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Research Article

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Uncertainty investigation of ^{134}Cs/^{137}Cs activity ratio method in determination of the spent nuclear fuel burnup

Zohreh Gholamzadeh*, Atiyeh Jozvaziri

Safety and Nuclear Research Reactor School, Nuclear Science and Technology Research Institute, Tehran, Iran

HIGHLIGHTS

- Fuel burnup determination is mandatory for spent fuel storage.
- ^{134}Cs/^{137}Cs activity ratio is a common method for SF burnup determination.
- ORIGEN code could be used to calculate ^{134}Cs/^{137}Cs activity ratio.

ABSTRACT

For storage of a spent fuel during the life of the facility, testing or evaluation may be required to determine the integrity of the spent fuel. In addition, various tests of the spent fuel are carried out at nuclear facilities for many purposes. Therefore, it is necessary to theoretically and experimentally evaluate the fuel burnups as one of the most important issues for nuclear spent fuel storage. The present study discusses the ^{134}Cs/^{137}Cs activity ratio method in determination of the spent nuclear fuel burnup, which is very important in view of the spent fuel storage as well as its dose rate calculation. For this aim, ORIGEN and MCNPX codes were used to calculate the radioisotopes' concentrations during the nuclear fuel burnup and different cooling times of Tehran Research Reactor (TRR) fuel assembly. The obtained results showed the burnup estimation might be maximum 2 times different from its real value by using this method if the fuel burnup history has not been regarded and the cooling time is unknown. For the specific cooling times, the discrepancy may be 36% if the fuel burnup history has not been modelled. The discrepancies between the estimated burnup and the real value may be less if the TRR fuel assembly burnups are less than 55%.

KEYWORDS

Spent nuclear fuel
^{134}Cs/^{137}Cs ratio
Burnup
ORIGEN code

HISTORY

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1 Introduction

The determination of spent nuclear fuel burnup is crucial for developing effective storage strategies and managing nuclear core fuel. Many nuclear centres have used different experimental set ups to determine the spent fuel burnup. In addition, many computational methods were used to determine the spent fuel burnup as possible as precise. In the following, an overview of some of these methods are presented.

Shin et al. (Shin et al., 2002) have determined burnups for 36 points of five rods in the G23 assembly of Kori unit 1 on the basis of gamma-ray spectrometric measurement of two isotopic ratios, ^{134}Cs/^{137}Cs and ^{154}Eu/^{137}Cs. They compared their results with the obtained ones by the SCALE4.4 SAS 2H module calculations. Their achieved results showed that burnups determined from ^{134}Cs/^{137}Cs are in agreement with the declared burnups in most cases within about 12% error (Shin et al., 2002).

Ezure (Ezure, 1990) calculated some atom ratios for some spent fuel assemblies of Japan Power Demonstration Reactor (JPDR-1). The obtained results were considered using a comparison between the gamma-ray spectrometry measured values as one non-destructive method and chemical methods; which by sampling from the spent fuel, the remained uranium enrichment after a burnup time could be measured. Their obtained results showed there were discrepancies between the calculated and the measured atom ratios of Pu/U and burnup. The found result by this center showed that the calculated atom ratios of ^{134}Cs/^{137}Cs and ^{154}Eu/^{137}Cs were somewhat less than the measured values for nearly all the fuel assemblies (Ezure, 1990).

Min et al. (Min et al., 1999) determined burnup, cool-

*Corresponding author: zgholamzadeh@aeoi.org.ir

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ing time and initial enrichment, which are considered as basic parameters of a spent fuel. They used measurement of activity ratios of some radioisotopes for this purpose. A high-resolution HPGe and ORIGEN-S calculations were used to investigate the ratios for the spent fuel burnup determination. The activity ratios considered in this work are ^{134}Cs/^{137}Cs, ^{154}Eu/^{137}Cs, and ^{106}Ru/^{137}Cs. They applied some equations to calculate the spent fuel burnup. Their results showed that by putting the cooling time and initial enrichment values into one of the equations, the fuel rod averaged calculated burnup would be 40.5 GWd/tU, which differs from the operator declared burnup of 39 GWd/tU by 3.8% (Min et al., 1999).

