A Characterization Study of Nuclear Reactors Through Xenon Isotopic Activity Ratios

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1. Introduction

Radioactive gases and particulate nuclides are collected in the atmosphere from control stations deployed through the International Monitoring System (IMS) established by the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO). Data on the release of atmospheric radioactivity from underground nuclear test explosions by neighboring countries in the past are important in developing and verifying methods of verification of the Comprehensive Nuclear-Test-Ban Treaty (CTBT). There has been a total of six nuclear tests from the neighboring country so far. So, there have been several research works to interpret potential nuclear explosion signals detected in the network using radioactive noble gases isotopes generated as by-products of nuclear activity and to identify nuclear activity in neighboring countries [1]. Of the noble gases, the xenon isotopes have special importance because they have been considered as strong indicator for nuclear activities. However, there are many possible facilities generating xenon isotopes such as different types of reactors and nuclear tests, which makes it difficult to identify the source of the xenon isotopic detection. Therefore, it is very important to devise a reliable indicator which can discriminate the source of xenon detection. The nuclear facilities operating in North Korea include IRT-2000 research reactor, nuclear fuel reprocessing facilities, uranium enrichment facilities, and 5MWe graphite reactor in Yongbyon [2].

In this work, the characteristics of xenon isotopic activity ratios are analyzed and discussed in detail for the various reactors including PWR, CANDU, IRT-2000, and MAGNOX reactors.

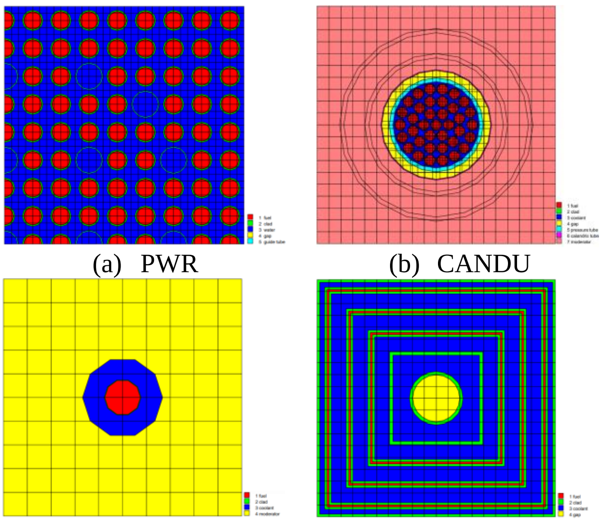
2. Methods and Results

2.1 Computational method

In this work, xenon isotopic analysis was performed using ORIGEN/TRITON module in SCALE 6.2 [3]. The ORIGEN code which was developed at Oak Ridge National Laboratory is widely used for irradiation and decay simulation for general purpose isotopic tracking. Key capabilities of ORIGEN are calculation of isotopic compositions and source terms for lots of nuclear applications. The primary function of TRITON is to simulate the time-dependent evolution of nuclide inventories of a reactor system through a series of multi-group transport calculations and depletion/decay calculations. Additionally, this module generates reactor-specific effective one-group cross section libraries for follow-on ORIGEN calculations which contain fuel assembly-averaged cross sections as function of burnup, enrichment, moderator density, boron concentration, and temperatures in the ORIGEN binary library file.

2.2 Modeling of Reactors

The first step is to generate the reactor-type specific effective one-group cross sections which are to be used in the depletion analysis using ORIGEN. As mentioned before, we considered PWR, CANDU, IRT-2000, and MAGNOX reactors. Rather than the whole core modeling, we generated the effective one-group cross sections by using lattice structures of these reactors with TRITON module to reduce the computational burden. The TRITON modelings of these reactors are shown in Fig. 1. For PWR, we modelled only quarter of typical 16x16 CE type assembly. For CANDU, we only one fuel bundle comprised of 37 fuel rods (natural uranium) surrounded by a large D_2O moderator region. For MAGNOX, we only considered one fuel channel in which the central fuel (U-0.5wt%Al) is sequentially surrounded by a CO_2 coolant region and a large graphite moderator region [4].



