UNCERTAINTY ANALYSIS FOR QUALITY CONTROL PROCEDURE IN FISSION MO-99 PRODUCTION

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ABSTRACT

Uncertainties of response parameters such as ^{99}Mo yield ratio, ^{239}Pu yield ratio, annual ^{99}Mo production rate, and minimum required decontamination factor were evaluated for fission Mo production in HANARO reactor with two kinds of fuel options, high enriched uranium and low enriched uranium fuel target. In this study, the reactor physics parameters of reactor core, manufacturing tolerance and composition tolerance of target were considered as sources of uncertainty. Crude Monte Carlo method was applied in a data sampling step. It was shown that there is no critical process to be controlled in case of HEU target option. Uncertainties of response parameters were relatively small. However, the inherent uncertainty from current fabrication process for LEU fuel film should be reduced for acceptable quality in Mo production. Uncertainty in annual production rate induced by this parameter showed about 20% in 95% confidence level.

1. INTRODUCTION

Molybdenum-99 is a parent isotope of technetium-99m which has been widely used in diagnostic medical imaging procedures.[1] Commercial production of ^{99}Mo, however, has been almost exclusively dependent upon Canadian company Nordion. Therefore, a need for a reliable regional supply facility had been recognized worldwide. Korea Atomic Energy Research Institute has been developed molybdenum-99 production technology with 30MW$_t$ reactor, HANARO as a national R&D program since 1997.

Current ^{99}Mo suppliers have used high enriched uranium (HEU) fuel target because of high ^{99}Mo yield ratio (Ci ^{99}Mo/gU) and low impurity level of α-emitters. RERTR, however, recommends the use of low enriched uranium (LEU) as a fuel or irradiation target in a research reactors.[2,3] It is expected that LEU has the larger uncertainties in the amount of Pu impurity and ^{99}Mo, because the LEU contains the 70 to 80 times as much ^{238}U as HEU and produces 30 to 50 times as much Pu in order to derive equivalent amount of Mo-99. Only if we know variation of production amount of Mo-99 and Pu-239 within the production procedure line for fission-induced molybdenum-99, we can achieve a high level of quality control procedure.

In this paper, the most important process for better quality control in a fission Mo production was derived by evaluating the uncertainties of the response parameters, such as Mo-99 yield ratio (Ci^{99}Mo/gU), Pu-239 yield ratio (Ci^{239}Pu/gU), annual production rate (Ci^{99}Mo/yr), and minimum required decontamination factor (MRDF) satisfying the U.S.P. standards (10^{-7}μCi^{239}Pu/mCi^{99}Mo). Section II discusses the analysis tool for better application to evaluate the exact isotope amount to the time. Section III represents the methodology to describe the equilibrium core that would be a reference condition for a target design and uncertainty analysis. Section IV is allocated to the results and discussion of uncertainty analysis on the performance of LEU and HEU target induced by the variation of irradiated conditions. Section V presents the summary and conclusions.

2. EVALUATION OF ANALYSIS TOOL

Uncertainty analysis performed in this work is based on the uranium fuel target irradiation in HANARO reactor that is built in Taejon as a multipurpose research reactor. As shown in Fig. 1, target will be irradiated at outer core region far from the center of the core, inside the heavy water reflector tank. Because outer region is supplied with neutrons from inner core, neutron spectrum in this region is shifted to thermal range compared with that in the core. Therefore, deterministic code system with few-group cross section library and diffusion theory is not reliable for the evaluation of the fission reaction rate in the target. In this study, full core 3-D calculation of MCNP-4/B code is done for the exact evaluation of fission power rate at the target. ORIGEN2 code was also used for the evaluation of isotope production and decay transient throughout the whole processes including post-irradiation chemical treatments.

