Assessments of radiation emission from molten salt reactor spent fuel: Implications for future nuclear safeguards verification

Vaibhav Mishra*[1], Erik Branger[1], Sophie Grape[1], Markus Preston[1], Zsolt Elter*[1,2], and Sorouche Mirmiran[2]

[1]*Department of Physics and Astronomy, Uppsala University, Uppsala, Sweden*
[2]*Seaborg Technologies, Titangade 11 2200, Copenhagen, Denmark*

Submitted: May 11, 2023

Abstract

Safeguards verification using non-destructive assay (NDA) techniques is an important pillar of the safeguards regime to ensuring that nuclear technology is not used for non-peaceful purposes. The methods and approaches for safeguards verification for conventional spent nuclear fuel (SNF) originating from the global fleet of water-cooled reactors are well-established. However, for reactors such as molten salt reactors (MSR), accountancy verification method of irradiated fuel salts is not quite well-established. This is primarily since the irradiated salt is in "bulk form" whereas more conventional LWR SNF encountered by the safeguards inspectors is in "item form". Moreover, much about the nature of such SNF still remains unknown due to the lack of operational MSRs and equipment adequate to further study, develop and test NDA verification methods.

As MSRs could play a complementary role with the existing fleet of reactors in the near future, verification methods concerning the nature of emissions from irradiated fuel salts is timely. Therefore, in this current study we aim to quantify and study the nature of gamma and neutron emissions as well as decay heat production in irradiated fuel salt from the Compact Molten Salt Reactor (CMSR) developed by Seaborg Technologies in Denmark. Simulations were carried out using the Monte-Carlo particle transport code Serpent as well as the code SOURCES 4C to compute nuclide inventories and the associated emission rates of gamma and neutron (from spontaneous fissions, or SF and from (α, n) reactions) emissions and decay heat calculations. These results will shed more light on the implications for nuclear safeguards verification for irradiated fuel salts and also highlight some of the challenges and opportunities associated with detecting and characterizing the emissions using NDA methods in the future for SNF of such unique nature.

Keywords: *Molten Salt Reactors, nuclear safeguards verification, Serpent, SOURCES 4C.*

1 Introduction

After years of research between 1940-1970 into the viability of Molten Salt Reactors (MSR), there has been a resurgence of interest in the scientific community in this concept. MSRs offer several advantages over conventional Light Water Reactors (LWRs) some of which include the ability to operate at much higher temperatures (thereby improving efficiency) without the risk of losing physical integrity of the fuel Elsheikh. Most MSRs use alkali-halide salts i.e. fluoride salts (using fluorine, beryllium, lithium) for instance FLiBe and FUNaK, chloride salts (similar to fluoride), nitride salts (proposed candidates for breeder-type MSRs) et cetera Sohal et al.. Nearly all of these salt compositions can dissolve the fuel material such as U^{235} which is beneficial as it eliminates the need for use of another medium to house the fuel thereby greatly simplifying the design. The same salt, without the fissile or fertile material, is often used in the secondary circuits for many proposed MSR designs for heat exchange. Although a lot of research has been directed towards understanding the reactor concept and the nature of molten salts, a lot remains to be explored, particularly from a nuclear safeguards point of view. This is mainly since molten salt fuel differs in its physical and chemical properties from conventional solid SNF Worrall et al., Worrall et al., Kovacic et al.. Additionally, since these reactors are so different (in terms of design and operation), the radiation emission from irradiated molten salts needs to be investigated in order to guide the development of future frameworks for safeguards verification to ensure states' compliance with international agreements. There is a consensus within the community

*Correspondence: vaibhav.mishra@physics.uu.se, zsolt.elter@seaborg.com

that the implementation of safeguards approaches in the future should not not hinder the deployment of such reactors while still provide safeguards assurances at a level similar to or superior to those for operating LWRs. Therefore, as a first step in this direction, the current study looks into investigating the nature of gamma and neutron emissions produced by irradiated salts from one such MSR concept being developed by Seaborg Technologies in Denmark. The concept, named the Compact Molten Salt Reactor (CMSR) Pater et al., Al-Dbissi, Dos et al. is presently in the detailed design stage and is interesting as it adds modularity and portability in addition to all advantages offered by a traditional MSR. The study will make use of the Monte-Carlo particle transport code Serpent Leppänen et al. and the code SOURCES 4C Wilson et al. for computing the nuclide inventories, radiation emission rates and other safeguards-relevant characteristics for a thorough qualitative and quantitative assessment. The findings from the study will help build a dataset or a fuel library with the use of the two calculation codes. It is expected that the results from this study will help us better understand safeguards-relevant properties of irradiated fuel salts after operation and by extension, the insights can be extrapolated to other similar type of MSR reactors that may operate in the future. Additionally, these findings will assist safeguards inspectors to frame NDA measurement routines to better suit this kind of SNF.

