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Determination of fission products of Ghana Research Reactor-1 at End of Cycle

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ABSTRACT

The fission product isotopic inventories have been estimated for a 90.2% highly enriched uranium (HEU) fuel core of the Ghana Research Reactor-1 (GHARR-1) using the ORIGEN2 code. The results indicate a gradual decrease in the ^{135}Xe inventory, and a build up in the ^{239}U and ^{239}Np inventory. The results will assist in assessing the bounding radiological consequences of a release of actinides from fuel material. It will also assist in the design of a nuclear spent fuel storage cask for GHARR-1 using local material (Serpentine).

Introduction

The core conversion studies of the Ghana Research Reactor Core started in 2006 with the objective of designing an LEU core with similar operational capabilities as the original HEU core and with acceptable safety margins under both normal and accident conditions. The burnup studies of the reactor is closely related to the neutronic calculations and the structural design. In the operation of nuclear reactors, the fuel is gradually depleted or used up. In this case, the number of nuclear fission reactions occurring in the fuel in the reactor core decreases; consequently, the effective multiplication factor reduces. To compensate for reactivity loss, control rods have to be withdrawn. For the special case of GHARR-1 which has only one control rod, loss of core excess reactivity due to fuel depletion is achieved by adding beryllium shim reflectors to the top part of the core to act as upper axial reflectors.

A major contributor to fuel depletion is the production of different fission fragments or products which invariably have large absorption cross sections. Their accumulation in the reactor core tends to reduce the core reactivity. In particular, since absorption cross sections decrease rapidly with increasing neutron energy, such fission product poisons are of great importance to thermal reactor design. The amounts and activities of individual fission products and the total fission product inventory in nuclear fuel during reactor operation and shutdown have important connotations. These quantities are used to evaluate radiation hazard from fission product dispersal to the environment, to determine the fission product radioactivity in the spent nuclear fuel after discharge from the reactor core, to estimate decay heat removal and finally to calculate the poisoning or parasitic capture by fission products that accumulate during reactor operation (Gladstone et al,1994). The two most important fission products of concern in reactor core design and analysis, and also to reactor operators are ^{135}Xe (a 1/v absorber) with an enormous thermal absorption cross section is 2.65b and a relatively large yield. The designed core lifetime estimated for GHARR-1 is ten years if it is operated continuously at peal power of 30kW for 2.5 hours a day. The measured cold clean core excess reactivity during the zero power criticality tests is 3.99mk. This value is quite small. Therefore, to properly utilize this core excess reactivity bank in order to realize the estimated core lifetime, adequate notice and care should be taken of core reactivity consuming poisons such as fission products. Nuclide inventory data were calculated with ORIGEN2 code package, a zero-dimensional isotope decay and transmutation code. The code solves the transmutation equations using libraries of radioactive decay data and 1-group cross section data. The base code libraries allow the tracking of over 100 actinides and nearly 900 fission product nuclides. The results of the burnup variations of the isotopic inventories of these fission products are presented. The results would in particular be helpful in performing decay heat removal calculations for the design of a spent fuel storage cask for the HEU fuel of GHARR-1.

Discription of gharr-1

The Ghana Research Reactor-1 (GHARR-1) is a commercial version of the Miniature Neutron Source Reactor (MNSR) and belongs to the class of pool-in-tank type reactors (Gao et al,1993) (Nyarko et al, 2009). It is under-moderated with an H/U atom ratio of 197. The thermal power rate of the facility is 30 kW with a corresponding peak thermal neutron flux of 1.0×10E12 n/cm2 s (Nyarko et al, 2009). For fresh core, its cold clean excess reactivity is about 4 mk. Cooling is achieved by natural convection using light water. Presently, the GHARR-1 core consists of a fuel assembly HEU (U-Al alloyed) fuel elements arranged in ten concentric rings about a central control rod guide tube, which houses the reactor's only control rod. The control rod's reactivity worth is about -7 mk, providing a core shutdown margin of 3 mk of reactivity. The small core has a low critical mass. However, its relatively large negative temperature coefficient of reactivity is capable of boosting its inherent safety properties (Nyarko et al, 2009). The small size of the core