Oleinik et al. (Oleinik et al., 2002) also used the previously mentioned method for determining the burnup of spent nuclear fuel. The method is based on analyzing of the results of spectrometric measurements performed at the Zaporozh'e nuclear power plant. This method differs from existing methods in that the initial enrichment of the nuclear fuel and the cooling time of the nuclear fuel after irradiation are resolute needed parameters (Oleinik et al., 2002).

Park et al. (Park et al., 2009) used ORIGEN-ARP code to estimate the spent fuel burnup of a PWR reactor using the ^{134}Cs/^{137}Cs method. They indicated their measurements using the gamma spectroscopy method were in good agreement with the theoretically calculated ones by 0.8 to 4.5% relative discrepancies (Park et al., 2009).

2 Materials and Methods

The present study uses ORIGEN2.1 code to calculate ^{134}Cs/^{137}Cs activity ratio in order to estimate the spent fuel burnup. ORIGEN 2.1 is a versatile point-depletion and radioactive-decay computer code for use in simulating nuclear fuel cycles of pressurized water reactors, boiling water reactors, liquid-metal fast breeder reactors, and Canada deuterium uranium reactor (Croff, 1980).

The code was used to calculate the ^{134}Cs/^{137}Cs activity ratio for TRR spent fuels. The TRR is an open pool, MTR type light water-moderated and cooled reactor with a thermal power of 5 MW. The reactor core is composed of two types of fuel assembly: standard fuel elements (SFE) and control fuel elements (CFE). The first considered core (No.83) consists of 28 SFEs containing 19 fuel plates and 5 CFEs containing 14 fuel plates. The core is fuelled with 20% enriched U3O8-Al MTR fuels. The TRR core is cooled with the mass flow rate of 500 m^3.h^{-1}. Two types of control rods are used in the TRR; which one made of Ag-In-Cd alloy, and the other of stainless steel. Both have a set of two control plates as a fork type shape (Mirvakili et al., 2012).

The present study investigates the effect of considering the real irradiation history of spent fuel on the precision of the ^{134}Cs/^{137}Cs method for the spent fuel burnup estimation by theoretical and experimental procedures.

To model the real irradiation history, the spent fuel position at any fuel cycle or core operation is very important so that its precise received power at any reactor run should be determined very sharply. MCNPX 2.6.0 code is a Monte Carlo-based code with ability of 34 particles transport and modelling different geometries powerfully. It is used for modelling different problems such as nuclear reactors, accelerators, neutron imaging systems and others (Pelowitz et al., 2005). MCNPX code is usually proper computational code to extract the real power of the specific fuel assembly.

The value is calculated by F7 tally of the computational code for each position that the fuel plate or fuel assembly has been placed at any core operation cycle. Power ratio is referred to the marked fuel assembly or fuel plate power (the one which its burnup is going to be calculated) than the whole core power. The value is calculated from the reactor start-up till the reactor power steady state. The data is used as a high-precision history in ORIGEN code.

The power (MWH) and the time (day) has been provided from logbook data reported by the reactor operators during different cycles of TRR core operation. These data provide enough required data for ORIGEN code which are the irradiation time history including both irradiation and cooling days during the fuel plate burnup as well as the received fuel plate power on MW. The data are used as IRP card of ORIGEN code which the first entry is time and the second one on this card is power (MW). Third and fourth entries on IRP card are the number of the vectors and the fifth entry is the time unit. ORIGEN code forms a matrix from these data in a vector space to solve Bateman equation as the following:

$$\begin{aligned}\frac{\mathrm{d}N_i}{\mathrm{d}t} = &\sum_j \gamma_{ij}\,\sigma_{f,j}\,N_j\varphi + \sum_k \sigma_{c,k\to i}N_k\,\varphi + \\ &\sum_l \lambda_{l\to i}N_l - \left(\sigma_{f,j}\,N_{ji}\varphi + \sigma_{a,i}\,N_{ji}\varphi + \lambda_i N_l\right)\end{aligned} \quad (1)$$

