SENSITIVITY AND UNCERTAINTY ANALYSIS FOR RADIOTOXICITY ESTIMATION OF THE HALDEN BWR SPENT FUEL

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Abstract

The main objective of the study is the theoretical estimation of long-lived actinide and fission product inventories by combining depletion calculations and non-destructive measurement techniques (measuring of gamma-ray emitting fission products). This is important on account of possible leakage from long-term disposal of spent nuclear fuel. Identification of the most radiotoxic actinides and fission products in different cooling times (especially at long-term) and their behaviour from radiological protection aspects is discussed in the study. The solubility and mobility of long-lived actinides and fission products in underground waters, rocks and soil is very important for the accurate prediction of environmental impacts in the case of leakages from long-term disposal facilities. The results of this research are expected to improve the estimation of long-lived alpha emitting actinides, which is usually measured by destructive methods.

The inhalation and ingestion radiotoxicities are calculated along with the concentration, radioactivity, thermal and gamma power, and neutron and gamma spectra in different cooling times (up to 106 years). Different parameters were varied, such as fuel enrichment, fuel temperature, burnup history, length of cycles, and local irradiation environment, in order to determine their influence on inventories. Variations of operational conditions and local irradiation environment during irradiation have a significant impact on isotopic inventories for the Halden reactor.

Investigation of inhalation and ingestion toxicities of specific actinides and fission products revealed that the larger variations are seen between 3000 – 100 000 years, which is relevant time frame for the postulated leakage from the long-term disposal according to the reported results from previous studies. Preliminary results of this study have shown that the long-lived actinides, such as ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{243}Am, ^{245}Cm, ^{236}Np, ^{237}Np, ^{235}U and ^{238}U and fission products, 79Se, 99Tc, 129I and 135Cs are the important radionuclides in the terms of radiotoxicity, as well as solubility and mobility.

The 6.0% enriched Halden driver fuel irradiated up to 60 GWd/tUO_2 is chosen for this study and is modelled by using SCALE-6.1 codes system. TRITON code is used for the building of Halden driver fuel specific cross section libraries and ORIGEN-ARP is employed for the depletion analysis. The 238-group ENDF/B-VII.0 neutron library is used for the calculations.

1. INTRODUCTION

Accurate estimation of long-lived actinides and fission-products inventories are important for the safety assessment of a geological disposal, and are required to assess the radiological impact on the public and environment from possible leakage of long-term disposal [1–9]. Radiotoxicity is the mostly used indicator in radioactive waste management, because it measures potential radiological hazard of nuclear material. Radiotoxicity is commonly expressed by inhaled or ingested toxicity. Inhalation radiotoxicity is important for fuel handling and transportation, as well as for short-term storage purposes. However, ingestion radiotoxicity is more significant for long-term disposal, because of the greatest biological hazard to humans and environment, when the radioisotope is leaked to nearby underground water. The solubility and mobility of long-lived actinides and fission products in underground waters, rocks and soil is important for the assessment of environmental impacts of spent nuclear fuel and development of accident scenarios [10–13].

Non-destructive techniques based on gamma-ray spectroscopy, using fission products as monitors have been extensively used for the spent fuel analysis for decades. Relatively longer lived fission products, such as

^{134}Cs, ^{137}Cs and ^{154}Eu are most commonly used for the verification of spent fuel time of discharge, burnup, irradiation history and cooling time [14–19].

The current study focuses on theoretical estimation of long-lived actinide and fission product inventories by combining depletion calculations and non-destructive measurement techniques, by measuring of gamma-ray emitting fission products. The ^{134}Cs, ^{137}Cs, ^{154}Eu and ^{95}Zr and their single ratios at 1 year have been used for the estimation of long-lived isotopic inventories at 104 years. The aim of the ongoing project is to demonstrate that non-destructive techniques can predict valuable information about long-lived actinide and fission product inventories after long decay times. Investigation of effects of variations of operational parameters on isotopic inventories is another objective of the study. Identification of the most radiotoxic actinides and fission products in different cooling times and their behaviour from radiological protection aspects are also discussed in the study.

The paper is organized as follows: in Section 2 the method, the driver fuel model and all studied cases are described. The results from depletion calculations and discussion of results are presented in Section 3. Conclusions drawn from the study are summarized in Section 4.

2. METHOD AND MODEL DESCRIPTION

2.1. Method description

The main objective of the present study is to develop the method for the accurate estimation of long-lived actinides and fission product inventories for Halden BWR spent fuel, by combining depletion calculations and non-destructive measurement techniques, based on measuring of gamma-rays emitted from long-lived fission products. Another objective is to investigate the influence of operating parameter variations on radionuclide inventories.

The Halden boiling heavy water reactor (HBWR) has been used for the study of fuel behaviour at high burnup and materials testing since 1959 and is the largest joint project of the OECD – Nuclear Energy Agency [20–21]. The 6.0% enriched Halden BWR driver fuel irradiated up to 60 GWd/tUO_2 was chosen as a Base Case and is modelled by using SCALE-6.1 codes system [22]. TRITON code was used for the building of Halden driver fuel specific cross section libraries and ORIGEN-ARP was employed for the spent fuel analyses [23–24]. The 238-group ENDF/B-VII.0 nuclear data library was used for the calculations [25].