(a) PWR (b) CANDU
(c) MAGNOX (d) IRT-2000
Fig. 1. The reactors modeled with SCALE 6.2

For IRT-2000 reactor, the lattice is comprised of three rectangular fuel plates of UO_2 (36.15wt% enriched uranium) and the water coolant fills the coolant regions between fuel plates. Table 1 summarizes the conditions for the depletion and cooling calculation using ORIGEN. For PWR, three cycles operation of 375 days depletion and 60 days cooling is considered with a specific power of 40 MW/MTU while only one cycle depletion of 700 days is considered with 19.5 MW/MTU for CANDU, giving 13.5 MWd/kg burnup. For IRT-2000, one cycle depletion over 350 days is considered with a very high specific power of 557.1 MW/MTU while one cycle depletion of 4000 days is considered with a low specific power of 0.50 MW/MTU [5]. For all the reactors, the simulation is performed up to 100 days after shutdown to consider the cooling time effect.

Table 1. Burn Cycle for each reactor

	PWR	CANDU	IRT-2000	MAGNOX
Irradiation Cycle (day)	375/60	700	350	4000
Number of Cycle	3	1	1	1
Specific power (MW/MTU)	40	19.5	557.1	0.502622
Burnup (MWd/MTU)	45,000	13,650	195,000	2,010
Simulation interval after shutdown (day)	100			

2.3 Result

For characterization of nuclear reactors using xenon isotopic activity ratios, we considered four xenon isotopes ($^{131m, 133m, 133, 135}Xe$). The half lives of ^{131m}Xe, ^{133m}Xe, ^{133}Xe, and ^{135}Xe are 11.9 days, 2.19 days, 5.24 days, and 9.14 hours, respectively. Usually, xenon isotopic ratios are widely used to characterize the nuclear reactor and nuclear test rather than their absolute inventories.

Fig. 2 compares the evolution of the isotopic ratio $^{135}Xe/^{133}Xe$ for PWR operation with different uranium enriched oxide fuels. As shown in Fig. 2, the different uranium enrichments lead to the different $^{135}Xe/^{133}Xe$ ratios at their equilibrium state and the higher uranium leads to the higher equilibrium $^{135}Xe/^{133}Xe$ ratio. Fig.3 compares the evolutions of this xenon isotopic ratio for the considered four reactors, where PWR results were obtained with 4.5wt% uranium enrichment. From this figure, it is shown that the xenon isotopic ratio at an equilibrium state for MAGNOX is significantly higher than the other cases, which makes it possible to discriminate MAGNOX from the other ones. The equilibrium $^{135}Xe/^{133}Xe$ ratios from CANDU and IRT-2000 are much lower than PWR with 4.5wt% uranium enrichment and it seems that CANDU and IRT-2000 can be discriminated using this xenon isotopic ratio from PWRs having conventional uranium enrichments of 3.0~5.0wt%. However, it is difficult to discriminate CANDU and IRT-2000 using this xenon isotopic ratio.

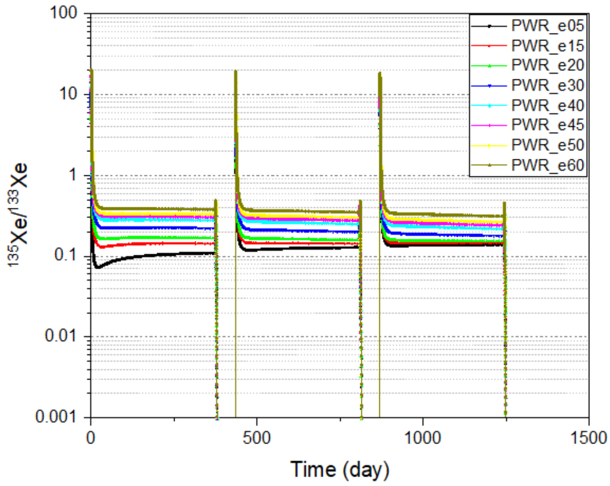


Fig. 2. Evolutions of $^{135}Xe/^{133}Xe$ ratio for PWR having different uranium enriched fuels