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facilitates neutron leakage and escape in both axial and radial directions. To minimize such loses and thereby conserve neutron economy, the core is heavily reflected, respectively, on the side and underneath the fuel cage by a thick annulus and slab of beryllium alloy material. Due to its inherent safety features, stability of flux and moderate cost, the MNSR has recently found enormous application in various fields of science (Kennedy et al,2000) particularly in trace elements in matrices of biological and environmental samples (Su-De, 1984) and soil fertility studies and geochemical mapping (Umar,2003).

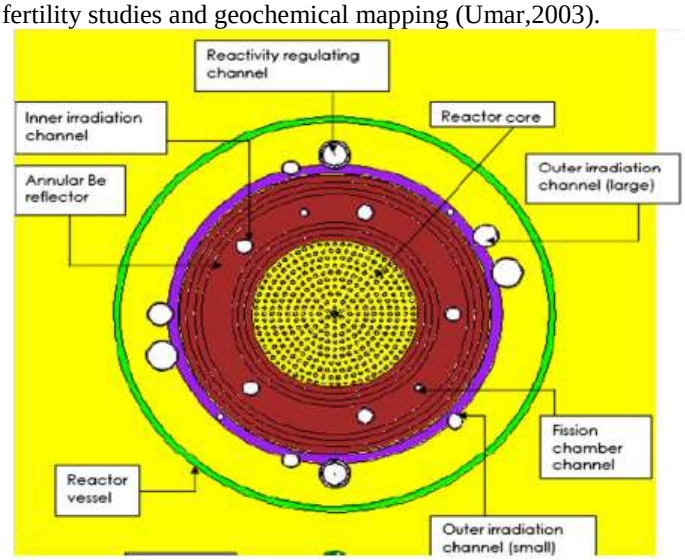

Fig 1: GHARR-1 core configuration showing fuel region.

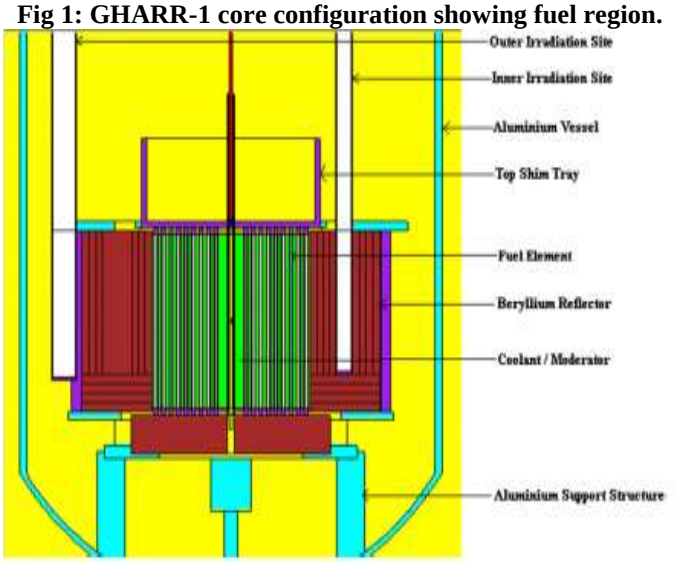

Fig 2: Vertical cross section of GHARR-1 reactor

Theory

In determining the time dependence of nuclide concentrations, ORIGEN2 is primarily concerned with developing solutions for the following equation

$\frac{dN_i}{dt}$ = Formation rate – Destruction rate – Decay rate (1)