The inhalation and ingestion radiotoxicities for the actinides and fission products were calculated along with the concentration, radioactivity, thermal and gamma power, neutron and gamma spectra in different cooling times (up to 106 years). Different parameters were varied, such as fuel enrichment, fuel temperature, local irradiation environment, burnup and power history, length and number of cycles, in order to study their influence on isotopic inventories. Variations of operational conditions and local irradiation environment during irradiation have a significant impact on isotopic inventories for the Halden reactor. Results related to the radiotoxicity is presented in the current paper, the rest of the study will be published elsewhere.

Firstly, total inhalation and ingestion radiotoxicities from different cases were analysed for the actinides and fission products and compared to the Base Case. Secondly, inhalation and ingestion radiotoxicities have been investigated for the specific actinides, such as ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{243}Am, ^{242}Cm, ^{244}Cm, ^{245}Cm, ^{246}Cm, ^{237}Np, ^{239}Np, ^{235}U, ^{237}U, ^{238}U, ^{230}Th and fission products, ^{79}Se, ^{93}Zr, ^{99}Tc, ^{129}I and ^{135}Cs, to evaluate the impact of variations on the specific actinide and fission product inventories and to determine when the largest variations on the inventory occurs. Lastly, the fission products, ^{134}Cs, ^{137}Cs, ^{154}Eu, ^{95}Zr and their single ratios, ^{134}Cs/^{137}Cs, ^{134}Cs/^{154}Eu, ^{154}Eu/^{137}Cs, ^{134}Cs/^{95}Zr, ^{137}Cs/^{95}Zr, and ^{154}Eu/^{95}Zr after 1 year of fuel discharge have been used for the comparison to the long-lived actinides and fission-products inventories at 104 years.

2.2. Description of the Base Case

The Halden BWR is a heavy water moderated and cooled research reactor with maximum thermal power of 25 MW and the water temperature of 240˚C, corresponding to an operating pressure of 34 bar [26]. The reactor core consists of about 110 UO_2 fuel assembles with various fuel enrichments, in an open hexagonal lattice configuration (i.e. each fuel assembly is surrounded by 3 fuel positions and 3 control rod positions). Each driver fuel assembly consists of 8 (or 9) fuel rods with 6.0% fuel enrichment. Within each fuel assembly, there is heavy water around the pins as well as in the centre. The main difference of Halden reactor from other BWR and

PWR types is that it has a more heterogeneous core configuration and a larger neutron migration area compared to the fuel assembly size. This results in a high sensitivity to fuel configurations in the local surroundings.

The driver fuel assembly with 8 fuel rods, in the centre of a hexagonal two-ring model was chosen for this study and modelled with the 2-dimensional lattice code TRITON. Fig.1 shows the target driver fuel assembly is placed in the middle position of the two-ring model and Table 1 summarizes the composition of the fuel analysed in the study.

Depletion calculations are performed by using ENDF/B-VII.0 nuclear data library, with 238 and 47 neutron and gamma energy groups, respectively. It should be noted that all calculations have been performed at typical irradiation conditions, not actual power history. The initial amount of fissile material corresponds to 1 metric ton of uranium with 6.0% enrichment. In the TRITON burnup simulations, as well as for the ORIGEN-ARP spent fuel analysis, the reactor power was assumed to be 40 MW/t and the burnup up to 60 GWd/tUO_2. The burnup of the target assembly was calculated in 13 different burnup levels, from 0 to 60 GWd/tUO_2 with five updates to the cross sections in each step. Halden driver fuel specific burnup-dependent cross section libraries computed by TRITON were later employed by ORIGEN-ARP for spent fuel analysis.