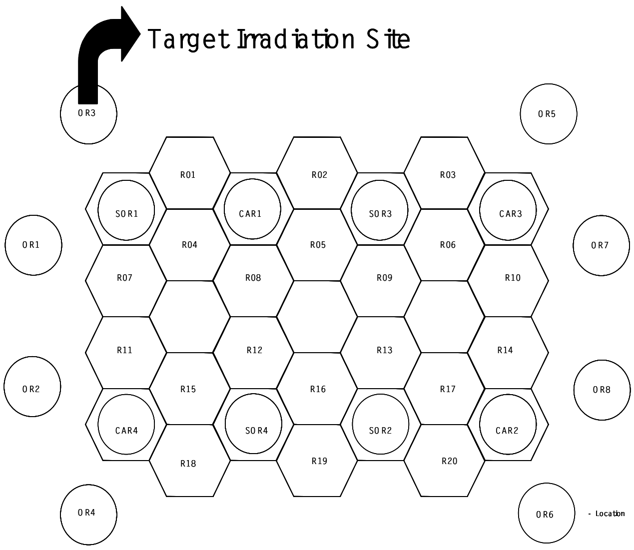

Fig. 1 HANARO Core and Target Irradiation Site

Production-destruction schemes of Mo-99 and Pu-239 were searched and characteristics of these schemes were analyzed to evaluate exact isotope amount by MCNP/ORIGEN2 code system. From this analysis, the amounts of ^{99}Mo and ^{239}Pu evaluated with modified cross section library of σ_f^{235}, σ_r^{238} were agree well with the results with library including the all modified cross section associated with production-destruction chain of ^{99}Mo and ^{239}Pu within 0.3% difference. Therefore, only these two cross sections generated by MCNP were modified, whenever the amount of isotope was evaluated.

3. TARGET DESIGN OPTIMIZATION FOR EQULIBRIUM CORE

3.1 CORE MODELING METHODOLOGY

An Equilibrium core was selected and modeled as a reference core for target design optimization and uncertainty analysis. To perform an analysis with the MCNP code, number density of all isotopes and neutron source distribution should be defined. An equilibrium burnup core condition was defined with WIMS/VENTURE design methodologies. Fig. 2 shows schematic diagram for equilibrium core modeling procedure proposed in this work for MCNP calculation. The isotope number density and source density to interest region of core geometry in MCNP was retrieved by the results of WIMS/VENTURE system, as shown in fig. 2.

