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Criticality safety calculations of storage canisters

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This report concerns a study which was conducted for SKB. The conclusions and viewpoints presented in the report are those of the author and do not necessarily coincide with those of the client.

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1 Introduction

In the planned Swedish repository for deep disposal of spent nuclear fuel the fuel assemblies will be stored in storage canisters made of cast iron and copper.

To assure safe storage of the fuel the requirement is that the normal criticality safety criteria have to be met. The effective neutron multiplication factor must not exceed 0.95 in the most reactive conditions including different kinds of uncertainties.

In this report it is shown that the criteria could be met if credit for the reactivity decrease due to the burn up of the fuel is taken into account. This study is a continuation of the work presented in reference 1.

2 Methods

All calculations were performed using Scale 4.4 (reference 2). Depletion calculations were performed using the Scale SAS2H control sequence and the criticality calculations were performed using Scale CSAS25 (Keno V.a) sequence. The calculations were performed using the Scale 44 – group (ENDF/B-V) library.

The SAS2 control module was originally developed for SCALE to provide a sequence that generated radiation source terms for spent fuel and subsequently utilized these sources within a one-dimensional (1-D) radial shielding analysis of a shipping cask. One of the principal uses of SAS2 over its history has been fuel depletion analyses to obtain radiation and heat generation sources and spent fuel isotopics to be used in subsequent analyses. The new, significantly enhanced version of the SAS2 control module is denoted as SAS2H. For each time-dependent fuel composition, SAS2H performs 1-D neutron transport analyses (via XSDRNPM) of the reactor fuel assembly using a two-part procedure with two separate lattice-cell models. The first model is a unit fuel-pin cell from which cell-weighted cross-sections are obtained. The second model represents a larger unit cell (e.g. an assembly) within an infinite lattice. The larger unit-cell zones can be structured for different types of BWR or PWR assemblies containing water holes, burnable poison rods, gadolinium fuel rods, etc. The fuel neutron flux spectrum obtained from the second (large) unit-cell model is used to determine the appropriate nuclide cross-sections for the specified burnup-dependent fuel composition. The cross-sections derived from a transport analysis at each time step are used in a point-depletion computation (via ORIGEN-S) that produces the burnup-dependent fuel composition to be used in the next spectrum calculation. This sequence is repeated over the operating history of the reactor.

The Criticality Safety Analysis Sequences (CSAS) were developed within the SCALE code system to provide automated, problem-dependent, cross-section processing followed by calculation of the neutron multiplication factor for the system being modeled. These control sequences activate the cross-section processing codes BONAMI and NITAWL-II to provide resonance-corrected cross-sections. KENO V.a uses the processed cross-sections and calculates the k-effective of three-dimensional system models. The geometric modeling capabilities available in KENO V.a coupled with the automated cross-section processing within the control sequences allow complex, three-dimensional systems to be easily analyzed. In this study the sequence CSAS25 was used.

Input to the CSAS25 sequence is geometric data as well as isotopic data. For fresh fuel these data are readily available. For irradiated fuel the isotopic composition in the fuel was calculated separately using the sequence SAS2H/Origen-S in Scale 4.4. Based on initial fuel geometry and composition the code calculates the isotopic compositions resulting from specified irradiation history. The isotopics were extracted for different burnups and enrichments and used as input in the CSAS25 model to calculate the $k_{\rm eff}$ for different burnups and enrichments. These data were then used to construct the limit curves for $k_{\rm eff}$ =0.95 for the BWR and PWR-canister for different sets of actinides and fission products. Finally the limit curves were compared with the inventory in CLAB.

3 Criticality safety criteria

The criticality safety criteria are based on the US NRC regulatory requirements for transportation and storage of spent fuel. The US NRC positions are found in several regulatory guides:

Regulatory guide 3.58 – Criticality Safety Criteria for the Handling, Storing and Transporting LWR Fuel at Fuels and Materials Facilities.

Regulatory guide 1.13 – Proposed revision 2 to Regulatory Guide 1.13 Spent Fuel Storage Facility.

The basic criticality criteria is that the effective neutron multiplication factor should not exceed 0.95 including uncertainties and the nuclear safety analysis should include considerations of all credible normal and abnormal operating occurrences. Credit for fuel burnup may be taken.

