Space and Time Distribution of Pu Isotopes inside The First Experimental Fuel Pin Designed for PWR and Manufactured in Indonesia

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Abstract. The first short fuel pin containing natural UO_2 pellet in Zry4 cladding has been prepared and planned to be tested in power ramp irradiation. An irradiation test should be designed to allow an experiment can be performed safely and giving maximum results of many performance aspects of design and manufacturing. Performance analysis to the fuel specimen shows that the specimen is not match to be used for power ramp testing. Enlargement by 0.20 mm of pellet diameter has been proposed. The present work is evaluation of modified design for important aspect of isotopic Pu distribution during irradiation test, because generated Pu isotopes in natural UO2 fuel, contribute more power relative to the contribution by enriched UO2 fuel. The axial profile of neutrons flux have been chosen from both experimental measurement and model calculation. The parameters of ramp power has been obtained from statistical experiment data. A simplified and typical base-load commercial PHWR profile of LHR history has been chosen, to determine the minimum irradiation time before ramp test can be performed. The data design and Mat pro XI materials properties models have been chosen. The axial profile of neutrons flux has been accommodated by 5 slices of discrete pin. The Pu distribution of slice-4 with highest power rate has been chosen to be evaluated. The radial discretion of pellet and cladding and numerical parameter have been used the default best practice of TU. The results shows that Pu 239 increased rapidly. The maximum burn up of slice 4 at upper the median slice, it reached nearly 90% of maximum value at about 6000 h with peak of 0.8%a Pu/HM at 22000 h, which is higher than initial U 235. Each 240, 241 and 240 Pu grows slower and ends up to 0.4, 0.2 and 0.18 % respectively. This results can be used for verification of other aspect of fuel behavior in the modeling results and also can be used as guide and comparison to the future post irradiation examination for Pu isotopes distribution.

1. Introduction

Experimental Fuel Elements Installation (EFEI) re-vitalization that began be built late 20th century needs to be improved and with future goals change of EFEI from only laboratory for CIRENE BHWR (Boiling Heavy Water Reactor) fuel technology Power Reactor Fuel type of Cirene, to gradually improved its ability of preparation of various specimens of fuel elements from different types of reactors, PHWR (Pressurized Heavy Water Reactor), especialy CANDU then in the end also will be one the first factory-owned BATAN to produce Power Reactor Fuel type of CANDU, if Candu type nuclear power plants have been later. Preparation direction has been initiated, and at times can realisa thereof can be started, if desired [1].

The first short fuel pin containing natural UO_2 pellet in Zry4 (Zircaloy-4) cladding has been prepared at the CNFT. The PRTF installed inside RSG-GAS (*Reaktor Serba Guna G.A.Siwabessy*) multi purpose 30 MW reactor has been revitalized and is ready for irradiation of fuel pin speciment. An irradiation test should be designed to allow the experiment can be performed safely and giving maximum results of many performance aspects of design and manufacturing. The present work is evaluation of important isotopic Pu generation-depletion and distribution during irradiation test, as Pu generated Pu during irradiation in natural UO2 fuel gives more power fraction than fuel containing enriched UO2. The constraint of axial profile of neutrons flux and the maximum generated integral power have been determined from both experimental measurement and modeling calculation. The contraint of maximum ramp power has been obtained from measured experiment data [2]. As natural uranium is used for fuel pellets, a simplified and typical base-load commercial PHWR profile of LHR (Linear Heat Rate) history has been chosen in determining the minimum irradiation time before ramp test can be performed. The obtained design and manufactured fuel pin data and measured and model material properties data and behavior and component have been used as input.

The natural-UO2 fuel geometry is in between former design of PWR fuel and new design of PHWR fuel. The plan of irradiation test will be at PWR coolant temperature & pressure and thermal neutron flux of PWR.