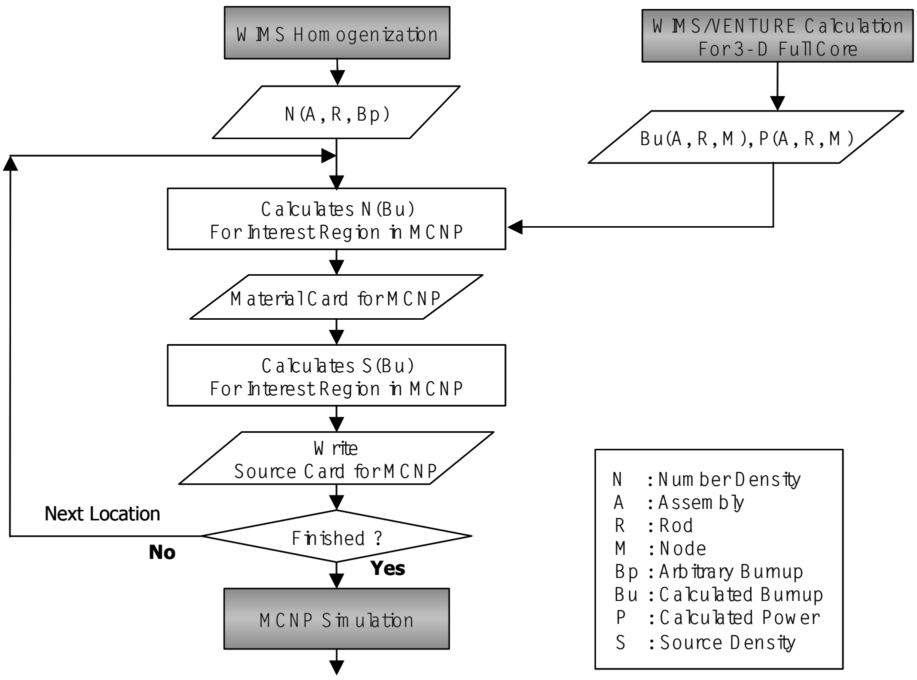

Fig. 2 Core Modeling Flow Chart by WIMS/VENTURE and MCNP

Relative power distribution calculated by MCNP was compared with the results calculated by WIMS/VENTURE code system. The calculated value from each code system is described in Fig. 3. As shown in Fig.3, MCNP model for equilibrium core was shown to be reliable, because root mean square error was small within 2.5%.

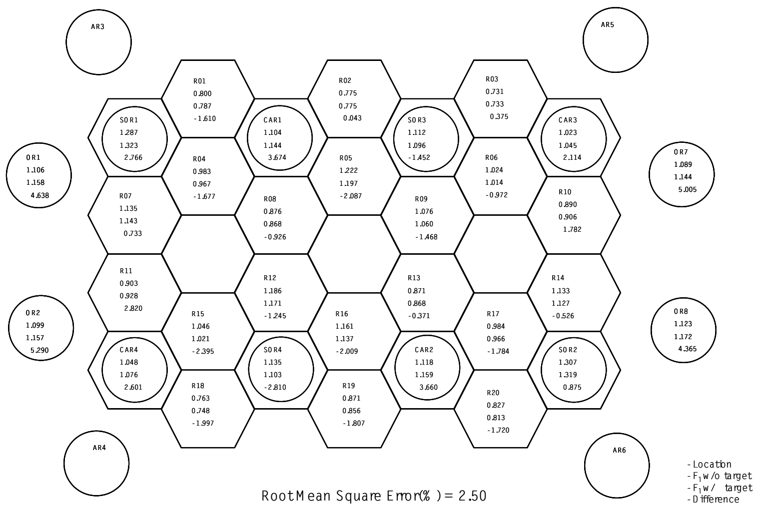

Fig. 3 Relative Power Distribution by WIMS/VENTURE and MCNP at Middle of Cycle

3.2 TARGET DESIGN OPTIMIZATION

The design optimization for LEU and HEU was performed. The proposed targets are as follows.

3.2.1 Proposed HEU Target

Design parameters such as axial irradiation location, fuel thickness, radius, axial length, the number of target, ^{99}Mo recoil loss, and various fuel material were analyzed and technical issues such as target loading to core, and target transportation from reactor building to hot cell under on-power reactor condition were considered for design optimization. The dimension and evaluated parameters of designed HEU target are described in Table 1 and Table 2, respectively. This target uses 93w/o enriched UO_2 fuel electrodeposited onto Ni recoil-barrier which is electrodeposited onto inner surface of SUS-304. The integrity of this target was proved through calculation for HANARO.[4]

3.2.2. Proposed LEU Target

The dimension and evaluated parameters of designed LEU target are also described in Table 1 and Table 2, respectively. This target uses 19.75w/o enriched U metal fuel embeded in both Al-cladding. The Ni recoil-barrier is coated in inner surface of outer cladding and in outer surface of inner cladding. This design reflects the concept proposed by Argonne National Laboratory to supply the world with LEU target.[5,6] For this target, further optimization will be required but modification will be so minor that this can't affect the results of uncertainty analysis.