2 Reactor design and specifications

The reactor design used in the present study is based on the CMSR which falls under the umbrella of liquid-fuelled type, Gen-IV reactors that will build on the concepts introduced in the 1960's for the Molten Salt Reactor Experiment (MSRE) that was once operational at the Oak Ridge National Laboratory (ORNL). The reactor will be a 250 MWth (and 100 MWe) fueled with HA-LEU (High Assay-Low Enriched Uranium) and moderated with liquid sodium hydroxide. The primary salt used in the core will be a FUNaK type fuel (chemical composition of NaF-KF-UF_4) which can also be used in the secondary circuit (without the fissile material dissolved in it). The structural material used in the reactor core is a proprietary high-nickel alloy (with a specialized coating) that is known to be corrosion resistant against the mixed-halide fuel as well as the moderator. The reactor is designed to operate on a specially-designed barge that can sail to the desired location, anchored, and supply electricity to the grid on-site. The reactor is fuelled to operate for a period of 12 years (including periods of shutdown and maintenance) and in order to achieve such a long fuel cycle, there are provisions for online removal of gaseous fission products by an Off-Gas System (OGS). Some of the important parameters of the reactor relevant to the study are shown in Table 1. It is important to point out that the findings in the present study are focused solely on the presence and nature of irradiated fuel salt material in the reactor core (and not outside the active core region). Further details of the CMSR design are also available in Pater et al., Al-Dbissi.

Table 1: Operational specifications of the CMSR relevant for this study.

Parameter Name	Parameter Value
Power rating	100 MWe (250 MWth)
Reactor type	Molten Fluoride Salt
Operating temperature	650℃
Neutron spectrum	Thermal
Moderator	NaOH
Convertor/breeder	Converter
Salt type	FUNaK
Primary driver	U^{235}
Structural material	High Ni-alloy
Salt density	4200 kg/m^3
Salt volume	25 m^3
Enrichment	HA-LEU
Time in reactor	12 years
Online refueling	No
Off-Gas System (OGS)	Present[1]

Table 2: Key parameter ranges used in the dataset.

Parameter Name	Parameter Range
Burnup (B)	0.0 – 29.0 MWd/kgU 75 steps in total
Initial enrichment (I)	10.0 – 20.0 wt. % ^{235}U 41 steps in total
Cooling time (C)	0 – 40 years 101 steps in total

*Correspondence: vaibhav.mishra@physics.uu.se, zsolt.elter@seaborg.com

3 Modelling and assumptions

The present study aims at modeling the design and operation of the CMSR core and computing emission rates of gamma, neutron (from SF, and (α, n) in the fuel), and decay heat production to compare them with more conventional Pressurized Water Reactor (PWR) spent fuel. These quantities are often relied upon by nuclear safeguards inspectors for verification of SNF using NDA methods. For instance, gamma activity measurements can be used to deduce fuel BU and CT while neutron measurements can be used to assay fuel's fissile content and thereby determine its IE. For the assessments carried out in the present study, the code SOURCES 4C Wilson et al. and the Monte-Carlo particle transport code, Serpent Leppänen et al. (version 2.1.28) were used. Serpent was used to create a 3-dimensional model of the CMSR core and to compute the nuclide inventories, decay heat (DH) and gamma emission rate (GE) after irradiation of the fuel salt. These quantities will be compared to those from LWR SNF (both UOX and MOX type). The code SOURCES 4C was thereafter used to calculate the (α, n) and SF neutron emission rates. The methodology and assumptions are explained in greater detail in the following sub-sections.

3.1 Isotopes of interest

From numerous previous studies conducted for different fuel types (Hermann and Alexander, Jansson, Fast et al., Preston et al., Tanskanen), it is well-established that certain nuclides fit the criteria for nuclear safeguards verification owing to their relative abundance in SNF, longer half-life and emissions that are detectable with traditional NDA instruments. For instance, Cs^{137} is an important nuclide that is indicator of fuel BU (as it is a direct fission product) and has been used over the decades to verify operator declarations of SNF. Some other nuclides that are important for verification of PWR SNF are listed in Table 3. Similarly, it is known that some isotopes in the SNF contribute more extensively to the decay heat produced after discharge than others Jansson. As before, Cs^{137} is again one of the primary contributors to decay heat and remains so for several hundred years. Other nuclides important to decay heat considerations surrounding PWR SNF are listed in Table 4.

Table 3: Gamma-emitting nuclides important for safeguards and their respective half-lives (data from Bé et al.). Table from Mishra et al..

Nuclide	Half-life	Nuclide	Half-life
Nb^{95}	34.991 *d*	Zr^{95}	64.032 *d*
$^{\dagger}Ru^{106}$	371.5 *d*	Cs^{134}	2.064 *y*
Cs^{137}	30.05 *y*	Ce^{141}	32.5 *d*
Ce^{144}	284.89 *d*	Eu^{154}	8.601 *y*

† In secular equilibrium with Rh^{106}.

Table 4: Isotopes important for decay heat prediction used in present study and their respective half-lives (data from Bé et al.). Table from Mishra et al. and list of isotopes from Jansson.

Nuclide	Half-life	Nuclide	Half-life
Sr^{90}	28.8 *y*	$\bigstar Y^{90}$	64.2 *h*
$^{\ddagger}Ba^{137m}$	2.6 *min*	Pu^{238}	87.7 *y*
Pu^{239}	24100 *y*	Pu^{240}	6561 *y*
Am^{241}	432.2 *y*	Cm^{244}	18.1 *y*

$\bigstar$ Daughter of Sr^{90}, ‡ Daughter of Cs^{137}.