The chosen model takes into account mat-pro XI [3] materials properties models, model for isotopes generation and depletion but ignores the period of power shut-down and restart-up. Typical high power rate has been chosen to be evaluated. The axial profile of neutrons flux has been accommadated by 5 slices of discret pin. The radial discretizatioon of pellet and cladding and numerical parameter have been used the default best paractice of TransUranus [4] code used. The code has been veried by diference theoretical and computation modeling and experiment data [5] The results of time distribution of Pu 239 increased rapidly. The maximum burnup of slice 4 at upper the median slice, it reached nearly 90% of maximum value at about 6000 h with peak of 0.8%a Pu/HM at 22000 h, wich is higher than initial U 235. Each 240, 241 and 240 Pu grows slower and ends up to 0.4, 0.2 and 0.18 % respectively. The axial distribution of Pu isotopes are proportionaly to the flux profiles of thermal neutron. This results can be used for verification of other aspect of fuel behavior in the modeling results and also can be used as guide and comparison to the future post irradiation examination for Pu isotopes distribution.

2. Theory / Calculation

Fig. 0 illustrates the chain of nuclear fission and decay of Pu isotopes [6]. The most common isotope formed in a typical nuclear reactor is the fissile Pu-239 isotope, formed by neutron capture from U-238 (followed by beta decay), and which yields much the same energy as the fission of U-235. Well over half of the plutonium created in the reactor core is 'burned' *in situ* and is responsible for about one third of the total heat output of a light water reactor (LWR). Of the rest, about one sixth through neutron capture becomes Pu-240 (and Pu-241). The approximately 1.15% of plutonium in the spent fuel removed from a commercial LWR power reactor (burn-up of 42 GWd/t) consists of about 53% Pu-239, 25% Pu-240, 15% Pu-241, 5% Pu-242 and 2% of Pu-238, which is the main source of heat and radioactivity [7]

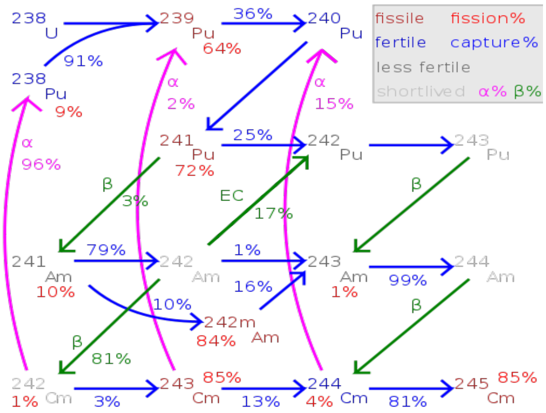

Fig. 0. Nuclear reaction chain of Pu isotopes during irradiation of natural UO2 pellet[6].

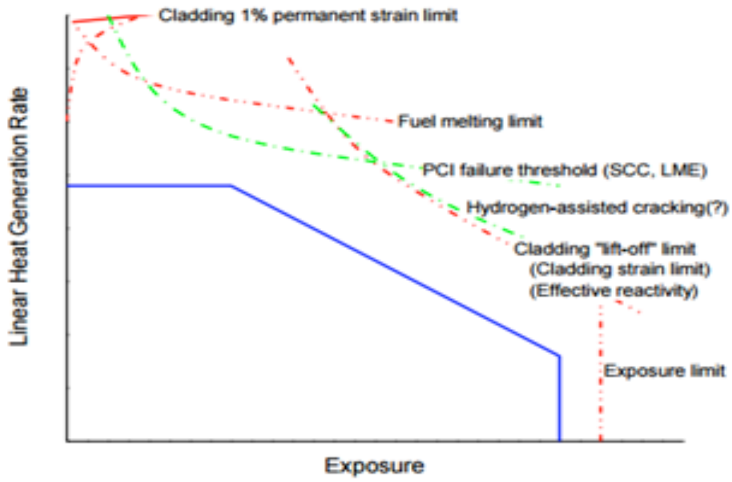

Fig.1. LHR profile chosen under LHR envelop and related its individual constraints [11]

3. Experimental Method

The natural UO2 fuel geometry is in between former design of PWR fuel [11] and new design of PHWR fuel of larger pellet and thinner fuel to cladding gap. The plan of irradiation test will be at PWR coolant temperature & pressure and thermal neutron flux of PWR. The main purpose of the irradiation test in the PRTF is to obtain the limite of power ramp can be applied to the fuel pin. The power ramp parameter to be considered for the testing must be determined according to the design and fabrication. Since the design is chosen need to be compatible to both the existing PTRF which are capability and duty limites and capability or mastering of fabrication of pin technology, while the main required fuel performance for the first NPP in Indonesia is: minimum fuel assemby discharge burnup 50 MWd/kgHE in case the reactor type is PWR. The comparative natural uranium fuel or HWR fuel burnup is about 8 MWd/kgHE. The problem is solved in the platfoem of Transuranus integral thermal mechanical behavior of water cooled reactor.