Table 1 Design Parameters of Optimized HEU and LEU target

Target Fuel	Axial Length (cm)	Target Cladding		Thickness(μ m)			U Loading (g/target)
		Outer Tube O.D.(cm)	Inner Tube I.D.(cm)	Clad.	Recoil Barrier	UO_2 Fuel	
HEU	15	4.32	-	800	10	10	3.786
LEU	10	4.40	4.088	1500	10	100	24.40

Table 2. Performance of Designed HEU and LEU Target

Target Fuel	Reactivity Worth $(\%\Delta\rho \pm 1\sigma)$	Yield Ratio $(Ci^{99}Mo/gU)$	Production $(Ci^{99}Mo/yr)$	Max. Clad. Temp.($^{\circ}C$)	Δ T($^{\circ}C$) (Outlet-Inlet)
HEU	0.0135±0.0135	32.37	4042	86.6	0.6
LEU	0.0761±0.0158	6.967	5440	88.8	1.05

4. UNCERTAINTY ANALYSIS ON FISSION MO TARGET

The many input parameters are used to evaluate the isotope amount to the time and they mean the actual value for ^{99}Mo production. These parameters, however, have uncertainty themselves. The sources of uncertainty for response parameters in ^{99}Mo production are as follows; The first, reactor physics parameters induces the uncertainty. This source of uncertainty arises from inaccurate core model in MCNP and insufficient consideration of all reactor condition when response parameter was evaluated. The second, manufacturing tolerance of fabrication and composition induce the uncertainty.

These sources of uncertainty arise from inaccurate target dimension and material composition. The third, chemical processing tolerance induces the uncertainty. This source of uncertainty arises from the condition of chemical experiment. The uncertainty in this study means that the scatter of response parameters, such as Mo-99 yield ratio, Pu-239 yield ratio, annual production rate, and MRDF satisfying the U.S.P. standards, induced by input parameters variation previously mentioned. Hereafter, we refer to these parameters as response parameters. The uncertainty caused by nuclide number density calculated by WIMS/VENTURE code system, MCNP code bias and chemical processing are not included in this study because it should be evaluated by comparison with experimental data.

4.1 DETERMINATION OF INPUT PARAMETERS

4.1.1 Reactor Physics Parameters

For proper simulation of neutron from core to irradiation site in MCNP code, exact generation of neutron source is indispensable. The neutron source generated in fuel region *i*, and energy interval *g* is expressed by

$$\begin{aligned} S_{ig} &= \sum_j [\chi_{ig}]^j C_i^j P_i \\ &= \sum_j [\chi_{ig}(\nu / E_r)]_i^j P_i \\ &= \sum_j [\chi_{ig}]^j \left([\nu / E_r]_i^j \frac{[\Sigma\phi]_i^j}{\sum_k [\Sigma\phi]_i^k} \right) P_i = \sum_j [\chi_{ig}]^j S_i^j P_i \end{aligned} \qquad (1)$$

where,

i = fuel region index,
j, k=fissile nuclide,
g = certain energy interval index,
χ_{ig}= fission source spectrum,
P_i = node power,
$C_i(\nu/E_r)$ = power-to-source conversion factor, C factor,
E_r = recoverable energy.[7]

If the parameters in eq. (1) were considered exactly, uncertainty from biased neutron source will be negligible. In this section, we elaborate on the uncertainties associated with terms of the source formulation.

A) Fission Source Spectrum

To produce the source exactly based on the eq. (1), fission source spectrum χ^j and S^j should be applied considering the fission reaction rate by each fissile isotopes in each region. Since fission spectra of ^{235}U is different from that of ^{239}Pu, determination of the energy of emitting source for each region should consider the ratio of fissile nuclides. It was possible to apply the S^j properly. However, it was difficult to do so for χ^j, and subsequently it was not applied for each region but for each assembly in MCNP code, as shown in eq. (2).

$$S_{ig} = \sum_j [\chi_{lg}]^j S_i^j P_i \qquad (2)$$

where, *l* denotes the each fuel assembly. Therefore, the uncertainty from fission source spectrum arose.

B) MCNP Source Density Modeling

To produce the neutron source properly for each energy interval and for each region, P_i in eq (1) should be exact value. However, P_i that was derived from WIMS/VENTURE code system had the uncertainty itself. This fact induces the uncertainty of response parameters.