where $\frac{\mathrm{d}N_i}{\mathrm{d}t}$ is time rate of change in concentration of isotope i, $\sum_j \gamma_{ij}\,\sigma_{f,j}\,N_j\varphi$ is production rate per unit volume of isotope i from fission of all fissionable nuclides, $\sum_k \sigma_{c,k\to i}N_k\varphi$ is production rate per unit volume of isotope i from neutron transmutation of all isotopes including (n,γ), $(n,2n)$, etc., $\sum_l \lambda_{l\to i}N_l$ is production rate per unit volume of isotope i from decay of all isotopes including β^-, β^+, α, γ, etc., $\sigma_{f,j}\,N_{ji}\varphi$ is removal rate per unit volume of isotope i by fission, $\sigma_{a,i}\,N_{ji}\varphi$ is removal rate per unit volume of isotope i by neutron absorption (excluding fission), $\lambda_i N_l$ is removal rate per unit of isotope i by decay (Fensin, 2008).

In this work, real history of a fuel plate during 2 years burnup at different positions of TRR core was modelled using MCNPX code to show its importance for gamma dose rate or burnup calculations. The history is used as ORIGEN code input to calculate ^{134}Cs/^{137}Cs activity ratio.

3 Results and discussion

It should be noted in the case of some research reactors, depending on the nuclear core operation strategy the core

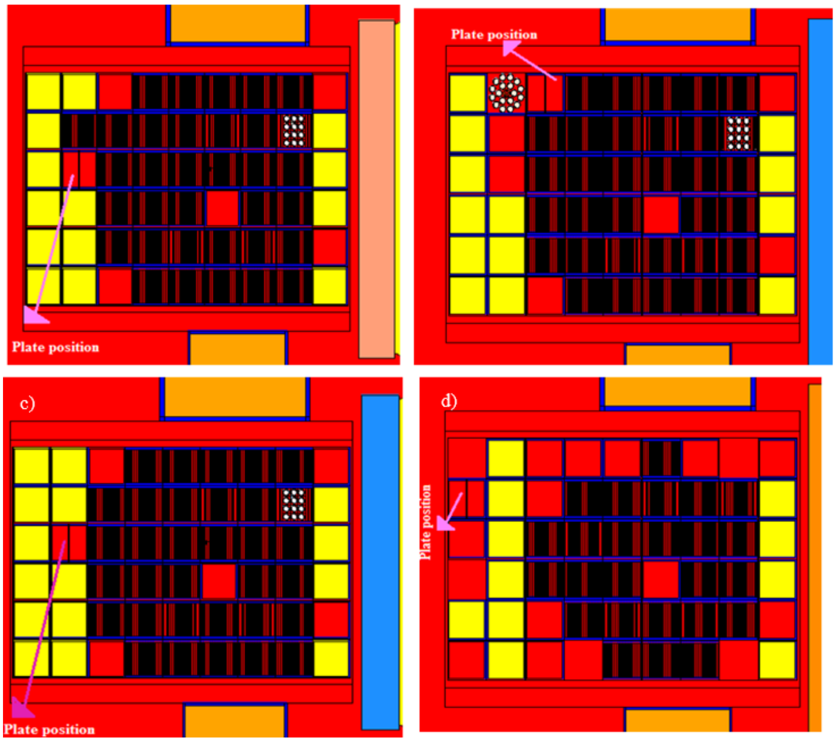

Figure 1: Different core configurations during a fuel plate irradiation history (for 2 years).

configuration as well as the operational power during a cycle is not constant so that this effects on the fuel burnup and Cs-134 as well as Cs-137 accumulation.

By modelling different configurations of the reactor core during a special irradiation history for a specific assembly, its real irradiation history would be obtained. For example, Fig. 1 shows real history of the modelled core configurations experienced by one of the fuel plates during its irradiation history. The first fresh fuel loading in the TRR core was modelled using MCNPX2.6.0 according to Fig. 1-a. Its power ratio was calculated using F7 card of the computational code. Then its second position for the next operation cycle of the TRR was modelled using the computational code and its power ratio was calculated using the same mentioned procedure (Fig. 1-b). This history preparation for the fuel plate during its 2-year burnup was followed according to Figs. 1-c and 1-d. Then the provided history after the carried-out calculations using MCNPX2.6.0 code was written very precisely according to Table 1.