The fuel pin design is presented in Table-1, which is sorted form originale of EdyS publication [8]. The LHR history is chosen a typical simple one under envelop described by Hagrman [9]. Despite the accompany lot properties of cladding exist, the prediction use the standard MatPro. Both properties of pellet and cladding are modelled according to MatPro xi, although the accompany lot properties of cladding exist. Since the existing accompanied properties of cladding lot is fulfil the ASTM B850 [10].

The axial profile of neutrons flux have been chosen from both experimental measurement and model calculation. The parameter of ramp power has been obtained from statistical experiment data. As natural uranium is used for fuel pellets, a simplified and typical base-load commercial PHWR profile of LHR history has been chosen, to determine the minimum irradiation time before ramp test can be performed. The obtained design and manufactured fuel pin data and measured and model material properties data and behavior and component have been used as input. The chosen model takes into account MatPro XI materials properties models, HWR model for isotope generation and depletion but ignores the period of power shut-down and restart-up. The axial profile of neutrons flux has been accommodated by 5 slices of discrete pin. The slice-4 with highest power rate has been chosen to be evaluated. The radial discretion of pellet and cladding and numerical parameter have been used the default best practice of TU code used

The LHR history has been chosen under LHR limite by combination of different constraint from begining of life (BOL) up to end of line (EOL) [11]

1) Cladding 1% permanent strain limit at BOL (Begining of Live); 2) Fuel melting limit; 3)PCI failure threshold; 4)Hydrogen assisted cracking ; 5)Cladding lift-off limit / Cladding strain limit / Effective reactivity 6) Exposure limit While the cooling temperature and pressure is typical base load of commercial irradiation. The important input of initial and boundary condition is presented by Table.2.

4. Results and Discussion

The results of time distribution of Pu 239 increased rapidly. The maximum burn up of slice 4 at upper the median slice, it reached nearly 90% of maximum value at about 6000 h with peak of 0.8%a Pu/HM at 22000 h, which is higher than initial U 235. Each 240, 241 and 240 Pu grows slower and ends up to 0.4, 0.2 and 0.18 % respectively. The axial distribution of Pu isotopes are proportionally to the flux profile of thermal neutron. This results can be used for verification of other aspect of fuel behavior in the modeling results and also can be used as guide and comparison to the future post irradiation examination for Pu isotopes distribution.

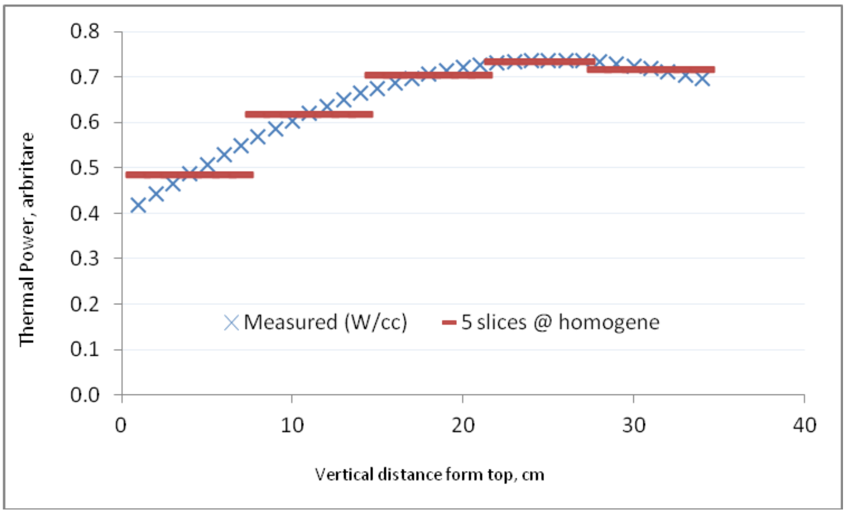

Fig. 2a. Axial (absis) distribution of power density computed form neutronic (x blue) and model of 5 slices (--red)