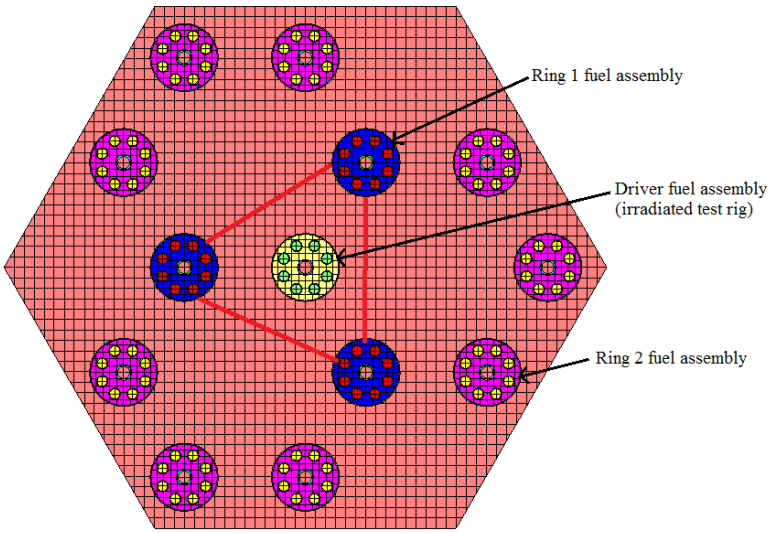


Fig.1. TRITON model of local surroundings

The ORIGEN-ARP calculations are performed by assuming that the driver fuel was irradiated for 1500 days in 3 cycles, with 500 days in each with an intermediate decay of 100 days to obtain the final composition of the spent fuel at discharge burnup of 60 GWd/tUO_2. After the discharge, the inhalation and ingestion radiotoxicities, the concentration, the radioactivity, the thermal and gamma power, the neutron and gamma spectra were evaluated for the actinides and fission products until 106 years.

TABLE 1. DRIVER FUEL SPECIFICATIONS

Fuel	
Material	UO_2
Enrichment, %	6.0
Density, g/cm^3	10.3
Temperature, K	700
Radius, cm	0.5335
Half pitch, cm	1.0
Cladding	
Material	Zr-2
Temperature, K	600
Radius, cm	0.6125
Coolant	
Material	D_2O, H_2O
Temperature, K	500
Density D_2O, g/cm^3	0.40
Density H_2O, g/cm^3	0.01

2.3. Description of the other cases

One of the objectives of this study is the investigation of effects of the operational parameter variations on radionuclide inventories. The following cases have been modelled and analysed in the study:

(a) Fuel temperature study

Fuel temperature study is important for the reliable prediction of fuel rod behaviour and for the overall fuel performance assessment. Two temperature cases have been investigated, 300K (case 1) and 1100K (case 2), respectively. The new cross section libraries were developed for both cases and employed for the spent fuel analysis.

(b) Fuel enrichment study

Taking into consideration the influence of initial enrichment on the build-up of isotopes in the fuel assembly, four enrichment cases have been analysed in the study and described in Table 2. The new cross section libraries were developed for all cases and employed for the ARP calculations.

TABLE 2. DESCRIPTION OF ENRICHMENT CASES

Case	Fuel enrichment, %
1	6.5
2	5.5
3	5.7
4	6.2

Surrounding study

Irradiation environment has significant impact on isotopic inventories of the Halden BWR. Two surrounding cases have been studied, by adding an extra (case 1) and three extra driver fuel assemblies to the Ring 1 (case 2), respectively. The number of fuel assemblies on Ring 2 has been kept the same as the Base Case. Fig.2 describes the TRITON model of the local surroundings of modelled cases. The new cross section libraries were developed for both cases and employed for the spent fuel analysis.

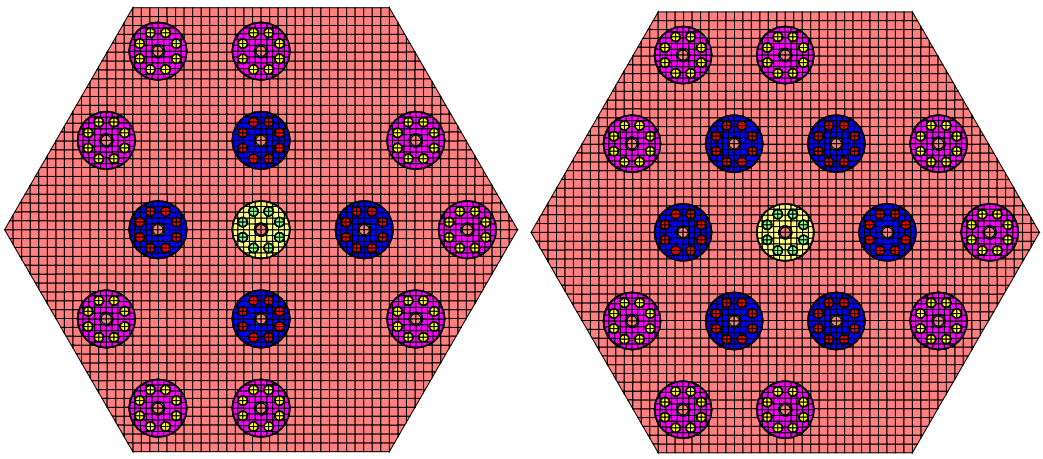


Fig.2. Surrounding study cases; Case 1) 4 fuel assemblies on Ring 1; Case 2) 6 fuel assemblies on Ring 1

Burnup study

Fuel burnup analysis is not only important for the assessment of fuel performance in the core, but also for overall reactor safety and for the design of long-term storage facilities. Seven burnup cases have been investigated in the study. It should be noted that the ARP spent fuel analyses performed by using the same cross section library as a Base Case. Details of variations made for burnup study are presented in Table 3.