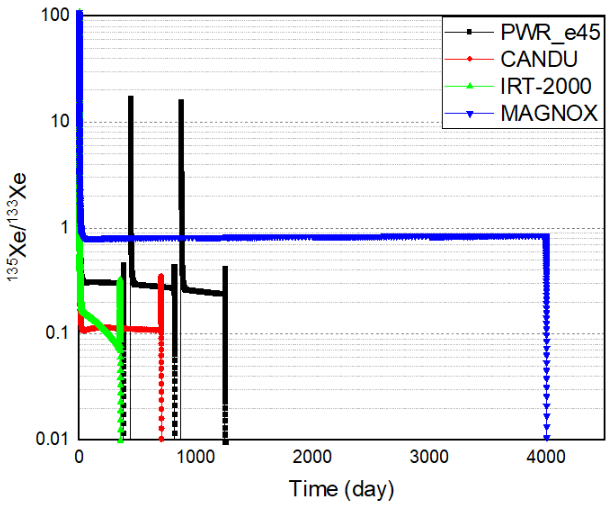


Fig. 3. Comparison of the $^{135}Xe/^{133}Xe$ ratios for the considered reactors

Next, we compared the trajectories of the fuel depletion in a plane where $^{133m}Xe/^{131m}Xe$ and $^{135}Xe/^{133}Xe$ ratios are used as x- and y-axis, respectively, for the considered reactors. The results are shown in Fig. 4.

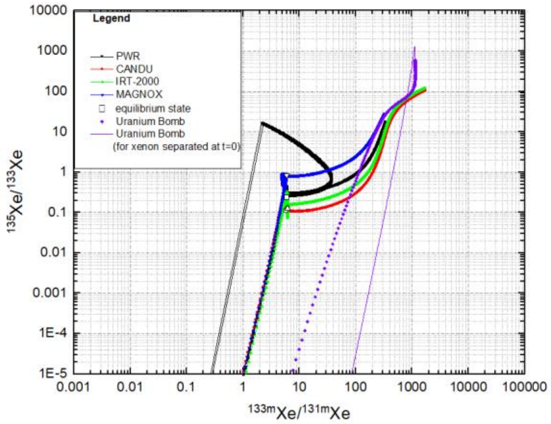


Fig. 4. Comparison of the trajectories in the $^{135}Xe/^{133}Xe$ and $^{133m}Xe/^{131m}Xe$ ratios-plane

In this figure, the trajectories for PWR are repeated for three cycles operation while the ones for the other reactors are given only for the one cycle. For example, the trajectory for PWR starts from upper and right side part modeling a reactor startup, approaches the equilibrium values designated with white rectangle symbols, describes the decrease of $^{135}Xe/^{133}Xe$ ratio as cooling, and then describes the increase of $^{135}Xe/^{133}Xe$ ratio as the reactor restarts up. So, the straight lines at the left and right sides represent the reactor startup and cooling by shutdown, respectively. As shown in this figure, the discrimination of the reactors using this figure is difficult except for the equilibrium states. In this figure, we also considered two uranium bomb cases with the fractionation of Xe isotopes (purple line) and without any fractionation (dashed purple line). In both cases, uranium enrichment is 90wt%. From this figure, it is shown that the uranium bomb with Xe fractionation can be discriminated from the nuclear reactor cases.