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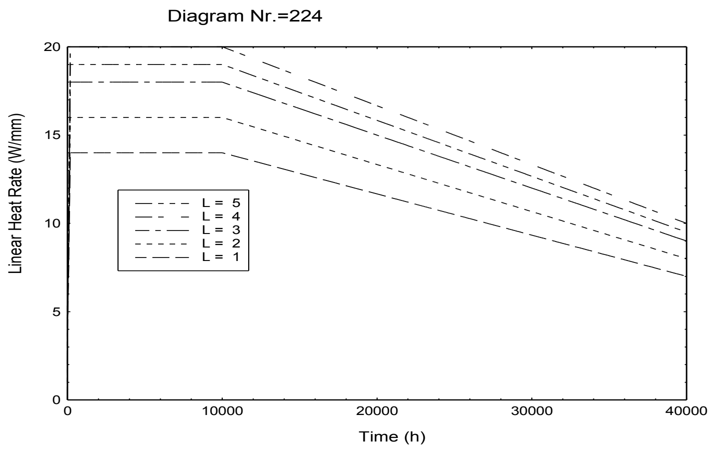

Fig.2b. Chosen input LHR history of different axial positions of fuel pin

Fig. 3 shows the temporal distribution of 5 isotopes and its total, of radially averaged slice-4 whichis highest power of axial position, from start-up to 20000 hr. The Pu 239 the highest concentration of Pu Isotope first grows rapidly up to about 5000 hr then grows more slowly accomponied to the growth of Pu240, followed by Pu241 and Pu242 consecutively. The fraction of Pu239 to the total Pu reach its peak at about 5000 hr, or in this case 6000 hr, which is 0.55, then slowly decreasing. This peak is chosen for Pu-239 production from natural UO2.

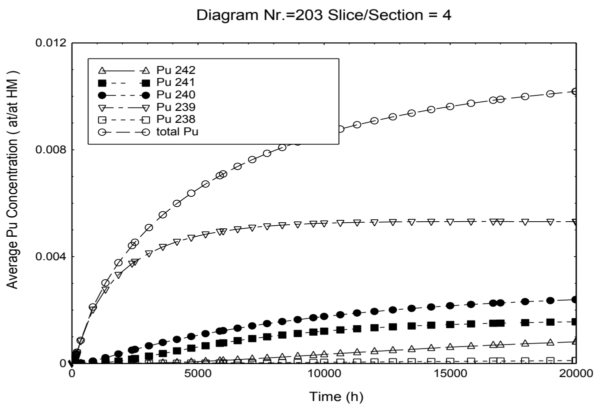

Fig.3. Generation and depletion of Different Pu Isotopes

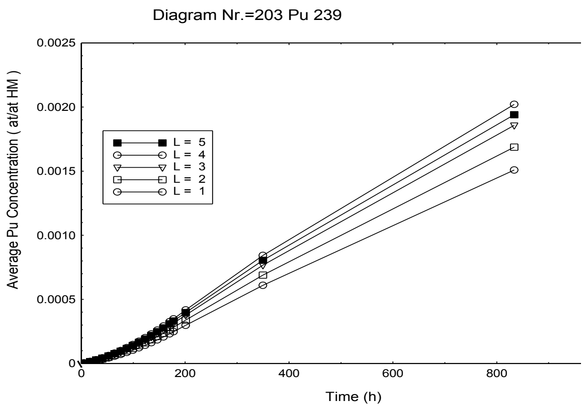

Fig.4. Time distribution of Different Pu 239 for different axial positions of fuel pin