The obtained data was used in ORIGEN2.1 input file which the irradiation time and the power ratio are used in IRP card of the computational code at its first and second terms respectively. For the cooling times between the reactor shut dawn and its start-up the DEC card of ORIGEN 2.1 code is used which the cooling time is used at its first term. After the spent fuel history writing in the ORIGEN code input, calculation of the different radioisotopes' build-ups is calling using the 7^{th} term of the OPTL/OPTF/OPTA cards of the computational code to determine the ^{134}Cs/^{137}Cs ratios from the code output at different situations (irradiation times, cooling times, powers, continuous and discontinuous burnups).

Table 1: History writing for a specific fuel assembly. MWH and Power ratio have been provided from TRR operator and MCNPX2.6.0 code, respectively.

Year	Date	MWH	Power ratio	Time (day)
2012	Feb 17	580	0.004009143	6.0694
2012	Feb 23	cooling		12.0694
	Feb 23	85.83	0.007110898	12.236
	April 12	cooling		60.896
	April 12	670.453	0.006925266	61.0626
	April 14	cooling		63.2605
	April 14	472.94	0.00384867	68.2605
	May 10	cooling		88.8327
	May 10	5	0.007110898	89.0236
	May 11	cooling		89.7736
	May 11	625	0.00384867	96.1902
	May 22	cooling		100.8568
2013	May 22	0.015	0.00384867	100.8811

Table 1 shows one example for irradiation history of a specific fuel plate. The table data are used as ORIGEN code input for precise estimation of ^{134}Cs/^{137}Cs activity ratio in the spent fuel after its final burnup. As before mentioned the power ratio has been calculated using MCNPX2.6.0 code regarding the fuel plate positions during 2-years burnup inside the TRR core.

To estimation the rough burnup without using any history modelling, which would be extremely time consuming, ^{134}Cs/^{137}Cs ratio was calculated for different burnups of TRR fuel assembly and different cooling times. Maximum fuel burnup of TRR fuel assemblies is about 55%. As Fig. 2 shows, there is about 36% relative discrepancy between continuous 55%-burnup calculation and the real history 55%-burnup for the different cooling times. If the

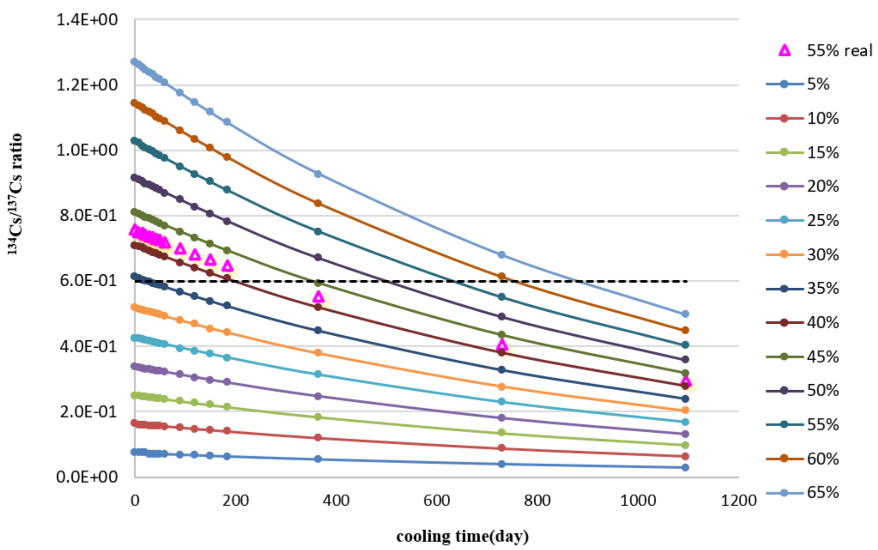

Figure 2: ^{134}Cs/^{137}Cs activity ratio investigation for different burnups and cooling times of TRR fuel assemblies.

cooling time is unknown, the estimated burnup value from the obtained ratio may differ even 2 times higher than its real value (See the 0.6 ratio cuts off 30% burnup curve as well as 65%-burnup curve at different cooling times).