C) Fissile Consumption in Target

In MCNP/ORIGEN code system, it was assumed that the fission reaction rate during 5-days irradiation was treated as a constant value, when nuclide isotope was evaluated. The fissile material in a target is reduced by nuclear fission, however, which makes the fission reaction rate decrease during the irradiation period. Inappropriate consideration of this phenomenon in MCNP/ORIGEN2 code system induces the uncertainty of response parameters.

D) Variation of Reactor Power Level

A absolute reactor power was assumed with 30MWt in MCNP/ORIGEN2 system. It is difficult to maintain the reactor 30MWt power, and subsequently the power level vibrates from the 30MWt power. This fact induces the uncertainty.

4.1.2 Manufacturing Tolerance of Target

If target were not fabricated in accordance with the dimension that was optimized, the variation of response parameters would be expected. The fabrication tolerances which should be considered are target fuel thickness and axial length of the fuel, and cladding. It is also difficult to make the material composition the same with designed value. For example, it is expected that the enrichment of the LEU fuel should be made 19.75w/o. The material with biased enrichment makes the response parameters have undesired value.

4.2 SCREENING BY SENSITIVITY ANALYSIS

It is difficult to perform the uncertainty analysis considering all parameters previously mentioned. The screening of input parameters whose effect on uncertainty of response parameters was negligible was done as a first step of uncertainty analysis. The screened parameters were thickness, axial length and composition variation of Al cladding in LEU target. The thickness, the axial length and composition variation of SUS cladding used in fabrication of HEU target were also eliminated. The uncertainty from these parameters was small enough to be neglected.

4.3 QUANTIFICATION OF INPUT PARAMETERS

The distribution of each parameter was assessed. The fission reaction rates by each fissile nuclides for each node were calculated to evaluate the uncertainty from improperly applied fission source spectrum. The distribution of fission reaction rate induced by Pu for each assembly was deduced from WIMS/VENTURE calculation. This distribution didn't indicate certain formal distribution and was assumed uniform distribution, because this distribution makes uncertainty conservative. The distribution of P_i in eq. (1) was assumed to have normal distribution with 4.5%(1σ) uncertainty according to final safety analysis report of HANARO. This means that relative assembly power calculated by WIMS/VENTURE can vibrate from exact value with ±4.5% difference for 1σ level. The distribution of reactor power level was derived with the minimum value of 97.4% and the maximum value of 102.6% of 30MWt. This data is based on the actual operating experience. The variation of fissile nuclides for irradiation period is described in Table 3. In Table 4 and 5, the fabrication tolerance and composition tolerance are described based on experience data, respectively.

The distributions associated with these parameters are assumed with uniform distribution, because it makes the uncertainty larger.[8]

Table 3. The Variation of fissile Nuclide for Irradiation Period

Enrichment	Amount of Fissile Nuclide in Target (#/barn-cm)		
	Min. Value	Max. Value	Median Value
HEU	2.197E-02	2.296E-02	2.247E-03
LEU	9.180E-03	9.613E-03	9.397E-03
Distribution	Uniform	Uniform	Uniform

Table 4. A Distribution of Input Parameters with Fabrication Tolerance

		HEU			LEU		
		Min.	Max.	*Median*	Min.	Max.	Median
Fuel	Thickness(μm)	19	21	20	80	120	100
	Axial Length(cm)	14.5	15.5	15	9.5	10.5	10
Cladding	Thickness(μm)	700	900	800	-	-	-
	Axial Length(cm)	16.5	17.5	17	-	-	-
Distribution		Uniform			Uniform		

Table 5. A Distribution of Input Parameters for Each Fuel

Material	Concentration [w/o]					
	U-235			U-238		
	Min.	Max.	Median	Min.	Max.	Median
HEU	92.75	93.25	93.0	8.75	9.25	9.0
LEU	19.50	20.00	19.75	80.00	80.50	80.25
Distribution	Uniform			Uniform		

