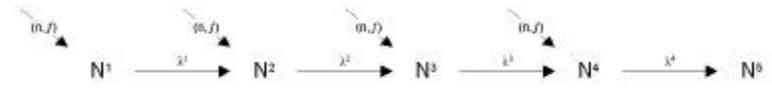


67

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 -

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

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

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

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

, 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

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

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

, 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,

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

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

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

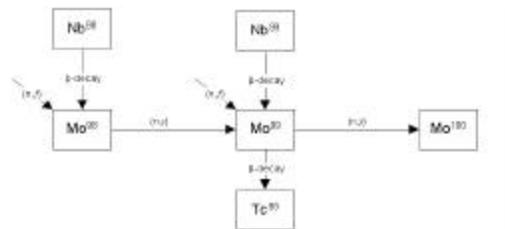
$$= \sigma_f \Phi N_0^{235} \gamma_a^{99} \quad (11)$$

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}

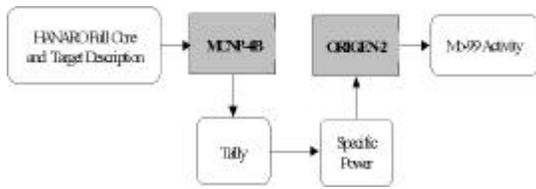
Mo-99

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

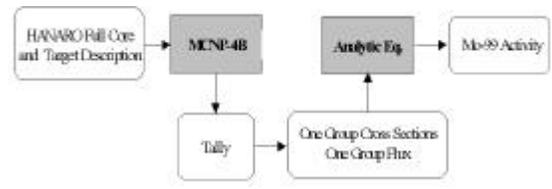
$$\begin{aligned}
N^{99}(t) = & \frac{\gamma_a^{99} N_0^{235} \sigma_f^{235} \Phi}{\lambda^{99} + \sigma_r^{99} \Phi - \sigma_a^{235} \Phi} (e^{-\sigma_a^{235} \Phi \cdot t} - e^{-(\lambda^{99} + \sigma_r^{99} \Phi) \cdot t}) \\
& + \frac{\gamma_a^{98} N_0^{235} \sigma_f^{235} \Phi \sigma_r^{98} \Phi}{(\sigma_r^{98}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

2.3 MCNP-ORIGEN 가

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



4. MCNP-ORIGEN



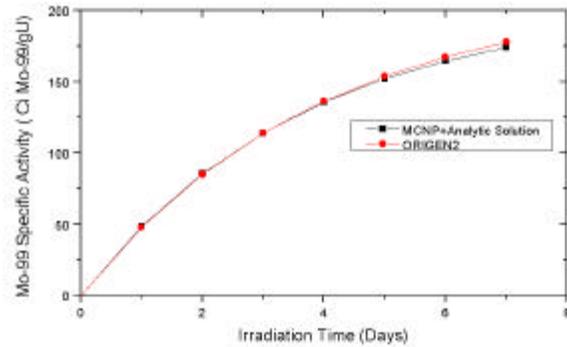
5. MCNP-Analytic Eq.

MCNP-ORIGEN-2 가 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-

3.

가 가 , 가

Mo-99 가 , , 가

1) 가(total reactivity worth) 1.25 %

2) 2.349 MW/m² 가 , 146.57 가

3) Mo-99 4,000Ci/yr

4) Mo-99 HEU Mo-99 (Ci ⁹⁹Mo/gU) 가 ,

5) , ,

4. HEU

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

Mo-99 가 가 가

Mo-99 가 가

가 가 가

(electro-deposited) 가 , (proven technology)