3.2 Calculations using Serpent

The BU[2] calculations performed in the current study were carried out in Serpent. The fresh[3] fuel salt was modeled in the 3-D model of the CMSR core and its irradiation was simulated over a broad range of BIC parameters (ranges shown in Table 2) in order to create a dataset of quantities such as nuclide inventories, gamma emissions etc. The core model comprised of 235 fuel channels surrounded by the structural material (proprietary cladding material and associated specialized corrosion-resistant surface coatings) as well as the moderator (NaOH) in a hexagonal arrangement. The fuel salt channel accommodates the FUNaK fuel salt belonging to the primary circuit and allows its flow through the core. The reactor core also accommodates control rods in designated locations however the control rods were not included in the Serpent model. As described above, input decks of all possible combinations of the BIC parameters from Table 2 were prepared and run on a Linux-based computer cluster. The JEFF-3.1 Koning et al. nuclear data library for cross sections along with ENDF/B-VIII.0 Herman decay and neutron fission yield libraries were used for the computations. A total of 400K neutron histories[4] were simulated for each BIC combination[5] to ensure convergence be keeping the nuclide inventory statistical errors below 1%. The Serpent runs produced output files that contain (among other quantities), nuclide inventories, and nuclide specific gamma source strength (in photons/second), decay heat (in Watts), and SF rates (in fissions/second). It is important to note that while the Serpent

*Correspondence: vaibhav.mishra@physics.uu.se, zsolt.elter@seaborg.com

calculations can provide an estimate of SF rates in the spent fuel, it does not directly compute the neutron yields from these fissions. This means that the user must scale them with the nuclide specific SF neutron multiplicities (ν_{SF}). The code SOURCES 4C however, computes the SF neutron emission rates directly while computing the (α, n) rates which is why both (α, n) and SF neutron emission rates were computed with SOURCES 4C instead.

3.3 Calculations using SOURCES 4C

SOURCES 4C was developed in 1981 by Los Alamos National Laboratory (LANL) and allows users to compute rates and energy spectra of (α, n) reactions in a user-supplied composition of low-Z materials and α sources. It can also compute SF neutron emission rates for the same user-supplied problem. SOURCES 4C does not require any details of the system geometry in order to carry out its computations for a homogeneous problem i.e. one in which the low-Z nuclides and α sources (such as Pu^{238}, Am^{241} et cetera) are mixed together. It uses four library files (or '*tapes*') which include stopping cross section coefficients, target (α, n) cross-sections, target (α, n) product level branching data and sources decay data that are used during the calculations. Users can choose among 19 low-Z nuclides and up to 107 decay nuclides that could serve as α-sources. Further details of the methodology used in SOURCES 4C and the nuances of the data and its sources are available in Wilson et al.. For the current study, the low-Z and α source nuclide inventories were obtained from the Serpent calculations described previously. SOURCES 4C requires the user to supply these quantities in a certain format along with some calculation-specific parameters (such as number of α energy groups) and in specific units which the user must be mindful of. It was noted that using lower number of α energy groups ($<$ 10) is generally preferable and the code produces an error if the input file consists of a composition with numerous α sources and low-Z targets. Once the (α, n) and SF neutron emission rates calculations were performed, they were combined with the dataset containing results from the Serpent calculations.

4 Description of dataset

Combining results from calculations carried out in Serpent and SOURCES 4C, a dataset comprising of properties of irradiated salt at various combinations of BIC was compiled. Based on information presented in Table 2, the dataset contains a total of $75 \cdot 41 \cdot 101 = 310,575$ entries of different combinations of salt BIC and the associated quantities of interest like nuclide inventories, and nuclide specific gamma source strength (in photons/second), decay heat (in Watts), (α, n) and SF neutron emission rates. The structure of the final dataset is shown in Table 5.

Table 5: Description of dataset for irradiated CMSR salt.

BU	**IE**	**CT**	**a**$_1$	**a**$_M$	**m**$_1$	**m**$_M$	**SF**$_1$	**SF**$_M$	**GE**$_1$	**GE**$_M$	**DH**$_1$	**DH**$_M$	**(α, n)**
BU_1	IE_1	CT_1	a$_{11}$	a$_{1M}$	m$_{11}$	m$_{1M}$	SF$_{11}$	SF$_{1M}$	GE$_{11}$	GE$_{1M}$	DH$_{11}$	DH$_{1M}$	(α, n)$_1$
...													
BU_N	IE_N	CT_N	a$_{N1}$	a$_{NM}$	m$_{N1}$	m$_{NM}$	SF$_{N1}$	SF$_{NM}$	GE$_{N1}$	GE$_{NM}$	DH$_{N1}$	DH$_{NM}$	(α, n)$_N$

a_{ij}: ith isotopic number density in atoms/barn·cm for each one of the M nuclides,
m_{ij}: ith isotopic mass density for jth BIC combination in g/cm^3 for each one of the M nuclides,
SF_{ij}: SF rate of the ith isotope and jth BIC combination in SF/sec for each one of the M nuclides,
GE_{ij}: gamma emission rate of the ith isotope and jth BIC combination in photons/sec for each one of the M nuclides,
DH_{ij}: decay heat of the ith isotope and jth BIC combination in Watts for each one of the M nuclides,
(α, n)$_i$: (α, n) neutron emission rate for ith BIC combination.