TABLE 3. DESCRIPTION OF BURNUP CASES

Case	Power (MW/t)	Burnup (GWd/t)	Number of cycles	Irradiation per cycle (days)	Intermediate decay (days)
1	40	60	5	300	100
2	40	60	3	500	-
3	20	60	10	300	-
4	44	66	3	500	100
5	36	54	3	500	100
6	44	60	3	454.6	100
7	36	60	3	555.6	100

3. RESULTS AND DISCUSSION

3.1. Total inhalation and ingestion radiotoxicity for actinides and fission products

Analyse of total inhalation radiotoxicities have shown that the radiotoxicity of actinides are notably higher than that of fission products for all studied cases and the studied cooling time range. In contrast, total ingestion radiotoxicity of fission products appears higher than the radiotoxicity of actinides under 102 years. However, afterwards, the contribution of fission products to total ingestion radiotoxicity decreases notably compared to actinides, which has seen in all compared cases.

The total inhalation and ingestion radiotoxicities are determined by the following radionuclides in different cooling times. Among the actinides, the primary contributors of radiotoxicity are ^{238}Pu, ^{241}Pu, ^{242}Cm, ^{244}Cm and ^{239}Np in the short-term (<102 years), ^{239}Pu, ^{240}Pu, ^{242}Pu, ^{241}Am, ^{243}Am and ^{230}Th in the medium-term (102–105 years) and ^{242}Pu, ^{237}Np, ^{229}Th, ^{230}Th and ^{233}U in the long-term (106 years and beyond). Among the fission products, the main contributors of radiotoxicity are ^{95}Zr, ^{95}Nb, ^{106}Ru, ^{134}Cs, ^{141}Ce and ^{144}Ce in the short-term (<10 years), ^{90}Sr, ^{90}Y, ^{121}Sn, ^{121m}Sn, ^{137}Cs, ^{151}Sm and ^{154}Eu in the medium-term (10–102 years) and ^{79}Se, ^{93}Zr, ^{99}Tc, ^{129}I and ^{135}Cs in the long-term (103–106 years).

Comparisons of total inhalation radiotoxicities from different cases to Base Case have shown that the variations make the difference between 25–40% on total actinide and 15–70% difference on total fission product inventories. While, the difference for total ingestion radiotoxicity of compared cases is between 20–70% for actinide and 10–80% for fission product inventories.

3.2. Actinide inventories

After the comparison of total inhalation and ingestion radiotoxicities, the specific actinides are analysed in order to find their contribution to the total radiotoxicities and investigate the influence of variations on specific radionuclide inventories.

The study of ^{239}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu have revealed that the plutonium isotopes have the largest contribution to the total inhalation and ingestion radiotoxicities and actinide inventories, as previous studies report [27–28]. Comparisons of different cases with Base Case have revealed that the variations make the difference between 50–300% on ^{239}Pu, 45%-130% on ^{240}Pu, 50%-150% on ^{241}Pu and 20%-40% on ^{242}Pu inventories. The largest variations for plutonium isotopes caused by the change of burnup conditions and irradiation history and are seen between 103–105 years.

Analysis of ^{241}Am and ^{243}Am has shown that they have the second largest contribution to the total inhalation and ingestion radiotoxicities, as well actinide inventories for the first 103 years. However, the long-term contributions of americium isotopes are still high. Comparisons of different cases with Base Case have revealed that the variations make the differences between 50%-150% on ^{241}Am and 80%-300% on ^{243}Am inventories and the largest variations for americium isotopes are seen between 103–105 years.

The study of curium isotopes has demonstrated that they have third largest contribution to the radiotoxicity and actinide inventories. Comparisons of different cases with Base Case have shown that the curium inventories are also mainly affected by burnup and irradiation history changes in the range of 50–230%.

According to the results the fourth largest contributor of radiotoxicity and actinide inventory is neptunium isotopes. Comparisons of different cases with Base Case have demonstrated that the variations make the difference between 10–70% on ^{237}Np inventory.

Analysis of other actinides has shown that at the long-term radiotoxicity and inventory is dominated by uranium isotopes and daughters along with ^{242}Pu, because of their long half-life. Comparisons of different cases with Base Case have shown that the variations make the 40–100% differences on uranium inventories.

Study of actinides has revealed that the variations make huge differences on specific actinide inventories and all analysed actinides are mainly sensitive to the change of burnup conditions and irradiation history.

3.3. Fission product inventories

Analysis of long-lived ^{79}Se, ^{93}Zr, ^{99}Tc, ^{129}I and ^{135}Cs has shown that they are primary contributors of long-term radiotoxicity and fission product inventories. The ^{9}9Tc and ^{129}I are major radiotoxic contributors among the long-lived fission products. Comparisons of different cases with Base Case have shown that the variations make around 10% difference on long-lived fission product inventories and are sensitive to the change of burnup conditions and irradiation history like actinide inventories.

3.4. Gamma-ray spectroscopy for the long-lived isotopic inventory estimation

High gamma-ray intensity and relatively longer half-life, as well as domination of the gamma-ray spectrum of spent fuel for the first 10 years of cooling time make the ^{134}Cs, ^{137}Cs and ^{154}Eu the best candidates for the spent fuel analysis by gamma spectroscopic methods. Because of the high gamma-ray intensity the short-lived ^{95}Zr was also included in the study. The ^{134}Cs, ^{137}Cs, ^{154}Eu and ^{95}Zr and their ratios after 1 year of discharge are used for the estimation of long-lived actinide and fission product inventories at 104 years, while operational conditions are varied.