ORIGEN (Bell, 1973) considers radioactive disintegration and neutron absorption (capture and fission) as the processes appearing on the right-hand side of Eq. (1). The time rate of change of the concentration for a particular nuclide, Ni, in terms of these phenomena can be written as

$$\frac{dN_i}{dt} = \sum_j \gamma_{ji}\sigma_{f,j}N_j\phi + \sigma_{c,i-1}N_{i-1}\phi + \lambda_i' N_i' - \sigma_{f,i}N_i\phi - \sigma_{c,i}N_i\phi - \lambda_i N_i \quad (2)$$

where (i = 1, ... I), and

$\sum_j \gamma_{ji}\sigma_{f,j}N_j\phi$ = yield rate of Ni due to the fission of all nuclides Nj;

$\sigma_{c,i-1}N_{i-1}\phi$ = rate of transmutation into Ni due to radiative neutron capture by nuclide Ni-1;

$\lambda_i' N_i'$ = rate of formation of Ni due to the radioactive decay of nuclides Ni′

$\sigma_{f,i}N_i\phi$ = destruction rate of Ni due to fission

$\sigma_{c,i}N_i\phi$ = is the destruction rate of Ni due to all forms of neutron absorption other than fission ((n,γ), (n,α), (n,p), (n,2n),(n,3n));

$\lambda_i N_i$ = radioactive decay rate of Ni

Equation (2) is written for a homogeneous medium containing a space-energy-averaged neutron flux, φ, with flux-weighted average cross sections, σ_f and σ_c, representing the reaction probabilities. The flux is a function of space, energy, and time is dependent upon the nuclide concentrations. The mathematical treatment in ORIGEN2 assumes that the space-energy-averaged flux can be considered constant over a sufficiently small time interval, Δt. Similarly, it is assumed that a single set of flux weighted neutron cross sections can be used over the same time step. For a given time step, these assumptions are necessary if Eq. (2) is to be treated as a first-order, linear differential equation. The time-dependent changes in the flux and weighted cross sections are simulated in ORIGEN2 by providing a capability of updating the values for the space-energy-averaged flux and, therefore, for the weighted cross sections for each successive time step, Δtk, Δtk+1, ..., Δtn. These values are derived from lattice cell analyses using physics transport methods to update cross sections that represent the lattice geometry, conditions, and the nuclide concentrations.

Methodology

An input deck was set up and run with the specification of the reactor. Pre-calculated libraries of 1-group cross section data are available for use in ORIGEN2 for several reactor systems. A standard library for an oxide-fueled LWR, the so-called bwrus library, represents the closest potential match to the basic neutronics characteristics of the core. It is preferable to replace cross section data for some or all of the nuclides in the library with data that are more appropriate for the particular system under analysis. For the present analysis, 1- group cross section data were calculated for the fuel rods using the WIMS-ANL code. Replacement cross section data were calculated for only a dozen also selected actinides and fission products. The calculated 1-group capture and fission cross sections at the mid-core life.

ORIGEN-2 code tracks radioactive nuclide inventories for 3 material groups, i.e., activation products, actinides and daughters, and fission products. The radioactive nuclide activities (in curies) are produced both during the irradiation period and cooling period afterward. A large amount of nuclide inventory data is available for analysis. For a bounding hypothetical accident radiological dose evaluation, the maximum value of the radionuclide activities over the whole core life history (including both irradiation period and cooling period) must be extracted if needed.

Table 1: Radioactivity in Curies of Nuclides in Peak Power Pin for HEU344 Core During Irradiation Period of Core Life (Halogens)