The end goal for creating this dataset is for the safeguards community to analyse properties of the fuel salt and to allow for the implementation of machine learning (ML) algorithms trained on safeguards-relevant signatures. Many ML algorithms (such as neural networks) require large amounts of data to train therefore, datasets of large dimensions are often a necessity. Such models can be used to predict salt's BIC parameters and also explore the development and use of new NDA signatures that could be interesting for MSR fuel. Such research has previously focused on PWR-type SNF (Grape et al., Bachmann et al., Mishra et al., Mishra, Borella et al., Borella et al.) using a variety of datasets that are built for this purpose (Elter et al., Rossa and Borella). These techniques may prove useful

*Correspondence: vaibhav.mishra@physics.uu.se, zsolt.elter@seaborg.com

for development of safeguards verification strategies for irradiated molten fuel salts as guidelines for such fuel are at a lower level of maturity compared to those for conventional LWR SNF.

5 Results

The following sections describe the results from the analysis of depletion studies conducted for the CMSR to determine the primary contributors to emissions from the irradiated salt to highlight the prevalence of any major differences in the nature and magnitude of emissions from irradiated CMSR salt and the conventional PWR SNF. Wherever results are presented and comparisons are drawn, the SNF is assumed to have properties listed in Table 6. These BU and IE values for PWRs were selected on the basis that they have become more prevalent in the recent times where operators aim for longer fuel cycles and higher terminal BU.

Table 6: Properties of fuel types compared in the study.

Fuel Type	**BU** ($\frac{MWd}{kgU}$)	**IE (wt. % U^{235}) (unless otherwise specified)**	**CT**
CMSR	15.0	15.0	variable
PWR-UOX	50.0	5.0	variable
PWR-MOX	50.0	5.0 wt. % ★IPC	variable

★ IPC: Initial Plutonium Content.

5.1 Gamma emissions

The gamma emission calculations for the CMSR were performed using Serpent and the values for PWR-UOX and PWR-MOX were obtained from the specific dataset produced for PWRs Elter et al.. The top ten contributors to gamma emissions (in photons/sec) from the irradiated MSR salt at six different cooling times are listed in Table 7. From the results it is rather evident that for fuels cooled between 1–40 years, Cs^{137} (where Ba^{137m} is a daughter of Cs^{137}) remains as the primary contributor to the gamma emissions from the irradiated CMSR salt. Other nuclides from Table 3 are also seen to be among the top ten contributors in Table 7. Whether or not some of these nuclides can be used for verification of irradiated salts will depend on many factors such as whether they have emissions of a suitably high energy to make them detectable with detectors used by inspectors. In comparison with emissions from PWR (of both UOX and MOX type), the gamma emission levels (as a percentage of total gamma emissions) are shown in Figure 3. It should be noted that only nuclides from Table 3 are shown in Figure 3. One may see that the nature of the gamma emissions (between CMSR and PWR) are largely comparable (owing to use of the same fissile material, U^{235}). However, some subtle differences such as the proportion of emissions from Cs^{137} after 10 years of cooling are noticeable. For all three fuel types, nearly 100% emissions are from Cs^{137} past 10 years of CT (owing to its long half-life). At shorter CTs ($1 \leq CT \leq 10$ years), emissions from short-lived nuclides like Nb^{95}, Ru^{106} and Cs^{134} amounts to greater than those from Cs^{137} which is not observed in the case of CMSR where Cs^{137} is always the dominant contributor to gamma emissions of the salt over the entire CT range. These anolmalies may arise from the large difference in IE for fresh fuel in CMSR compared to that used in PWR-UOX or PWR-MOX.

Table 7: Top 10 contributors to gamma emissions for irradiated CMSR salt.

CT(y)	Rank 1	Rank 2	Rank 3	Rank 4	Rank 5	Rank 6	Rank 7	Rank 8	Rank 9	Rank 10
0	Np^{239}	U^{239}	La^{140}	I^{135}	Ce^{143}	Te^{132}	Xe^{135}	Xe^{133}	Nb^{95}	Zr^{95}
0.5	Nb^{95}	Ba^{137m}	Zr^{95}	Ce^{144}	Cs^{134}	Rh^{106}	Ru^{103}	Ce^{141}	Pr^{144}	Sb^{125}
1	Ba^{137m}	Ce^{144}	Cs^{134}	Nb^{95}	Rh^{106}	Zr^{95}	Sb^{125}	Pr^{144}	Eu^{154}	Eu^{155}
5	Ba^{137m}	Cs^{134}	Eu^{154}	Sb^{125}	Ce^{144}	Rh^{106}	Eu^{155}	Te^{125m}	Eu^{152}	Am^{241}
10	Ba^{137m}	Cs^{134}	Eu^{154}	Sb^{125}	Eu^{155}	Am^{241}	Eu^{152}	Te^{125m}	Pu^{239}	Pu^{238}
40	Ba^{137m}	Am^{241}	Eu^{154}	Eu^{152}	Pu^{239}	Pu^{238}	Pu^{240}	Y^{90}	Eu^{155}	Kr^{85}