Clearly uncertainty of the method could be observed by taking an attention on the dash line in the Fig. 2. If the measured ^{134}Cs/^{137}Cs activity ratio is 0.6 and cooling time of the spent fuel is completely known, for low cooling times less than 1 week the estimated burnup would be 35% which its discrepancy with the real value (55%) is 36%. If the cooling time of the spent fuel is longer for example 365 days, the estimated burnup would be 45% which its discrepancy with the real value (55%) is 18%. If the cooling time of the spent fuel is longer for example 730 days, the estimated burnup would be 60% which its discrepancy with the real value (55%) is 9%. If the cooling time of the spent fuel is longer for example 1095 days, the estimated burnup would be 65% which its discrepancy with the real value (55%) is 18%. Then if the burnup is completely unknown the method has maximum uncertainty 36% relative discrepancy for TRR spent fuels whereas maximum burnup of them is 55% before permanently storage in spent fuel pool. Values of real history modelling (triangle symbol in Fig. 2) have 36% difference than the 55% continuous burnup (dark bule circled-line in Fig. 2) theoretical calculations without any burnup history simulation.

The obtained calculated curves would help the operators to estimate the spent fuel burnup if the cooling time is accurately known. In low burnups the real history application to estimate the spent fuel burnup has not significant weight as it is seen in Fig. 3. The data observed in Fig. 3 shows there is a relative discrepancy of 1% between the real history simulation than the values obtained without any spent fuel history modelling.

Dependence of the ^{134}Cs/^{137}Cs activity ratio to the reactor power was investigated for a 10.5% burnup TRR fuel assembly by considering the TRR core operation at 5 MW, 2.5 MW and 1 MW respectively. The carried-out investigation showed there is about 9% relative discrepancy between the 10.5% TRR fuel assembly burnup obtained by 5 MW core operation and 1 MW core operation (Fig. 4).

Caruso et al. (Caruso et al., 2007) showed the ^{134}Cs/^{137}Cs activity ratio is not very precise in the high burnup ranges and may result in errors of a few percent in the range of low and medium burnups, while much higher discrepancies are observed between the calculations and measurements (even 23%) for high burnups (Caruso et al., 2007).

Harp et al. (Harp et al., 2014) showed the accuracy of the non-destructive burnup evaluation from gamma spectrometry for TRISO fuel compacts across a burnup range of approximately 10 to 20% FIMA (fissions per initial heavy metal atom) and also validated the approach used in the physics simulation of the AGR-1 experiment while there are 3.1% to 5.3% relative uncertainties (Harp et al., 2014).

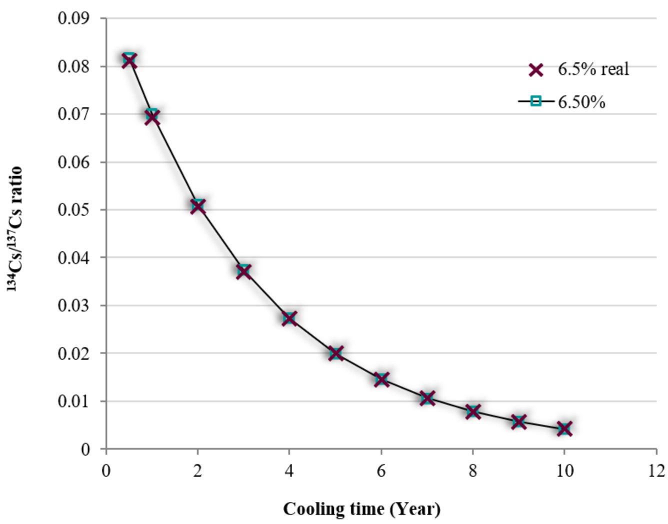

Figure 3: ^{134}Cs/^{137}Cs activity ratio investigation for low burnups and cooling times of TRR fuel assemblies.