4 Canister design and fuel types

4.1 Canister design

A canister consists of an insert of cast iron with a diameter of 946 mm with a 50 mm thick outer shell of copper. The outside diameter of a canister is 1,050 mm. During the casting process channels for the fuel assembly's storage positions are formed by square-formed tubes of steel. The wall thickness of these tubes is 10 mm. In the BWR-insert 12 storage positions are formed with the inner measures of $160 \text{ mm} \times 160 \text{ mm}$. In the PWR-insert four storage locations are formed with the inner measures of $230 \text{ mm} \times 230 \text{ mm}$.

In Table 4-1 below the main parameters of the canisters are presented.

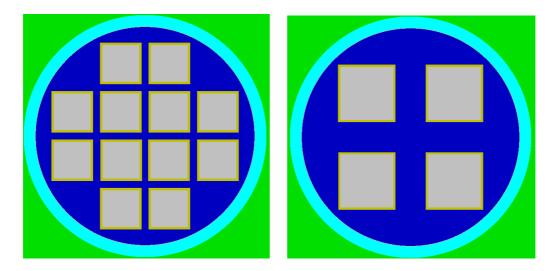


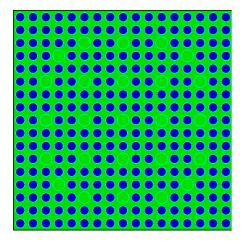
Figure 1. Cross-section of the BWR- and PWR-canister.

Table 4-1. Main parameters for the canisters.

Parameter	BWR	PWR
No positions	12	4
Pitch between channels (mm)	210	380
Channel size, inner (mm)	160	230
Channel tube wall thickness (mm)	10	10
Insert diameter (mm)	946	946
Canister diameter (mm)	1050	1050
Insert material	Cast Iron	Cast Iron
Insert material density (kg/dm³)	7.85	7.85
Steel density (kg/dm³)	7.85	7.85
Tube material	Steel	Steel
Shell material	Cu	Cu
Copper density (kg/dm³)	8.93	8.93
Length of channel (mm)	4470	4470
Length of canister (incl. Cu shell)	4833	4833

4.2 Fuel types

The criticality calculations were based on the fuel assembly types that were used as reference fuel in the safety analysis of CLAB. For BWR the fuel type is Svea 64 and for PWR F17×17. The cross-sections of the fuel are shown in Figure 2 and characteristics in Table 4-2.



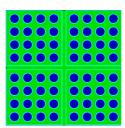


Figure 2. Cross-section of PWR- and BWR-fuel assemblies.

Table 4-2. Main parameters for the fuel assemblies.

Parameter	BWR	PWR
Fuel type	Svea64	17×17
No of fuel rods	64	264
No of guide thimbles	0	25
Max enrichment (% U235)	3.6	4.2
UO2 density (g/cm³)	10.47	10.45
Temperature (°C)	20	20
Fuel rod pitch (mm)	15.8	12.6
Fuel rod diameter (mm)	12.25	9.5
Cladding thickness (mm)	0.8	0.57
Pellet diameter (mm)	10.44	0.819
Channel wall thickness (mm)	1.1	-
Central cross width (mm)	4	-
Central cross wall thickness (mm)	0.8	_
Cladding material	Zr 2	Zr 2
Active fuel length (mm)	3680	3658

4.3 Irradiation history of the fuel assemblies

In order to calculate the isotopic composition of the fuel at different burnups the fuel had to be subjected to different burnup histories. The main parameters for the depletion calculation are shown in Table 4-3.

Table 4-3. Main parameters for the depletion calculation.

Parameter	BWR	PWR	
Assembly power (MW)	4	15	
Avg. fuel temperature (°C)	567	621	
Channel temperature (°C)	347	-	
Coolant pressure (bar)	70	155	
Coolant temperature (°C)	286	320	
Coolant density (kg/dm³)	0.388	0.676	
Cycle length (days)	345	312	
Shutdown length (days)	20	53	
Decay time (yrs)	10	10	

5 Analysis with fresh fuel

5.1 Underground storage position

Models for the reference fuel assemblies and final storage canisters were developed to investigate the criticality conditions for fresh fuel. The canister data from Table 4-1 and fuel data from Table 4-2 were used.

In the nominal case it is assumed that unirradiated fuel is stored in canisters. Each fuel assembly is located in the center of the storage location. The fuel and canister measures are nominal. The canisters are assumed to be stored in the underground storage surrounded by 60 cm bentonite in an infinite lattice. The bentonite is saturated with water.