5.2 Decay heat production

Jansson, Gauld and Ryman, Broadhead have previously looked into the importance of nuclides that play a key role in the production of decay heat from LWR fuel after discharge. Some of these nuclides

*Correspondence: vaibhav.mishra@physics.uu.se, zsolt.elter@seaborg.com

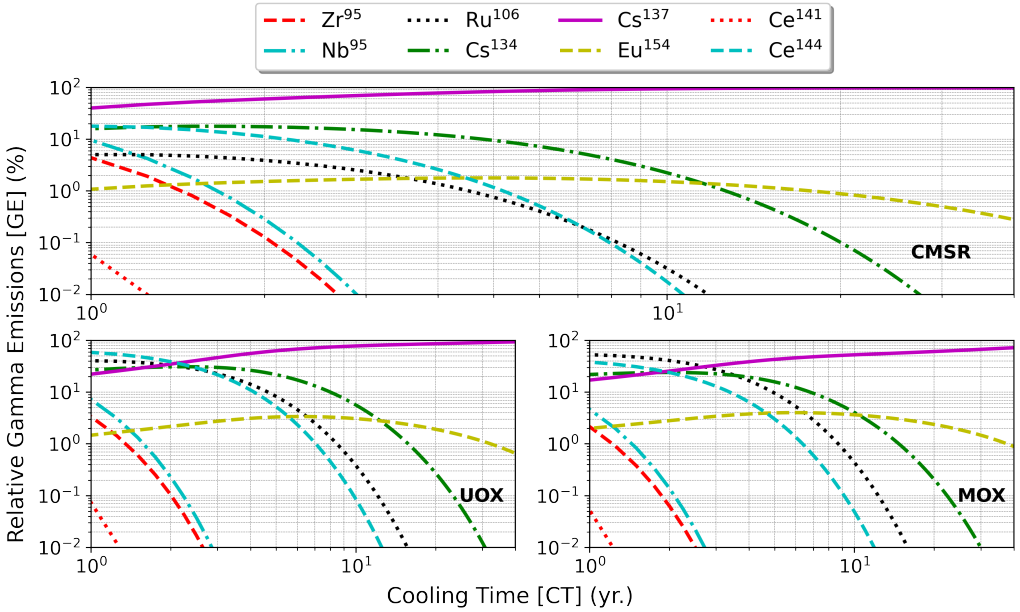


Figure 1: Comparison of percentage contributions from major gamma emitters in irradiated CMSR, PWR-UOX, and PWR-MOX fuels as a function of CT.

are also listed in Table 4 and the trends in nuclide importance given in Jansson, Gauld and Ryman are consistent with those observed in the present study for irradiated CMSR salt. As before, the decay heat calculations were performed using Serpent. The top ten contributors to decay heating in irradiated CMSR salt are listed in Table 8. One can say that for freshly discharged salt, the decay heat production is dominated by very short-lived nuclides such as U^{239} (half-life 23 minutes) and its decay product, Np^{239}. While for salt with $1 \leq CT \leq 10$ years, nuclides such as Y^{90}, Sr^{90}, Cs^{137}, Ba^{137m}, and Pr^{144} are major contributors. These trends are also readily apparent in Figure 2. Findings from Jansson peg the average contribution of fission products (and their daughter nuclides) at just above 60% and just about 40% for actinides over $10 \leq CT \leq 50$ years. While for the CMSR salt, this is split between fission products and actinides in the proportion 51:49. Overall, isotopes such as Y^{90} and Cs^{137} alone contribute to over 50% of the decay heating after $CT \geq 10$ years with contributions from actinides such as Pu^{238}, Pu^{239} and Am^{241} gradually becoming more important for $CT \geq 10$ years.

Table 8: Top 10 contributors to decay heat for irradiated CMSR salt.

CT(y)	Rank 1	Rank 2	Rank 3	Rank 4	Rank 5	Rank 6	Rank 7	Rank 8	Rank 9	Rank 10
0	U^{239}	Np^{239}	La^{140}	I^{135}	Zr^{95}	Nb^{95}	Mo^{99}	Ce^{143}	Xe^{135}	Y^{91}
0.5	Pr^{144}	Nb^{95}	Y^{90}	Rh^{106}	Ba^{137m}	Zr^{95}	Cs^{134}	Y^{91}	Ce^{144}	Cs^{137}
1	Pr^{144}	Y^{90}	Ba^{137m}	Rh^{106}	Cs^{134}	Ce^{144}	Cs^{137}	Sr^{90}	Nb^{95}	Zr^{95}
5	Y^{90}	Ba^{137m}	Cs^{137}	Sr^{90}	Cs^{134}	Pr^{144}	Pu^{239}	Rh^{106}	Pm^{147}	Kr^{85}
10	Y^{90}	Ba^{137m}	Cs^{137}	Sr^{90}	Pu^{239}	Am^{241}	Pu^{238}	Kr^{85}	Cs^{134}	Eu^{154}
40	Y^{90}	Ba^{137m}	Cs^{137}	Sr^{90}	Am^{241}	Pu^{239}	Pu^{238}	Pu^{240}	Kr^{85}	U^{234}