4.4. UNCERTAINTY ANALYSIS METHODOLOGY

The Crude Monte Carlo method was applied as a uncertainty analysis methodology.[8,9] The input variable set is composed by random sampling for each input parameter, and then this is applied for the complicated computer model. Repeating this procedure, we can obtain the exact distribution of response parameter. Therefore, it is sometimes treated as a true value in the benchmark calculation. However, this methodology has the disadvantage that the main source of uncertainty can't be deduced. Therefore, to clarify the main source of uncertainty, uncertainty analysis from each input parameters should be performed. When we make input variable set of power distribution (P_i) in this work, permission/rejection method was applied to compensate for correlation of each assembly. The only input variable set which makes the sample mean the average linear power rate is applied for computer model. In Table 6, the #1 is rejected and #2 is accepted based on this method.

Table 6. Permission/Rejection Sampling Chart

Variable / Sampling Set	X_1	X_2	X_3	~	X_k	$\frac{1}{k}\sum_{j=1}^{n}\sum_{i=1}^{k} X_{ij}$
#1	X_{11}	X_{21}	X_{31}	-	X_{kn}	$1.015\times \overline{X}$
#2	X_{12}	X_{22}	X_{32}	-	X_{k4}	$1.000\times \overline{X}$
#3	X_{13}	X_{23}	X_{33}	-	X_{k3}	$0.998\times \overline{X}$

If the many response parameters is calculated, the confidence interval is quantified by eq (3) with the weighting of the statistical error.

$$\overline{C} - T_{95,95}S_c \le X \le \overline{C} + T_{95,95} \qquad (3)$$

where, $\overline{C} = \dfrac{\sum_{i=1}^{n} \dfrac{C_i}{S_{C_i}^2}}{\sum_{i=1}^{n} \dfrac{1}{S_{C_i}^2}}$, $S_c = \dfrac{n}{n-1}\left(\dfrac{\sum_{i=1}^{n}\left(\dfrac{C_i}{S_{c_i}}\right)^2}{\sum_{i=1}^{n}\left(\dfrac{1}{S_{c_i}}\right)^2} - \overline{C}^2 \right)$,

and, $T_{95,95}$ represents two-sided tolerance limit factor.[10]

4.5. RESULTS

Table.7 shows results of uncertainty analysis on each response parameter through the simulation of 100 input variable sets for LEU and HEU target, respectively. The distribution of each response was shown to satisfy the normal distribution by Kolmogorov-Smirnov goodness of fit. In the analysis of HEU, the 95% confidence interval to Mo-99 yield ratio, Pu-239 yield, annual production rate, and MRDF from total input parameter variation was shown to be 29.70□34.3Ci^{99}Mo/gU, 3,450□4,256 Ci^{99}Mo/yr, 2.564E-6□3.504E-6Ci^{239}Pu/gU and 179□237, respectively. This shows the uncertainty of 6%, 9%, 13%, and 12% in 2σ level for each response parameter. In the analysis of LEU, the 95% confidence interval to Mo-99 yield ratio, Pu-239 yield, annual production rate, and MRDF from total input parameter variation was shown to be 6.23□7.70Ci^{99}Mo/gU, 4047□6382Ci^{99}Mo/yr, 1.381E-5□ 1.900E-5Ci^{239}Pu/gU and 4834□5596, respectively. This also shows the uncertainty of about 10%, 20%, 14%, and 7% 2σ level for each parameter.