In nominal conditions the canister is leak-tight and the atmosphere in the storage positions is dry nitrogen. In this case with no water present the effective neutron multiplication factor is less than 0.5 and the system is indeed subcritical.

If it is assumed that the canister is leaking and that the canister storage positions and the fuel assemblies are water filled the reactivity will increase. With all storage locations in the canister filled with water the following results are found:

BWR: k_{eff} =0.9050±0.0012

PWR: $k_{eff}=1.0550\pm0.0012$

5.2 Dry storage

During handling of the loaded canisters at the encapsulation plant they will be stored in a dry storage prior to transport to the final repository.

In the dry storage the canisters are stored in air.

Maximum reactivity in the dry storage will occur when the canisters are filled with water and the space between the canisters still consists of air.

In this position the following results are found:

BWR: $k_{eff}=0.9242\pm0.0012$

PWR: k_{eff}=1.0868±0.0012

In all these cases the fuel assemblies are located in the centre of the storage channels, which is an ideal situation. From reactivity standpoint the worst case is when the assemblies are located towards the centre of the canister. See Figure 3.

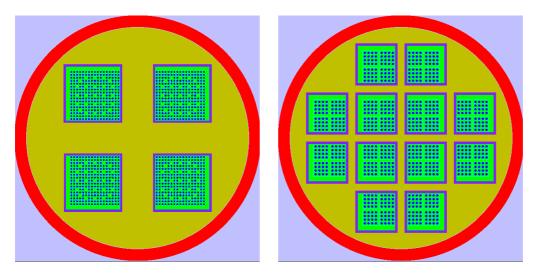


Figure 3. Cross-section of the BWR- and PWR-canister with the assemblies in eccentric placing.

In this geometry, waterfilled canisters that are located in the dry storage, criticality calculations give the following results:

BWR: k_{eff} =0.9319±0.0012

PWR: k_{eff} =1.0903±0.0012

These values represent the situation with the highest reactivity. Margin is also needed to cover various uncertainties such as manufacturing tolerances and calculational uncertainties. The uncertainties are analysed in section 7.3 later in this report.

At this point it is concluded that the reactivity criteria could not be met with the conservative assumption that the fuel is fresh.

A more realistic assumptions would be to take credit for the burnup of the fuel, which decreases the reactivity. In the following section it is investigated if the burnup of the fuel will give enough decrease of the reactivity to meet the reactivity criteria.

6 Burnup credit - selection of nuclides

In the irradiation process around 1,300 isotopes are created in the irradiated fuel.

All isotopes can not be represented in the models and it is needed to establish a limited set to use in burnup credit.

To select which isotopes to credit the following requirements could be established as criteria:

1. The isotopes should contribute to the reactivity decrease

The importance of different isotopes has been assessed in several studies. The reactivity worth of different isotopes vary with fuel design, initial enrichment, operational history and cooling time. The important nuclides seem, however to remain the same. In Appendix 3 a review of the international work and state of the art was done. Different sets of and fission products were examined in order to establish a set of nuclides that could be used in burnup credit.

2. Knowledge of nuclear data of the isotopes

Nuclear data such as neutron cross-sections and half-lifes have to be well known in order to be able to predict the isotopic contents in the fuel.

3. Knowledge of their chemical form, physical form and characteristics, solubility and volatility

Subcriticality in the final repository needs to be verified for very long time periods. During this time it has to be certain that the isotopes are stable in the fuel.

4. The calculated isotopic content in the spent fuel should be verified

The calculation of isotopic composition in spent nuclear fuel should be verified by comparison with experimental data if the nuclides are used in burnup credit. This provides a robust basis for canister design and final storage.

If these positions the following nuclides could presently be used for burnup credit in final storage canisters. The basis for the selection is found in Appendix 3.

U234, U235, U238, Pu 239, Pu 240, Pu 241, Pu 242, Am 241

Criticality calculations are performed with credit only for the above limited number of isotopes to assess if burnup credit still is sufficient to control reactivity. If not, efforts should focus on validating additional actinides and fission products such as:

Am243, Np237 Nd143, Nd145, Sm147, Sm149, Sm150, Sm151,Sm152 Eu151, Eu153, Gd155 Te99, Rh103

It is recommended that the development in these areas is followed up in order to include more nuclides in burnup credit for the final storage canisters.