5.3 Neutron emissions

Neutron emissions from spent fuel can be attributed to many sources. However, SF in heavy elements ($Z \geq 90$) and neutrons originating from (α, n) reactions in the fuel matrix are among the two most important sources Ensslin. As the name suggests, SF refers to the non-induced fission of a heavy nucleus and just like induced fission, SF produces fission fragments and additional neutrons that can be used to assay the fuel material. There are numerous heavy nuclides such as transuranics which are prone to SF and therefore spent fuel will show much higher rates of SF than fresh fuel (which only contains nuclides such as U^{234}, U^{235}, and U^{238}). Some unstable nuclei can also undergo α-decay which can further interact with low-Z elements in the fuel matrix such as oxygen (in LWR fuel) or fluorine (in CMSR salt) and produce additional neutrons by means of an (α, n) reaction. The ratio between neutron emissions from (α, n) and SF is termed as the 'α-ratio' Langner et al. and is typically unique

*Correspondence: vaibhav.mishra@physics.uu.se, zsolt.elter@seaborg.com

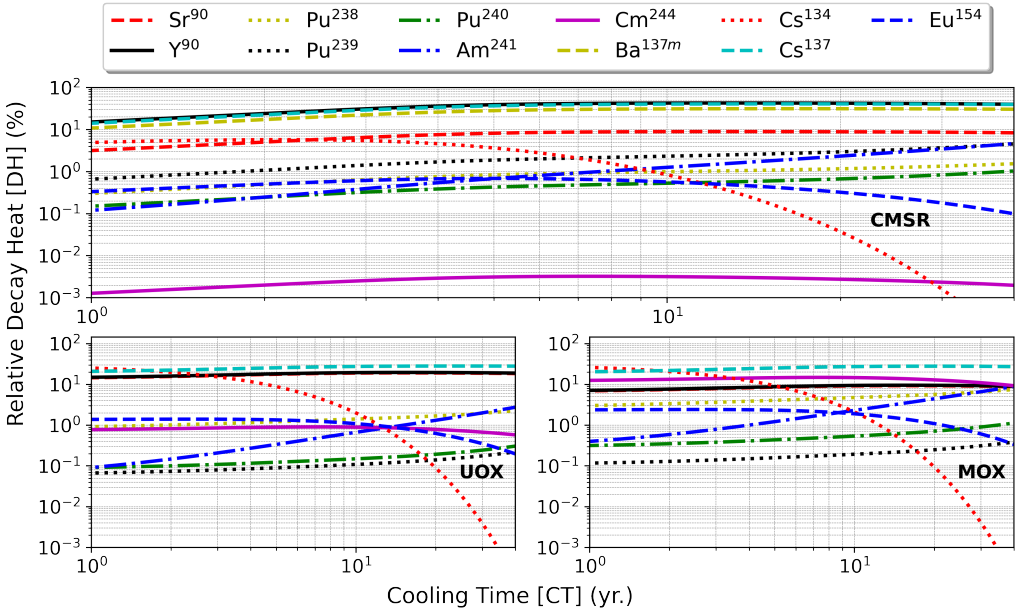


Figure 2: Comparison of percentage contributions from major decay heating nuclides in irradiated CMSR, PWR-UOX, and PWR-MOX fuels as a function of CT.

for a reactor and fuel type. The results in this section will therefore be presented in terms of the α-ratio for all three fuel types.

5.3.1 Spontaneous fission neutron emission

Previous work Preston et al., Richard et al. have looked into the magnitudes and spectra of SF neutrons originating from spent fuel produced by conventional LWRs and MYRRHA Abderrahim et al. fuel respectively. The present study will look into the nature of SF neutron emissions for irradiated CMSR salt. In a manner similar to previous sections, the calculations were performed SOURCES 4C. As done in the previous sections, the top ten contributors to SF neutrons are given in Table 9. The SF rates were computed with both, Serpent and SOURCES 4C and were found to be in good agreement with each other. It is evident from Table 9 that for irradiated salt with ($1 \leq CT \leq 10$ years), nearly all emissions are dominated by curium and plutonium isotopes such as Cm^{242}, Pu^{240}, Cm^{244} et cetera and to a lesser extent by isotopes of uranium such as U^{234} and U^{236}. Further nuances of the differences in the nature of SF neutron emissions between LWR fuels and CMSR salt are shown in Figure 2. It is seen that while the trends look similar between PWR-UOX and PWR-MOX fuels with Cm^{244} being the dominant contributor to nearly 100% SF neutrons, for the CMSR, Pu^{240} appears to be the primary contributor for $5 \leq CT \leq 40$ years. For PWR-UOX and PWR-MOX type fuels, contributions from Cm^{242} never exceed 15% over the entire CT range and show a marked dip after CT ≥ 1 years. A similar dip (of greater magnitude) is also observed in the case of CMSR.