HALOGENS	75.0D	150.0D	225.0D	300.0D	375.0D	450.0D	525.0D	600.0D	675.0D	750.0D
BR 84	2.54E+02	2.54E+02	2.54E+02	2.54E+02	2.54E+02	2.54E+02	2.54E+02	2.54E+02	2.54E+02	2.54E+02
BR 85	3.16E+02	3.16E+02	3.16E+02	3.16E+02	3.16E+02	3.16E+02	3.16E+02	3.16E+02	3.16E+02	3.16E+02
BR 86	2.39E+02	2.39E+02	2.39E+02	2.39E+02	2.39E+02	2.39E+02	2.39E+02	2.39E+02	2.39E+02	2.39E+02
BR 86M	2.41E+02	2.41E+02	2.41E+02	2.41E+02	2.41E+02	2.41E+02	2.41E+02	2.41E+02	2.41E+02	2.41E+02
BR 87	5.44E+02	5.44E+02	5.44E+02	5.44E+02	5.44E+02	5.44E+02	5.44E+02	5.44E+02	5.44E+02	5.44E+02
I131	7.19E+02	7.20E+02	7.20E+02	7.20E+02	7.20E+02	7.20E+02	7.20E+02	7.20E+02	7.20E+02	7.20E+02
I132	1.08E+03	1.08E+03	1.08E+03	1.08E+03	1.08E+03	1.08E+03	1.08E+03	1.08E+03	1.08E+03	1.08E+03
I133	1.69E+03	1.69E+03	1.69E+03	1.69E+03	1.69E+03	1.69E+03	1.69E+03	1.69E+03	1.69E+03	1.69E+03
I134	1.91E+03	1.91E+03	1.91E+03	1.91E+03	1.91E+03	1.91E+03	1.91E+03	1.91E+03	1.91E+03	1.91E+03
I135	1.58E+03	1.58E+03	1.58E+03	1.58E+03	1.58E+03	1.58E+03	1.58E+03	1.58E+03	1.58E+03	1.58E+03
I136	7.68E+02	7.68E+02	7.68E+02	7.68E+02	7.68E+02	7.68E+02	7.68E+02	7.68E+02	7.68E+02	7.68E+02
TE132	1.07E+03	1.07E+03	1.07E+03	1.07E+03	1.07E+03	1.07E+03	1.07E+03	1.07E+03	1.07E+03	1.07E+03

Table 2: Radioactivity in Curies of Nuclides in Peak Power Pin for HEU344 Core During Irradiation Period of Core Life (Noble gases)

NOBLE GAS	75.0D	150.0D	225.0D	300.0D	375.0D	450.0D	525.0D	600.0D	675.0D	750.0D
KR 85M	3.19E+02	3.19E+02	3.19E+02	3.19E+02	3.19E+02	3.19E+02	3.19E+02	3.19E+02	3.19E+02	3.19E+02
KR 87	6.45E+02	6.45E+02	6.45E+02	6.45E+02	6.45E+02	6.45E+02	6.45E+02	6.45E+02	6.45E+02	6.45E+02
KR 88	9.12E+02	9.12E+02	9.12E+02	9.12E+02	9.12E+02	9.12E+02	9.12E+02	9.12E+02	9.12E+02	9.12E+02
KR 89	1.16E+03	1.16E+03	1.16E+03	1.16E+03	1.16E+03	1.16E+03	1.16E+03	1.16E+03	1.16E+03	1.16E+03
XE133	1.69E+03	1.69E+03	1.69E+03	1.69E+03	1.69E+03	1.69E+03	1.69E+03	1.69E+03	1.69E+03	1.69E+03
XE135	1.52E+03	1.52E+03	1.52E+03	1.52E+03	1.52E+03	1.52E+03	1.51E+03	1.51E+03	1.51E+03	1.51E+03
XE135M	2.85E+02	2.85E+02	2.85E+02	2.85E+02	2.85E+02	2.85E+02	2.85E+02	2.85E+02	2.85E+02	2.85E+02
XE137	1.50E+03	1.50E+03	1.50E+03	1.50E+03	1.50E+03	1.50E+03	1.50E+03	1.50E+03	1.50E+03	1.50E+03
XE138	1.57E+03	1.57E+03	1.57E+03	1.57E+03	1.57E+03	1.57E+03	1.57E+03	1.57E+03	1.57E+03	1.57E+03

Table 3: Radioactivity in Curies of Nuclides in Peak Power Pin for HEU344 Core During Irradiation Period of Core Life (Alkaline metals)