Cintichem 7

7 OR-3, 5 7.5cm ,

BOC(Bottom 25cm) 가 Mo-99 가 40cm ,

가 Mo-99가

Mo-99

4.1

가

1
가

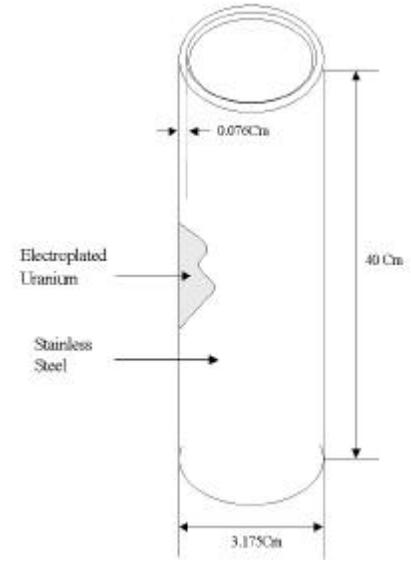
가

가

가

가

가

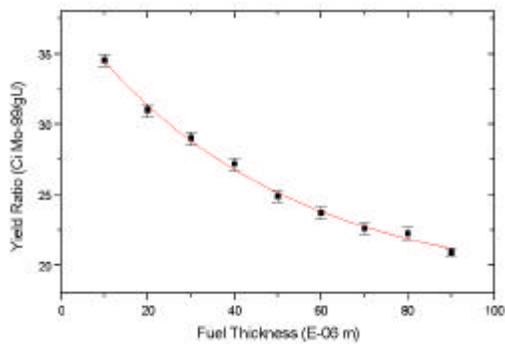


7. HEU Mo-99 Target

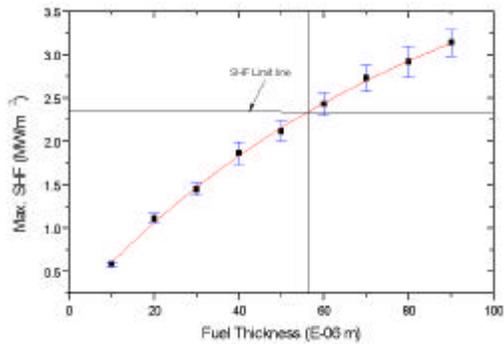
8 가 10 20 μ m 10
 μ m 10.3 \pm 1.83 % 가 가 70 80 μ m 1.6
 \pm 2.5 % 가
 9 가
 60 μ m 2.349 MW/m² 가
 60 μ m 가

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 , (removal)
 (source term) , q_i(x)

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

$$= p_i \quad (\text{for } x > \mu) \tag{14}$$
 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

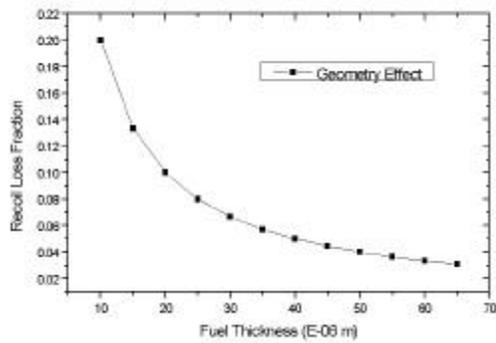
가

Mo-99

가

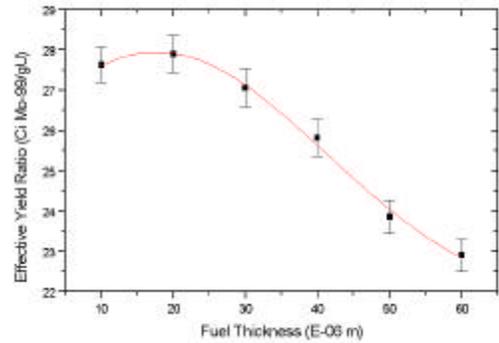
(effective yield)

(Ci ⁹⁹Mo/gU)



10.

Mo-99



11.

11

가

가

가

가

Mo-99

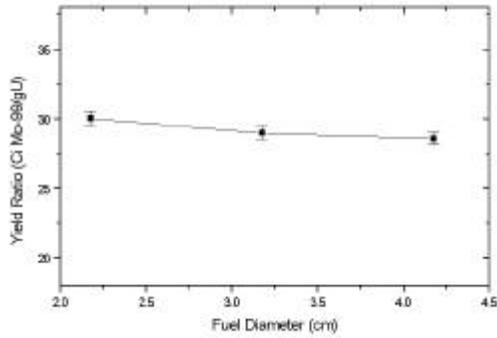
(effective yield ratio)

, 가

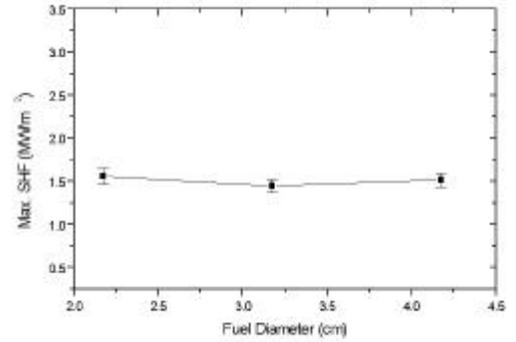
20 μ m

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 0.75 ,
0.5 , SUS 가 6kg

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	with 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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