5.3.2 (α, n) emissions

The calculation code SOURCES 4C was used for computing the rates of emissions of (α, n) neutrons from the irradiated fuel. It should be noted that the α-ratio (ratio between (α, n) and SF neutron emissions) were computed for all three fuel types (PWR-UOX, PWR-MOX and CMSR) at three different BU values using SOURCES 4C and the results are shown in Figure 4. It can be seen that for both, PWR-UOX and PWR-MOX fuel types, the α-ratio is always below unity for all BU values and remains so throughout the entire range of CT. This indicates that for conventional LWR fuel, SF is the dominant source of neutrons when compared to those from (α, n) reactions. Whereas in the case of irradiated CMSR salt, the α-ratio is noticeably over unity (closer to 200 in all cases) and rises further with increasing CT. This is quite a striking dissimilarity between the fuel types as it indicates that the neutron yield from (α, n) is significantly higher than that from SF from the irradiated CMSR salt. This can however be explained by the fact that CMSR fuel matrix has eight low-Z atoms with non-zero cross-sections for (α, n) reactions for every heavy nuclide that might undergo α-decay compared to

*Correspondence: vaibhav.mishra@physics.uu.se, zsolt.elter@seaborg.com

Table 9: Top 10 contributors to the emission of SF neutrons for irradiated CMSR salt.

CT(y)	Rank 1	Rank 2	Rank 3	Rank 4	Rank 5	Rank 6	Rank 7	Rank 8	Rank 9	Rank 10
0	Cm^{242}	Pu^{240}	Cm^{244}	Pu^{238}	U^{238}	Pu^{242}	Pu^{239}	U^{236}	Am^{241}	U^{234}
0.5	Cm^{242}	Pu^{240}	Cm^{244}	Pu^{238}	U^{238}	Pu^{242}	Pu^{239}	U^{236}	Am^{241}	U^{234}
1	Cm^{242}	Pu^{240}	Cm^{244}	Pu^{238}	U^{238}	Pu^{242}	Pu^{239}	U^{236}	Am^{241}	U^{234}
5	Pu^{240}	Cm^{244}	Pu^{238}	U^{238}	Cm^{242}	Pu^{242}	Pu^{239}	Am^{241}	U^{236}	U^{234}
10	Pu^{240}	Cm^{244}	Pu^{238}	U^{238}	Pu^{242}	Cm^{242}	Pu^{239}	Am^{241}	U^{236}	U^{234}
40	Pu^{240}	Pu^{238}	Cm^{244}	U^{238}	Pu^{242}	Cm^{242}	Pu^{239}	Am^{241}	U^{236}	U^{234}

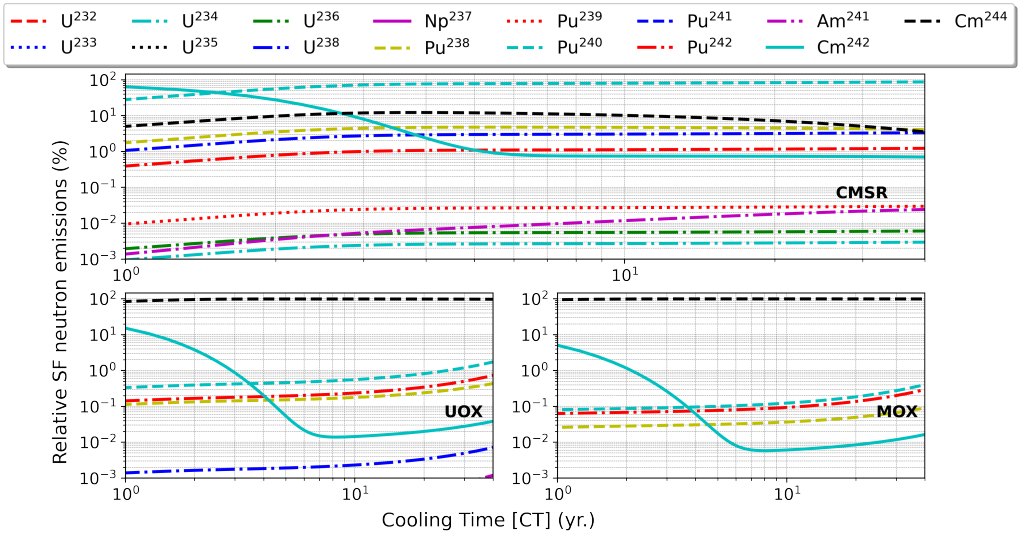
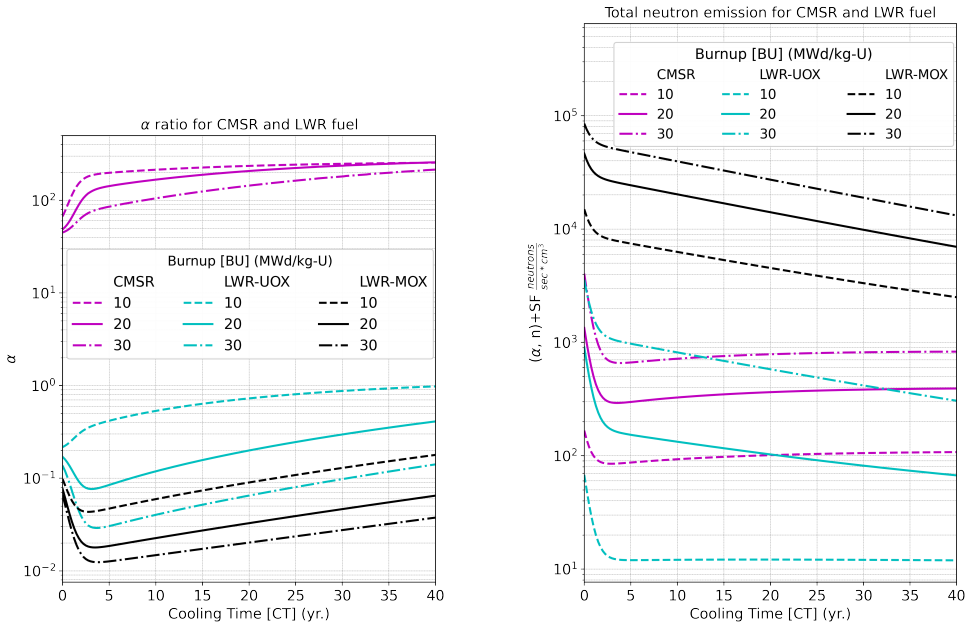