ALKALINE METALS	75.0D	150.0D	225.0D	300.0D	375.0D	450.0D	525.0D	600.0D	675.0D	750.0D
BA140	1.53E+03	1.56E+03	1.56E+03	1.56E+03	1.56E+03	1.56E+03	1.56E+03	1.56E+03	1.56E+03	1.56E+03
CE141	1.18E+03	1.42E+03	1.47E+03	1.48E+03	1.48E+03	1.48E+03	1.48E+03	1.48E+03	1.48E+03	1.48E+03
CE143	1.48E+03	1.48E+03	1.48E+03	1.48E+03	1.48E+03	1.48E+03	1.48E+03	1.48E+03	1.48E+03	1.48E+03
CE144	2.28E+02	4.17E+02	5.75E+02	7.07E+02	8.16E+02	9.08E+02	9.84E+02	1.05E+03	1.10E+03	1.14E+03
LA140	1.54E+03	1.57E+03	1.57E+03	1.57E+03	1.57E+03	1.57E+03	1.57E+03	1.57E+03	1.57E+03	1.57E+03
MO 99	1.50E+03	1.50E+03	1.50E+03	1.50E+03	1.50E+03	1.50E+03	1.50E+03	1.50E+03	1.50E+03	1.50E+03
NB 95	4.72E+02	1.01E+03	1.32E+03	1.48E+03	1.55E+03	1.58E+03	1.60E+03	1.60E+03	1.61E+03	1.61E+03
ND147	5.67E+02	5.72E+02	5.72E+02	5.72E+02	5.72E+02	5.72E+02	5.72E+02	5.72E+02	5.72E+02	5.72E+02
PM147	2.39E+01	5.27E+01	7.98E+01	1.05E+02	1.30E+02	1.52E+02	1.74E+02	1.94E+02	2.13E+02	2.31E+02
PR143	1.41E+03	1.45E+03	1.45E+03	1.45E+03	1.45E+03	1.45E+03	1.45E+03	1.45E+03	1.45E+03	1.45E+03
RU103	5.81E+02	7.35E+02	7.76E+02	7.87E+02	7.90E+02	7.91E+02	7.91E+02	7.91E+02	7.91E+02	7.91E+02
SR 89	7.74E+02	1.05E+03	1.15E+03	1.18E+03	1.20E+03	1.20E+03	1.20E+03	1.20E+03	1.20E+03	1.20E+03

Table 4: Radioactivity in Curies of Nuclides in Peak Power Pin for HEU344 Core During Irradiation Period of Core Life (Actinides)

ACTINIDES	75.0D	150.0D	225.0D	300.0D	375.0D	450.0D	525.0D	600.0D	675.0D	750.0D
U234	6.86E-02	6.85E-02	6.84E-02	6.84E-02	6.83E-02	6.82E-02	6.82E-02	6.81E-02	6.80E-02	6.79E-02
U235	2.13E-03	2.12E-03	2.12E-03	2.11E-03	2.11E-03	2.10E-03	2.09E-03	2.09E-03	2.08E-03	2.07E-03
U237	2.41E+00	4.63E+00	6.86E+00	9.11E+00	1.14E+01	1.36E+01	1.59E+01	1.82E+01	2.05E+01	2.28E+01
U239	4.56E+01	4.57E+01	4.59E+01	4.60E+01	4.61E+01	4.63E+01	4.64E+01	4.65E+01	4.67E+01	4.68E+01
NP238	1.84E-03	7.32E-03	1.65E-02	2.93E-02	4.59E-02	6.62E-02	9.04E-02	1.19E-01	1.51E-01	1.87E-01
NP239	4.56E+01	4.57E+01	4.59E+01	4.60E+01	4.61E+01	4.63E+01	4.64E+01	4.65E+01	4.67E+01	4.68E+01

Table 5: Peak Power Rod Actinide Activities after Constant Operation for 750 FPED for GHARR-1 followed by 1 Year of Cooling (Ci)