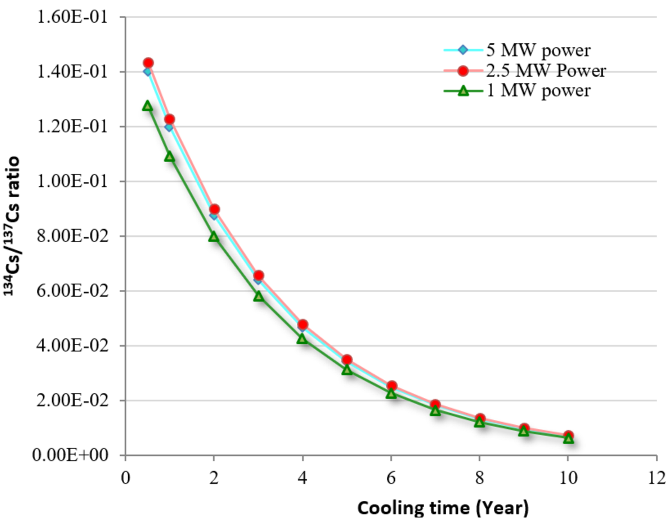

Figure 4: ^{134}Cs/^{137}Cs activity ratio investigation for different core operation power and cooling times of TRR fuel assemblies.

Joshi et al. (Joshi et al., 2019) explained the absolute measurement of fission product gamma activity or the ratio of fission product activities (such as ^{134}Cs/^{137}Cs or ^{154}Eu/^{137}Cs) has been used previously to measure or verify burnup of a commercial reactor spent fuel with a known reactor power history. If a cooling time of a spent fuel is less than a year, burnup can be measured by La-140, Zr-95, Ce-144, or Ru-106 absolute gamma-ray activity determination using a high-resolution HPGe. If a cooling time is greater than 1 year, then absolute measurement of Cs-137 or the ratios could be used (Joshi et al., 2019).

It should be noted in the case of research reactors that the power is not constant during an operation cycle or different cycles during several years, absolute Cs-137 activity determination method is less valid than ^{134}Cs/^{137}Cs method for burnup estimation of a fuel plate or fuel assembly. Clearly Cs-137 build up value is directly dependent to absolute power of the reactor core while the ratio has less dependency to it. In overall it can be said the ^{134}Cs/^{137}Cs ratio method has less discrepancies with the real values in low burnups while the discrepancy enhances in high-burnups. The best approach that research reactors can take in this regard is to automatically and accurately record the history of each fuel using a software designed for this goal by nuclear programmers. This recorded data can be used to estimate the fuel burnup of a given plate or assembly using both absolute value and ratio methods. Without such history recording, it is suggested any research reactor provide its own ratio curve according to Fig. 2 that at least helps to operators to estimate the spent fuel burnup with some uncertainty if the cooling time after the final storage of the spent fuel is completely known (it is usually recorded in storage pool logbooks).

4 Conclusions

All the nuclear reactor operators are trying to calculate and measure the spent fuel burnups as possible as close to its real value. Computational methods are used to help the burnup value be predicted as precise as possible. The most used method in this regard is ^{134}Cs/^{137}Cs ratio measurement by gamma spectroscopy. The present research aimed to investigation of the method uncertainty. The carried-out work showed the measured value without consideration of the spent fuel irradiation history might cause a maximum 36% discrepancy for 55%-burnup in the case of TRR fuel assemblies while the uncertainty is less than 1% in the case if 6.5% burnup of the spent fuel plate. It should be mentioned that for low burnups there are fewer relative discrepancies as it is seen for 6.5% burnup. It can be said in the case of power reactors which their configuration or power has not much fluctuation the method could be concluded in less uncertainties because their operation history is not complicated. In the case of research reactors which their operation cycle and power depends on costumers, there is more complicated operation history and noticeable power and core configuration fluctuations which drastically effect on the ^{134}Cs/^{137}Cs method uncertainty. It should be taken in attention, computational method applied in the present work helps the operators to predict an unknown burnup if the cooling time is absolutely known.

Conflict of Interest

The authors declare no potential conflict of interest regarding the publication of this work.

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