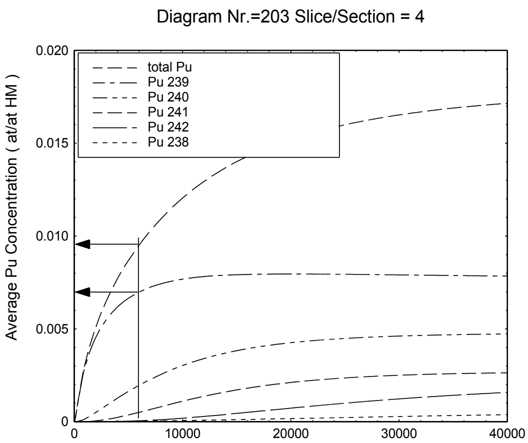

Fig. 3 (b) Zoom of Fig. 3Pu_i in slices-4 up to 40.000 h {40000 0.017153 Pu-total}

Table-1. Fuel pin Data [12]

Fuel Pin Parameter	**Value**	**Unit**
Pellet:		
Material	Nat. UO_2	-
Length	12	mm
Outside diameter	9.55	mm
Density	95	% ThD
Cladding:		
Material	Zry-4	-
Thickness	1	mm
Number of Pellet per pin	20	-
Rod:		
Length	244	mm
Outside diameter	13,2	mm

Fig. 4 shows the evolution of radially averaged of slices for Pu239. Slice-4 produces highest Pu239, as it is the highest generating power.

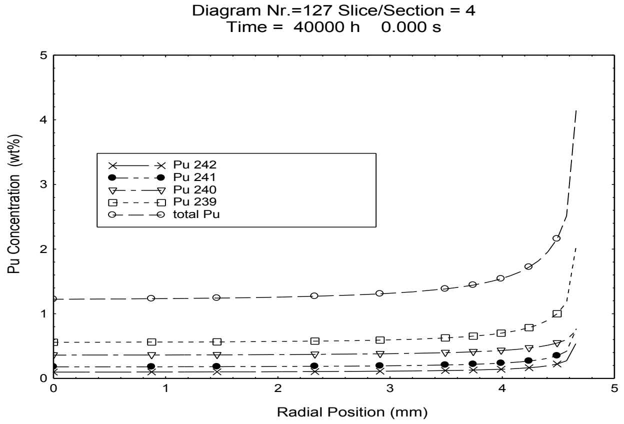

Fig.5. Radial distribution of Different Pu Isotopes at the end of irradiation

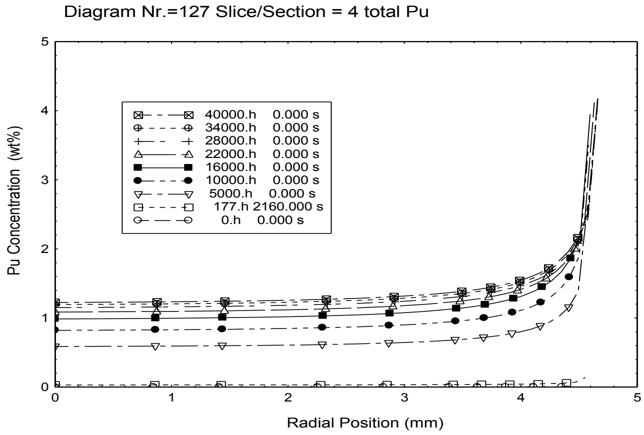

Fig.6. Radial distribution of total Pu Isotopes at different irradiation times

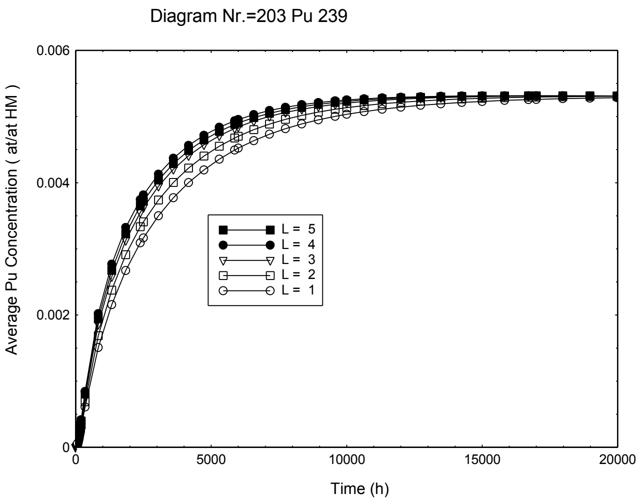

Fig.7. Evolution of average concentration of Pu 239 in different slices up tp 20.000 h