To study the correlation between fission product gamma emissions and long-lived actinide and fission product concentrations the Pearson correlation coefficients are calculated and presented in Table 4 for the fission products and in Table 5 for their ratios. When the coefficient value lies between ±0.50 and ±1, then it is strong correlation, which means linear relation between two variables. When the value is between ±0.30 and ±0.50, it is moderate correlation and below ±0.30 is weak correlation. Values closer to zero indicates no correlation. Correlation plots for some actinides and fission products are demonstrated in Fig. 3 and Fig. 4, respectively.

TABLE 4. ESTIMATION OF LONG-LIVED ISOTOPIC INVENTORIES AT 10^4 YEARS VERSUS FISSION PRODUCTS AT 1 YEAR

Isotope	^{134}Cs	^{137}Cs	^{154}Eu	^{95}Zr
^{239}Pu	0.27	0.18	0.64	0.14
^{240}Pu	0.58	0.73	0.63	-0.01
^{241}Pu	0.77	0.76	0.86	0.12
^{242}Pu	0.84	0.83	0.75	0.29
^{241}Am	0.77	0.76	0.86	0.12
^{243}Am	0.76	0.80	0.80	0.08
^{245}Cm	0.83	0.78	0.86	0.27
^{237}Np	0.52	0.47	0.82	0.21
^{235}U	-0.73	-0.75	-0.60	-0.28
^{238}U	-0.77	-0.86	-0.76	-0.37
^{230}Th	0.67	0.62	0.91	0.20
^{79}Se	0.76	0.88	0.74	0.42
^{93}Zr	0.83	0.95	0.70	0.41
^{99}Tc	0.80	0.93	0.66	0.42
^{129}I	0.92	0.96	0.87	0.37
^{135}Cs	0.08	0.13	0.42	-0.26

According to the Table 4, ^{134}Cs, ^{137}Cs and ^{154}Eu have shown better correlation on the estimation of most analysed actinides and fission products. ^{95}Zr has shown weak correlation with actinides, but moderate correlation with long-lived fission products. ^{154}Eu gives better correlation on estimation of plutonium isotopes in comparison of ^{134}Cs and ^{137}Cs. ^{95}Zr has shown weak correlation with plutonium isotopes and no correlation with ^{240}Pu. Because of depletion a strong but negative correlation is seen for ^{235}U and ^{238}U. The ^{137}Cs has shown

better correlation with both uranium isotopes. The ^{134}Cs, ^{137}Cs and ^{154}Eu have shown strong correlation with all long-lived fission products estimation, except ^{135}Cs.

TABLE 5. ESTIMATION OF LONG-LIVED ISOTOPIC INVENTORIES AT 10^4 YEARS VERSUS FISSION PRODUCT RATIOS AT 1 YEAR

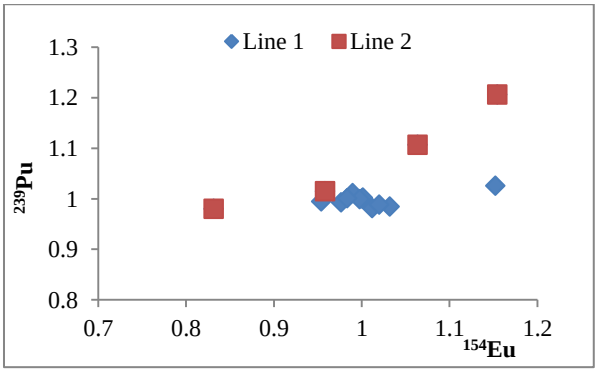
Isotope	^{134}Cs/^{137}Cs	^{134}Cs/^{154}Eu	^{154}Eu/^{137}Cs	^{134}Cs/^{95}Zr	^{137}Cs/^{95}Zr	^{154}Eu/^{95}Zr
^{239}Pu	0.31	-0.19	0.82	0.02	-0.11	0.06
^{240}Pu	0.47	0.32	0.39	0.43	0.19	0.26
^{241}Pu	0.71	0.40	0.70	0.37	0.03	0.17
^{242}Pu	0.78	0.67	0.51	0.75	0.40	0.56
^{241}Am	0.71	0.40	0.70	0.37	0.03	0.17
^{243}Am	0.69	0.46	0.58	0.43	0.09	0.20
^{245}Cm	0.79	0.50	0.70	0.74	0.37	0.67
^{237}Np	0.52	-0.06	0.89	0.45	0.17	0.68
^{235}U	-0.68	-0.67	-0.35	-0.65	-0.36	-0.43
^{238}U	-0.69	-0.53	-0.50	-0.64	-0.34	-0.52
^{230}Th	0.66	0.12	0.89	0.61	0.31	0.76
^{79}Se	0.67	0.55	0.47	0.63	0.32	0.49
^{93}Zr	0.73	0.74	0.35	0.70	0.38	0.44
^{99}Tc	0.69	0.73	0.30	0.66	0.36	0.40
^{129}I	0.84	0.67	0.60	0.80	0.43	0.64
^{135}Cs	0.07	-0.40	0.53	0.21	0.37	0.56