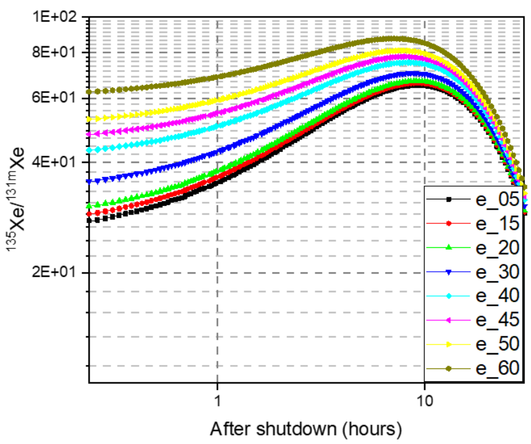


Fig. 5. Comparison of the $^{135}Xe/^{131m}Xe$ isotopic ratio evolutions for PWRs with different uranium enrichments

Next, we analyzed the $^{135}Xe/^{131m}Xe$ isotopic ratios after shutdown of the reactors in detail to understand the possibility of the discrimination of reactors during shutdown period. Fig. 5 compares the evolution of the $^{135}Xe/^{131m}Xe$ isotopic ratios for PWRs with different uranium enriched fuels. As shown in Fig. 5, the uranium enrichment gives a significant effect on the $^{135}Xe/^{131m}Xe$ isotopic ratio. In particular, the initial values of this ratio are distributed over a wide range of 20~60.

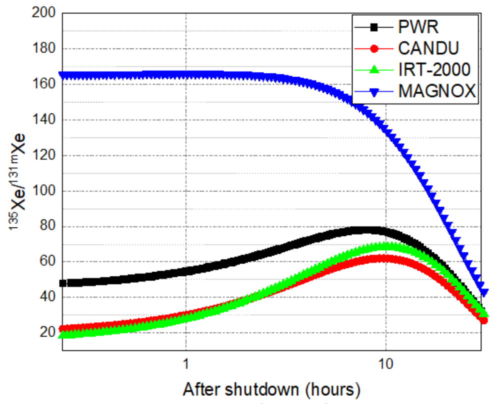


Fig. 6. Comparison of the $^{135}Xe/^{131m}Xe$ isotopic ratio evolutions for different reactors (PWR with 4.5wt% uranium enriched fuel)

Fig. 6 compares the evolutions of the $^{135}Xe/^{131m}Xe$ isotopic ratios versus time after shutdown for the considered reactors. As shown in this figure, it is possible to discriminate MAGNOX reactor from the other reactors and to discriminate IRT-2000 and CANDU from the other reactors except for the PWRs having very low uranium enrichments less than ~1.0wt% within several hours after shutdown.

3. Conclusions

In this work, we analyzed the characteristics of the xenon isotopic ratios for four different nuclear reactors which are operated or assumed to be operated in our country and neighboring countries. These analyses were performed using depletion calculation with ORIGEN and with the effective one-group cross sections generated with TRITON module of SCALE 6.2 for their lattice structures. The basic purpose of the analysis is to show that it is possible to discriminate these reactors using the information of the detected xenon isotopic ratios. From the analysis, it was shown that the MAGNOX reactor can be discriminated from the other reactors using $^{135}Xe/^{133}Xe$ ratio at the equilibrium state, and that CANDU and IRT-2000 reactor can be discriminated using this xenon isotopic ratio from PWRs having conventional uranium enrichments of 3.0~5.0wt%. However, it is difficult to discriminate CANDU and IRT-2000 using this xenon isotopic ratio. Also, the trajectory analysis of the fuel depletion using $^{133m}Xe/^{131m}Xe$ and $^{135}Xe/^{133}Xe$ ratios can be used to discriminate MAGNOX from the other reactors. Also, it was shown that the uranium bomb with Xe fractionation can be discriminated from the nuclear reactors. Finally, the analysis of the $^{135}Xe/^{131m}Xe$ ratio after shutdown showed that MAGNOX reactor can be discriminated from the other reactors and that IRT-2000 and CANDU can be discriminated from the other reactors.

4.Acknowledgment

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