Actinides	Discharge	1day	5days	10days	30days	90days	365days
U-237	6.64E-02	5.99E-02	3.97E-02	2.38E-02	3.05E-03	6.43E-06	7.76E-11
Np-238	5.42E-04	3.91E-04	1.05E-04	2.05E05	2.93E-08	4.83E-14	4.80E-14
Np-239	1.36E-01	1.02E-01	3.15E-02	7.22E-03	2.01E-05	5.38E-13	1.08E-13
Pu-238	2.92E-06	2.93E-06	2.95E-06	2.95E-06	2.95E-06	2.95E-06	2.93E-06
Pu-239	7.58E-06	7.59E-06	7.61E-06	7.61E-06	7.62E-06	7.62E-06	7.62E-06
Pu-240	4.23E-07	4.23E-07	4.23E-07	4.23E-07	4.23E-07	4.23E-07	4.23E-07
Pu-241	3.32E-06	3.32E-06	3.32E-06	3.31E-06	3.31E-06	3.28E-06	3.16E-06

Results and discussion

Calculations were performed by the ORIGEN2 code to obtain the inventory of halogens, noble gases, alkaline metals, and actinides in the peak power fuel rods for GHARR-1 operated at 30kw. The core peak burnup fuel rods were evaluated. The inventory data reported in Appendix tables 1 – 4 can be used in SSDOSE code for radiological assessment of accident scenarios involving the release of radioactive material (e.g., maximum hypothetical accidents, or MHA).

The bounding maximum radioactivity values are always found near the beginning (75 FPEDs) of the irradiation period for halogens and noble gases. However, the bounding maximum activities for alkaline metals and actinides and daughters are almost always found at the end of irradiation period (750 FPEDs).

For the halogen and noble gas radioactivities, the concentrations (and so, activity) of these shorter-lived nuclides reaches a saturation point within ~ 100 days of operation, as production from fission and destruction by radioactive decay balance. For the alkaline metals and actinides, the concentrations do not generally reach saturation during the irradiation because of their longer half-lives. Therefore in order to obtain the bounding hypothetical accident radiological dose evaluation, the maximum values of the radionuclide activities over the whole core life history (including both Irradiation and cooling period) must be examined.

U-238 undergoes a neutron capture to form U-239, which quickly decays by β^- emission to Np-239. Subsequent transmutations of Np-239 lead to even higher actinides and their daughters.

The major contributors (Np-239 and U-237) to the actinide activities at end of irradiation are short-lived radionuclides with half-lives on the order of days. Within a few weeks after shutdown, these nuclides will have largely decayed to longer-lived Pu-238 and Pu-239. . This can be seen in Table 5 which provides the actinide activity inventories in the peak burnup rods after one year of post-irradiation cooling.

For the purpose of assessing the bounding radiological consequences of a release of actinides from fuel material, the maximum doses would be obtained at the time of discharge. Unless the hypothetical accident scenario assumes that it happens right at the precise moment at discharge, it should be reasonable to consider some level of cooling has occurred for accidents involving any kind of operational core access events or maintenance activities. Therefore any level of cooling assumed after discharge would be helpful to reduce the radiological dose levels due to the decay away of those short lived actinides.

The isotopic inventory obtained will also be helpful in the neutronic dose calculation analyses of the spent fuel cask that would be used to store the fuel. The composition of the spent fuel will assist in precise criticality calculations.

Conclusion

The isotopic inventories of some fission products relevant for the safe operation of GHARR-1 have been estimated for its 90.2% HEU fuel. ORIGEN2 code package, a zero-dimensional isotope decay and transmutation code was used to perform the core depletion analysis. The results indicate that the ^{135}Xe isotopic inventory decreases gradually as the fuel is depleted.

However, a build-up trend was observed for fission products ^{239}U and ^{239}Np.

The results would be validated with other cell depletion codes and used as input data in performing detailed 3-D reactor core depletion analysis to adequately estimate the burnup history of the GHARR-1 core, as well as in the analysis of the decay heat removal from the spent fuel storage cask, for which criticality safety and shielding analysis would be performed.

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