The ^{134}Cs/^{137}Cs, ^{134}Cs/^{154}Eu and ^{154}Eu/^{137}Cs ratios have shown better correlation on the estimation of most actinides and fission products compared to the ratios with ^{95}Zr, as demonstrates Table 5. The ^{154}Eu/^{137}Cs ratio has shown stronger correlation with ^{239}Pu, ^{237}Np and ^{230}Th. However, the ^{134}Cs/^{137}Cs ratio has shown better correlation with other Pu isotopes, as well as with rest of actinides. The ^{134}Cs/^{137}Cs, ^{134}Cs/^{137}Cs and ^{134}Cs/^{95}Zr ratios have stronger correlation with long-lived fission products.

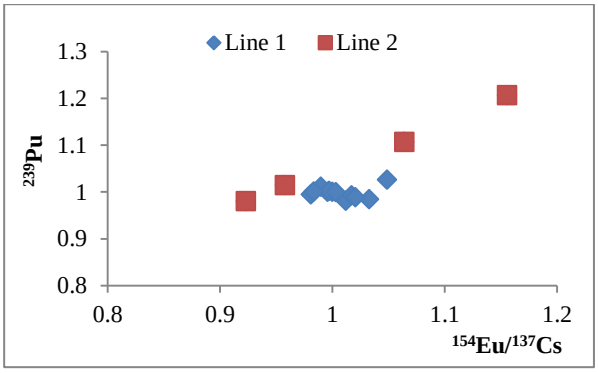
Analysis of the correlation between fission products have shown that the ^{134}Cs, ^{137}Cs and ^{154}Eu correlates better with each other compared with ^{95}Zr, according to Table 6.

TABLE 6. CORRELATION COEFFICIENT OF FISSION PRODUCTS

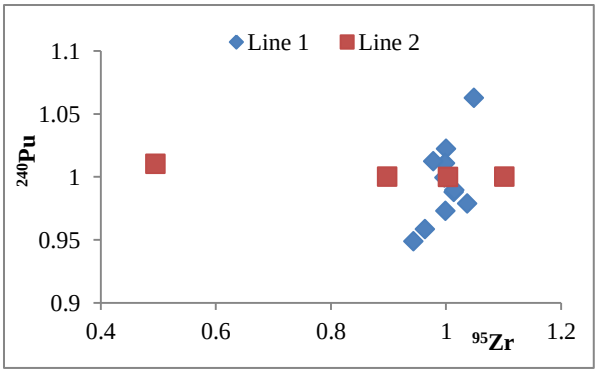
Fission products	Correlation
^{134}Cs vs ^{137}Cs	0.91
^{134}Cs vs ^{154}Eu	0.85
^{154}Eu vs ^{137}Cs	0.80
^{134}Cs vs ^{95}Zr	0.50
^{137}Cs vs ^{95}Zr	0.38
^{154}Eu vs ^{95}Zr	0.19