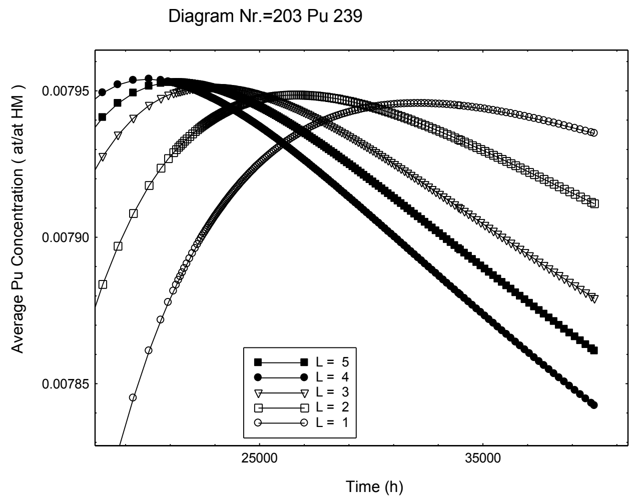

Fig.8. Zoom Fig 7. Arround 25000 hr, the volution of average concentration of Pu 239 in different slices during (20 – 40) 000 h

Fig. 5 shows the radial distribution of Pu Isotopes accentuating with time irradiation or burnup for all isotope at 10-15% of outer zone of pellet as shown by Fig 5 for BOL. Fig. 6 shows this is not only at BOL but aaccentuating all irradiation time. It related to penetration limit of epithermal neutron, and termal flux distribution. The data can be used for minimizing and optimizing the post irradiation examination.

Table-2. Important initial boundary data

No. of section or slice		1	2	3	4	5
Axial coordinate	(mm)	31.96	95.88	159.80	223.72	257.65
Relative position	(%)	12.31	36.93	61.55	86.18	99.24
Linear rating	(kW/m)	0.00	0.00	0.00	0.00	0.00
Fuel temperature	(C)	20.00	20.00	20.00	20.00	20.00
Coolant temperature	(C)	20.00	20.00	20.00	20.00	20.00
Steam content	(%)	0.00	0.00	0.00	0.00	0.00
Heat transfer coeff.	(W/m**2*K)	0.11E+05	0.11E+05	0.11E+05	0.11E+05	0.11E+05
Heat transfer coeff.tot	(W/m**2*K)	0.11E+05	0.11E+05	0.11E+05	0.11E+05	0.11E+05
Melt fraction fuel	(%)	0.00	0.00	0.00	0.00	0.00
Melt fraction clad	(%)	0.00	0.00	0.00	0.00	0.00
Fuel inner radius	(mm)	0.000	0.000	0.000	0.000	0.000
Fuel outer radius	(mm)	4.515	4.515	4.515	4.515	4.515
Thickness HBS, transition	(mm)	0.000	0.000	0.000	0.000	0.000
Thickness HBS, fully develop.	(mm)	0.000	0.000	0.000	0.000	0.000
Clad inner radius	(mm)	4.665	4.665	4.665	4.665	4.665
Clad outer radius	(mm)	5.375	5.375	5.375	5.375	5.375
Total clad outer radius	(mm)	5.375	5.375	5.375	5.375	5.375