The demand of fission-produced Mo-99 is constant during a relatively short period, namely, a half of one year, and this means stable and reliable supply of Mo-99 is crucial. A larger uncertainty induces overproduction to ensure the certain confidence of supply. This is because the 95% confidence interval of annual production rate of Mo-99 with 4,000□4,200Ci is superior to the interval of 4,000□ 5,000Ci. The latter case means that overproduction of 5,000Ci may be produced to supply the 4,000Ci/yr of Mo-99 with the 95% confidence level. From this point of view, LEU has twice uncertainty as large as HEU. This fact makes the economics worse. The ^{239}Pu is fissile material. Its large uncertainty can make disposal facility larger. The uncertainty of ^{239}Pu yield ratio of LEU and HEU target was shown to be 13.0% and 14%, respectively. Though the difference between the uncertainty of LEU and that of HEU target is small, the standard deviation of LEU target is 6 times larger as HEU target. Considering the ^{238}U loading amount it represents the about 30 times larger uncertainty as HEU target. Since the production amount of Pu in LEU and HEU target was shown to be small quantitatively, the larger uncertainty of LEU doesn't affect on the economics of waste disposal. If the decontamination factor (DF) of Pu for each step, namely, dissolution/precipitation, purification I, and purification II in Cintichem process were 10, the total DF would be deduced about 1000.

Table 7. Results of Uncertainty Analysis

Target	Response Parameter	Mean	Std.	2.5%	97.5%	Rel. Err.(%) (2σ level)
HEU	$^{c}Mo-99(Ci/gU)$	145.448	5.267	133.668	157.228	7.2
	$^{c}Pu-239(Ci/gU)$	3.0329E-6	2.1130E-7	2.5602E-6	3.5056E-6	13.9
	$^{c}MRDF$	207.326	12.202	180.030	234.623	11.8
	$^{R}Mo-99(Ci/gU)$	32.0583	1.1585	29.467	34.650	7.23
	$^{R}Production\ Rate\ (Ci^{99}Mo/yr)$	3869.72	178.87	3467.35	4272.09	9.24
LEU	$^{c}Mo-99(Ci/gU)$	31.591	1.497	28.247	34.934	9.48
	$^{c}Pu-239(Ci/gU)$	1.6402E-5	1.1620E-6	1.3807E-5	1.8998E-5	14.17
	$^{c}MRDF$	5215.32	170.60	4834.19	5596.44	6.54
	$^{R}Mo-99(Ci/gU)$	6.9627	0.3299	6.2255	7.6996	9.48
	$^{R}Production\ Rate\ (Ci^{99}Mo/yr)$	5215.67	521.987	4049.55	6381.79	20.02

C: The value evaluated at chemical processing, R : The value evaluated at 6-day reference

Therefore, it was shown that HEU has no problem considering the uncertainty of MRDF. In the case of LEU, however, more purification would be required considering the uncertainty. But, the uncertainty of LEU is only 6% in 95% confidence level, this fact means that the quality of $^{99}Mo/^{99m}Tc$ generator would not vary. Therefore, it was shown that clarification of main source of annual production rate only for LEU was needed. And then, the uncertainty caused by each input parameters was evaluated. The main source of uncertainty to Mo-99 revealed to be the fuel film thickness. The uncertainty from this parameter was shown to be 17% in 95 % confidence level.
On the basis of above results, we can conclude that there is no process whose uncertainty should be reduced in the viewpoint of quality control procedure for HEU target. Because current fuel film fabrication technology gives rise to large uncertainty in LEU target, however, technology to reduce the variation of thickness should be developed.

CONCLUSIONS

The uncertainty analysis for better quality control in fission Mo production was performed. In the analysis for HEU target, the uncertainties of response parameters such as Mo-99 yield ratio($Ci^{99}Mo/gU$), Pu-239 yield ratio($Ci^{239}Pu/gU$), annual production rate($Ci^{99}Mo/yr$), were small within 5% for 1σ level. In the analysis for LEU target, the result of uncertainty analysis indicates that LEU has twice uncertainty as large as HEU, which makes the economics worse. And, more purification would be required considering the uncertainty because the 95% confidence interval of

MRDF was 4834□5596. But, the uncertainty of LEU is only 6% in 95% confidence level, this fact means that the quality of ^{99}Mo/^{99m}Tc generator would be ensured in clinical procedure. From these results, it is concluded that there is no process to be seriously controlled for HEU target. However, the inherent uncertainty of current fabrication process of LEU fuel film should be reduced for better quality of Mo production.

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