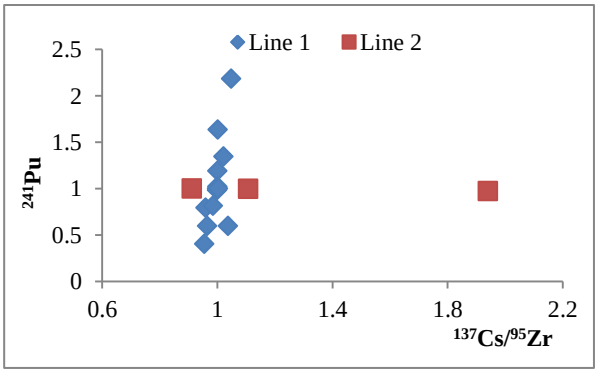
a) Strong correlation



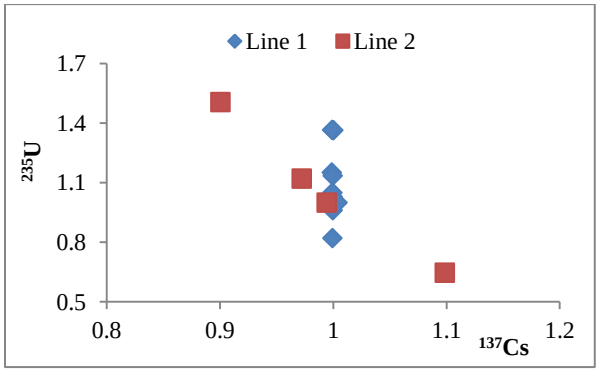
b) Strong correlation



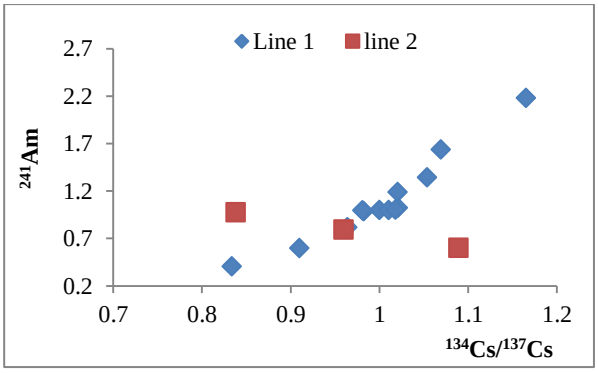
c) No correlation



d) Weak correlation



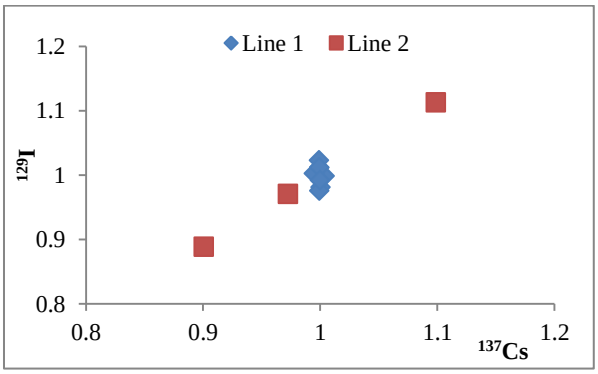
e) Strong correlation



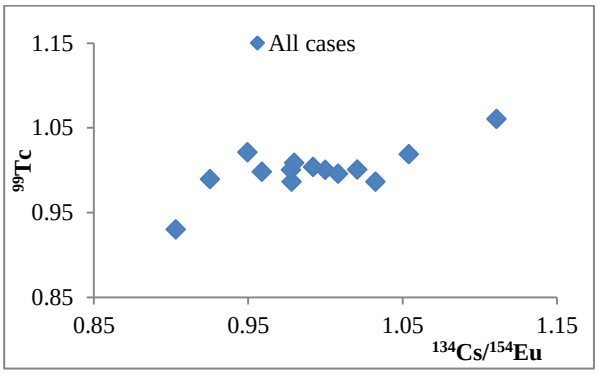
f) Strong correlation

Fig.3. Examples of actinides estimation: a (and b)) Line 1- the other cases, Line 2- Surrounding case 1 and 2, Enrichment case 1 and Bu case 5; c) Line 1- the other cases, Line 2- Bu cases 2, 3, 6 and 7; d) a) Line 1- the other cases, Line 2- Bu cases 3, 6, 7; e) Line 1- the other cases, Line 2- Bu cases 1, 4, 5 and 7; f) Line 1- the other cases, Line 2- Bu cases 1,3 and Enrichment case 2

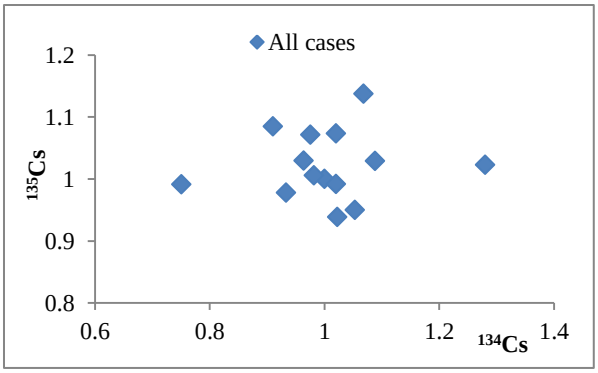
Figures 3 and 4 demonstrate that there are two distinct types of behaviour in most of plots with large uncertainties, which means the build-up of actinides and fission products is a very complex process and sensitive to the variations. According to the Fig. 3, the ^{239}Pu and ^{241}Am inventories are sensitive to surrounding, enrichment and irradiation history variations. ^{240}Pu, ^{241}Pu and ^{235}U inventories are sensitive only to the burnup and irradiation history changes. Fig. 4 demonstrates that long-lived fission products have also shown similar behaviour to the actinides, except ^{135}Cs. Investigation of long-lived fission products revealed that they are also sensitive to the burnup and irradiation history changes as most of actinides.



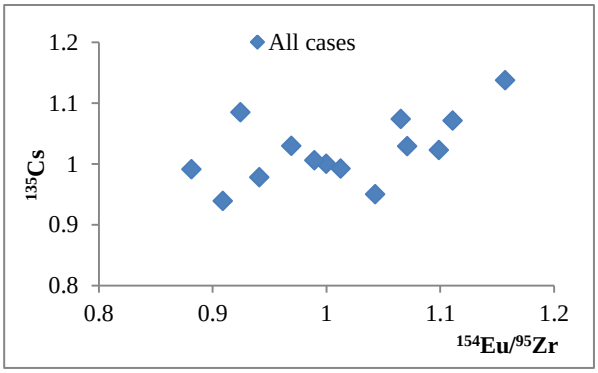
a) Strong correlation



b) Strong correlation



c) Weak correlation



d) Weak correlation

Fig.4. Examples of fission products estimation: a) Line 1- the other cases, Line 2- Bu cases 1, 4 and 5; b, c and d) All cases

Analysis of plots has revealed that the production of ^{137}Cs is linearly dependent on the neutron flux and the burnup, while the build-up of ^{134}Cs and ^{154}Eu quadratically depends on flux density and the burnup. To explain the behaviour seen in the plots, the production and destruction mechanisms of actinides and fission products will be further analysed.