Outer clad corrosion layer	(um)	0.00	0.00	0.00	0.00	0.00
Relative radial clad deformation:						
inner radius	(%)	0.00	0.00	0.00	0.00	0.00
outer radius	(%)	0.00	0.00	0.00	0.00	0.00
Average strains in cladding:						
effective creep strain	(%)	0.00	0.00	0.00	0.00	0.00
effective plastic strain	(%)	0.00	0.00	0.00	0.00	0.00
permanent tangent.strain	(%)	0.00	0.00	0.00	0.00	0.00
Cavity pressure	(MPa)	0.10	0.10	0.10	0.10	0.10
Clad inner loading	(MPa)	0.10	0.10	0.10	0.10	0.10
Outer pressure	(MPa)	0.10	0.10	0.10	0.10	0.10
Fluence	(neutrons/(cm**2))	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
Gap conductance	(W/m**2*K)	894.	894.	894.	894.	894.
Gap size (radial)	(um)	149.64	149.64	149.64	149.64	149.64
Centre grain size	(um)	10.00	10.00	10.00	10.00	10.00
Burn-up	(MWd/tU)	0.	0.	0.	0.	0.
Fission gas release	(%)	0.00	0.00	0.00	0.00	0.00
He production	(umol/mm)	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
He release	(%)	0.00	0.00	0.00	0.00	0.00

Fig. 7 shows average Pu239 concentration of each 5 slices. It shows the slice-4 is the highest, but also the first both increasing and in Fig. 8 decreasing, as its U238 is first exhausted because the original fissile is homogeneously distributed, but the LHR and burn-up are the highest.

Table 3. Typical concentration of Pu isotopes as % of fissile content at different reactor type and burn-up

Reactor type	Mean fuel burn-up (MW d/t)	Percentage of Pu isotopes at discharge Pu-238	Pu-239	Pu-240	Pu-241	Pu-242	Fissile content %
PWR	33000	1.3	56.6	23.2	13.9	4.7	70.5
	43000	2.0	52.5	24.1	14.7	6.2	67.2
	53000	2.7	50.4	24.1	15.2	7.1	65.6
BWR	27500	2.6	59.8	23.7	10.6	3.3	70.4
	30400	N/A	56.8	23.8	14.3	5.1	71.1
CANDU	7500	N/A	66.6	26.6	5.3	1.5	71.9
AGR	18000	0.6	53.7	30.8	9.9	5.0	63.6
Magnox	3000	0.1	80	16.9	2.7	0.3	82.7
	5000	N/A	68.5	25.0	5.3	1.2	73.8

Pu240/Pu239 increases along irradiation time. Higher content in Pu240 is better for proliferation resistance, since difficulty in Pu239 purification. After 20000 hr the Pu 239 content decreases continuously while other Pu isotopes increases. The maximum Pu239 concentration 20000 hr reach its maximum 7.95% at/at of HM

Fig. 8 shows the temperature of outer cladding and bulk coolant of first slice, as well as inner cladding and outer surface of pellet. At about 18000 hr both surface temperature of pellet and inner cladding increases sharply then drops slowly, but both temperature of outer cladding and bulk coolant slightly decreasing. The inner temperature of cladding reached 1305 ºC, but the outer one is still 250 ºC. This temperature is still under its melting point, but the related cladding creep need to be considered. Yes the cladding creep is positive and the lift-off is out of limit.

5. Conclusion

CNFT has prepared a short pin as fuel pin prototype based on natural UO_2 pellet. An irradiation test will performed in the PRTF. A simulation of irradiation test has been performed for to support testing license. The spatial and temporal distribution of generated and depleted Pu 239, 240, 241 and 242 and 238 isotopes has been analyzed as Pu239 produced and partially fissioned resulting important additional power.

Axial distribution of radially averaged concentration of Pu isotopes at certain burn up the axial power.

Radial distribution of Pu isotopes by slice-4 for different irradiation time has been visualized. All radial distribution has typical having peak at the surface of pellet proportionally to radius position of U burn-up. All peak of different isotopes tend to accentuates as irradiation time or burn-up accumulated.

Temporal distribution of different Pu isotopes is characterized by highest and sunner appearance of Pu-239 concentration. The ratio of Pu239 to total Pu isotope reach its peak at 6000 hr, corresponding to optimum irradiation time for Pu -239 production.

These visualization of radial – axial and temporal distribution of different Pu isotopes which are calculated for distribution of power, temperature, mechanical properties and eventually to chemistry behavior of cladding thermal and mechanical behavior of fuel pin can be used to benchmarking of post irradiation examination.