Figure 3: Comparison of percentage contributions from major emitters of SF neutrons in irradiated CMSR, PWR-UOX, and PWR-MOX fuels as a function of CT.



(a) Comparison of α-ratio for CMSR, PWR-UOX, and PWR-MOX fuel types for different BU values as a function of CT.

(b) Comparison of total neutron emissions ((α, n) and SF) in irradiated CMSR, PWR-UOX, and PWR-MOX fuels as a function of CT for different BU values.

Figure 4: Comparison of α-ratio and overall neutron emission rates between fuel types.

just two (O-atoms in PWR fuel matrix) in the case of PWR fuels. For sake of validation, selected values of the α-ratios for PWR-UOX and PWR-MOX were compared to those reported in Preston

*Correspondence: vaibhav.mishra@physics.uu.se, zsolt.elter@seaborg.com

et al.[6] and they were found to be in good agreement. The primary sources for the (α, n) neutrons were found to be those resulting from α-decay of Cm^{242} on F^{19} and Na^{23} while in the case of PWR fuel, from α-decay of Cm^{242} on O^{17} and O^{19}. The absolute magnitude of total neutron emissions (from (α, n) and SF) for the three fuel types is shown in Figure 4. It can be seen from the results that the total emissions for a typical MOX fuel are significantly higher than both, UOX and CMSR fuel for all three BU values. It can also be noted that while total neutron emissions are rather strongly dependent on both BU and CT for LWR-UOX and LWR-MOX fuel types (as was also noted in Preston et al.), the dependence of neutron emission levels for the irradiated CMSR salt on CT is rather weak compared to the other two fuel types. Interestingly, the total neutron emission rates increase slightly with increasing CT which is in stark contrast with UOX and MOX fuel types. This may be attributed to the buildup of nuclides like Am^{241} and their tendency to undergo (α, n) with increasing CT.

6 Conclusions and discussion

The present study has aimed to highlight some of the key characteristics of gamma and neutron emissions and decay heat production in spent fuel from a selected MSR concept. The findings have been presented in a comparative fashion to facilitate a one-to-one comparison of the nature of radiation emissions from the irradiated CMSR salt against those from conventional PWR SNF. With the help of a large database of safeguards-relevant quantities, the study has provided a preliminary qualitative and quantitative evaluation that can be useful to regulators and inspectors alike to devise safeguards verification strategies for such SNF. These findings include information on primary contributors to gamma and neutron emissions which could be useful in verification using conventional DA and NDA methods. Despite existence of subtle differences in the characteristics of gamma emissions, it appears that the overall nature of emissions is relatively similar to conventional PWR SNF and existing safeguards approaches should remain applicable with little adaptation. Differences in nature of neutron emissions between CMSR and PWR SNF e.g. relatively high levels of (α, n) compared to SF, may have safeguards implications. Some other findings from the assessment relating to levels of production of decay heat by the salt are also expected to help facilitate storage and transport of the irradiated salts in the future.

Acknowledgments

The authors thank the Swedish Radiation Safety Authority or Strålsäkerhetsmyndigheten (SSM) for the financial support under the contract SSM2017-5980 that made this work possible. We are also grateful to our industry collaborators from Seaborg Technologies for sharing models, specifications and their feedback that was crucial for this work to come to fruition.

Notes

[1]The CMSR can remove specific radionuclides via an OGS however, it was not included in the Serpent model.

[2]Nuclear loss calculations performed herein due to transformation to another element or isotope, aka BU for SNF.

[3]Fresh CMSR salt is modeled to include U^{234} which is expected due to the use of HA-LEU fuel. Modeled fresh LWR-UOX does not account for the presence U^{234}.

[4]200 batches with 2000 neutrons per batch and first 50 generations skipped to ensure fission source convergence.

[5]BU - For BU < 1.0 $\frac{MWd}{KgU}$ – steps increasing by a factor of 2.5 For BU ≥ 1.0 $\frac{MWd}{KgU}$ – steps of 0.5 $\frac{MWd}{KgU}$. IE - 41 values in steps of 0.25. CT - 101 steps and step size range is 0.25 years up to 10 years and in steps of 0.5 years thereon.

[6]Present work uses SOURCES 4C to compute (α, n) and SF rates while Preston et al. uses (α, n) yields from Simakov and Van den Berg.

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*CORRESPONDENCE: VAIBHAV.MISHRA@PHYSICS.UU.SE, ZSOLT.ELTER@SEABORG.COM