3.5. Radiological impacts of long-lived actinides and fission products

Results of this study have shown that radiotoxicity of actinides is notably higher than that of fission products. Most preferred final disposal of spent nuclear fuel from radiation protection and environmental consideration is deposition of highly radioactive and radiotoxic fuel in canisters and in capsules in deep underground repository. The strongest environmental impact would be expected if radionuclides escaped to the groundwater at the earlier stages of disposal, when it contains short and medium lived radionuclides.

The spent fuel in an underground repository can interact with groundwater only if the engineered barriers fail. The geochemical mobility of actinides largely depends on redox conditions. Oxidation state has a dramatic effect on solubility, mobility, bioavailability and toxicity of actinides. The formation of local oxidizing conditions in the spent fuel repositories induced by the radiolysis or some other processes is not probable under normal conditions, because the host medium contains reducing agents. The UO_2 stability is very important for the repository safety. When the geological medium becomes the only barrier retaining the radionuclide release, the repository safety in most cases will be determined by the intensity of the Pu and Am which escapes from the spent fuel. If the oxidizing conditions are present, the Pu and Am migration in dissolved form in concentrations hazardous for the environment will be precluded by their rapid sorption on minerals. The Np and Cm are the following important actinides with risk to the environment, after the Pu and Am. U has very low solubility and mobility compared to the above stated four actinides [29-31].

Long-lived fission products appear to be less radioactive and radiotoxic compared to the actinides, but they are more soluble and mobile in the environment than actinides, especially ^{99}Tc, ^{129}I and ^{135}Cs. ^{99}Tc has a long half-life and relatively high fission yield is the most abundant, highly soluble in water and in soil and a key mobile radionuclide in high level radioactive waste [32]. Technetium is a redox sensitive radionuclide and its migration behaviour depends on its oxidation state. The long-term mobility and radiotoxicity (the most radiotoxic when ingested) makes ^{129}I the second hazardous long-lived fission product [33]. The distribution and long-term environmental mobility of iodine have been studied by many researchers in order to predict its build up and potential radiological impact. Other long-lived, chemically and radiologically toxic fission products are ^{135}Cs and ^{79}Se [34-36]. Because of the high solubility and mobility of caesium and selenium in oxidizing environments, their behaviour is important in safety analysis of radioactive waste repositories.

4. CONCLUSIONS

The current study focuses on theoretical estimation of long-lived actinide and fission product inventories for the Halden BWR spent fuel by combining depletion calculations and gamma-ray spectroscopy. ^{134}Cs, ^{137}Cs, ^{154}Eu and ^{95}Zr and their single ratios at 1 year have been used for the estimation of long-lived

isotopic inventories at 104 years. It is concluded that ^{137}Cs and ^{154}Eu are better candidates on the isotopic inventory estimation compared to ^{134}Cs and ^{95}Zr. These promising preliminary results of this ongoing study presented in this paper are based on simulated data only. However, the approach taken in this study for the estimation of long-lived actinide and fission product inventories will be further analysed by using more variations and multivariate analysis and validated against experimental data.

Sensitivity and uncertainty analysis is performed by the variations of operational parameters in this study. Fifteen cases have been studied in comparison with the Base Case. The study has shown that all actinides and fission product inventories are sensitive to the burnup and irradiation history variations. Study has revealed that the ^{239}Pu inventory is sensitive to surrounding variations, due to the larger neutron migration area in heavy water moderated reactors. ^{239}Pu inventory is also sensitive to enrichment variations. However, more realistic smaller enrichment variations (under ±0.5%) do not make visible deviations on actinide inventories. The fuel temperature variations analysed also have insignificant effect on the actinide and fission product inventories. The resonance absorption processes are less dominant in the HBWR compared to interactions with the thermal neutrons.

Study of total inhalation radiotoxicities of the cases compared have shown that the variations make the difference between 25%-40% on total actinide and 15%-70% difference on total fission product inventories. While, the difference for total ingestion radiotoxicity of compared cases is between 20–70% for actinide and 10–80% for fission product inventories. The primary contributors of radiotoxicity of actinides and fission products are identified at short, middle and long-term. Study of specific actinides has revealed that the variations of parameters make the difference 10% to 300% on actinide inventories, for the long-lived fission product inventories is around 10%.

Review of literature has shown that long-lived actinides and fission products remain radiologically toxic after several thousand years and can cause harm to humans and environment. Radiotoxicity of actinides is notably higher than that of fission products, but they are less soluble and mobile compared to the long-lived fission products. Oxidation state has a dramatic effect on solubility, mobility and toxicity of both actinides and fission products.

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