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References

[1] Suwardi, 2013, Parametric Study on irradiation test of CNFT fuel prototype, Proc. WWER Fuel Performance Modeling and Experiment Support, Sandanski Bulgaria, 2013
[2] Anonym, 1983, Manufacturing CIRENE fuel by ENEA, Comb. IEEC, Nuclear Fuel Manufacturing Division, ITALY.
[3] H.Sudirdjo et al. 2005, Revitalization of Power Ramp Test Facility of RSG-GAS Reactor, Proc. Seminar Research Reactor Technology and Application, 2005, BATAN.
[4] Anonym, 2011, Transuranus Handbook, European Commission, Joint Research Centre-JRC, ITU 2011.
[5] P. VAN UFFELEN and M. SUZUKI, 2011, Chapter 71 "Fuel Performance Modeling and Simulations: Oxide Fuel Performance Modeling and Simulation" in Comprehensive Nuclear Materials, edited by R. Konings, accepted for publication in J. Nucl. Mat., 2011.
[6] A. SCHUBERT, P. VAN UFFELEN, J. van de LAAR, C. T. WALKER, W. HAECK, 2008, Journal of Nuclear Materials,Vol. 376 (2008) 1-10.
[7] P. VAN UFFELEN, Cs. GYÖRI, A. SCHUBERT, J. van de LAAR, Z. HÓZER, G. SPYKMAN, 2008, Journal of Nuclear Materials, Vol. 383 (2008) 137-143.

[8] T.Taryo and T.M. Sembiring, 2014, Analysis of Neutron Flux Distribution in RSG-GAS Reactor with U-Mo Fuels, Center for Development of Research Reactor Technology-BATAN, Tangerang, Indonesia, 2014.
[9] B.Arbie, 1987, Bakri ARBIE and Sutaryo SUPADI, Status Report on the Indonesian Multipurpose Reactor RSG-GAS, BATAN, Indonesia, 1987.
[10] J. Iman, 2015, privat communication, RSG-GAS, Serpong, Indonesia.
[11] J. Susilo, 2015, privat communication, Center for Nuclear Technology and Reactor Safety Serpong, Indonesia.
[12] L.P. Hong and T.M. Sembiring, Nuclear Engineering and Design 240 (2010)1433-1442.
[13] Edy Sulistyono and Tri Yulianto. J.Tek.Bhn.Nuk vol 9, No.2 (2013), pp 55-66
[14] K. Lassmann, TRANSURANUS: a fuel rod analysis code ready for use, J. Nucl. Mater., 188 (1992), pp. 295–302
[15] B. D. Murphy, Prediction of the Isotopic Composition of UO2 Fuel from a BWR: Analysis of the DU1 Sample from the Dodewaard Reactor, Computational Physics and Engineering Division, OAK RIDGE NATIONAL LABORATORY, 1998
[16] G. Kyriazidis, P Marimbeau, J. Cl Lefebvre. IMPACT OF IRRADIATION HISTORY ON INVENTORY PREDICTION DATA FOR PWR FUEL. PHYSOR 1994 - Re-actor Physics Faces 21st Century , Apr 1994, Knoxville - Tennessee, United States. Actes de la conference. <cea-01122087>
[17] L. Holta, b, , , U. Rohdea, M. Seidlc, A. Schubertd, P. Van Uffelend, R. Macián-Juanb Development of a general coupling interface for the fuel performance code TRANSURANUS – Tested with the reactor dynamics code DYN3D, Annals of Nuclear Energy, Volume 84, October 2015, Pages 73–85
[18] B. D. Murphy, Prediction of the Isotopic Composition of UO2 Fuel from a BWR: Analysis of the DU1 Sample from the Dodewaard Reactor, Computational Physics and Engineering Division, OAK RIDGE NATIONAL LABORATORY, 1998
[19] S. Azzaoui, SCALE-6 fuel depletion analyses:Application to the ARIANE program, EXTERNAL REPORT OF THE BELGIAN NUCLEAR RESEARCH CENTRE, August, 2010, SCK.CEN, Boeretang 200, Belgium