7 Calculations

7.1 Calculation of isotopics

The isotopic composition was calculated for BWR – Svea 64 and PWR-F17×17 fuel using the different irradiation histories. The irradiation was simulated in reactor conditions. Specific power, cycle history and reactor condition from Table 4-3 were used.

SAS2H was used to calculate the isotopic concentrations and the program Opus was used to extract the isotopic number densities from the SAS2H output files.

For BWR one cycle was assumed to be 345 days followed by a 20 days outage. This gives a burnup increase of 7 MWd/kgU per cycle, which is a realistic value.

PWR irradiation period of 320 days was used followed by 53 days outage. This gives a burnup increase of 10 MWd/kgU per cycle, which is a realistic value for PWR.

The irradiation histories for different burnups are shown in Table 7-1. The per-assembly power during operation was 4 MW for a BWR-assembly and 15 MW for a PWR-assembly.

In the BWR-case an average relative moderator density of 0.388 was used, which corresponds to a conservative estimate of the exit void fraction in a hot assembly in a BWR-core. In the PWR-case the moderator density was set to correspond to the core exit temperature. The temperatures of fuel and materials during irradiation are based on generic data.

For both BWR and PWR the enrichments 4.2%, 3.6% and 3.0% U235 were analysed.

The fuel was decayed 10 years after the last cycle before the isotopics were extracted for the criticality calculations.

Table 7-1. Irradiation histories.

No cycles	Burnup (MWd/kgU)		
	BWR	PWR	
1	7	10	
2	14	20	
3	22	30	
4	29	40	
5	37	50	
6	45	-	
7	53	-	

Tables of isotopic concentrations were extracted from the depletion calculations for all enrichments and burnup for both PWR and BWR according to Table 7-1. The following nuclides were extracted:

Set 1: U234, U235, U238, Pu239, Pu240, Pu241, Pu242, Am241

Set 2: Set 1+ Am243, Np237, Nd143, Nd145, Sm147, Sm149, Sm150, Sm151,Sm152, Eu151, Eu153, Gd155

Set 3: Set 2+ Tc99, Rh103

The isotopic concentrations were formatted so they could be used in the canister model from section 5.2.

7.2 Criticality calculations

The isotopics were used in the canister model from section 5.2. A canister model for each enrichment and burnup was established. Criticality calculations were done with CSAS25 sequence.

The resulting k_{eff} for a canister for PWR fuel is presented in Appendix 1 for each enrichment and burnup and the different sets of nuclides. The corresponding diagrams for BWR fuel are shown in Appendix 2.

From these curves the burnup for different enrichments for the k_{eff} that is required for BWR fuel and for PWR fuel could be picked out.

These burnups should correspond to k_{eff} =0.95 including uncertainties and will be the minimum required burnup that a fuel assembly with a certain initial enrichment should have to assure sufficient margin to criticality.

To get these values assessment of the uncertainties is needed.

7.3 Uncertainties

The results are based on the most reactive location of the canisters during handling and the most reactive location of the fuel assemblies in the canister.

For other parameters where the worst case is not calculated the uncertainties need to be evaluated:

Fuel types and fuel data

In the study the reference fuel types from the CLAB safety analysis have been used. These cover the fuel types in the Swedish program. If it is shown that another fuel type has non-conservative depletion behavior, this could be handled by using a penalty on the initial enrichment for the specific fuel type. No uncertainly factor to cover this is used in this study.

Operation history

The operation history for the fuel assemblies is modeled based on realistic power histories for BWR- and PWR-reactors. In reality the power history for an assembly could be more complex. In reference 3 it is indicated that the effect of different operating histories could be $0.002 \Delta k$ at 5 years decay time.

Declared burnup

The declared burnup is based on the plant heat balance, which normally predicts the thermal power within 2%. Additional uncertainty when going from global reactor power to assembly power is estimated to be 1%, giving a total of 3%. These values are estimations based on experiences from the power plants.

With the diagrams in Appendices 1 and 2 it can be can calculated that a change of 1 MWd/kg burnup corresponds to an average of $0.0075~\Delta k$ for both PWR-fuel and $0.0079~\Delta k$ BWR-fuel for set 3. This gives an uncertainty of $0.009~\Delta k$ for PWR at 40~MWd/kgU and 0.005~for BWR at 22~MWd/kg.

Power density

The power density during irradiation is different for different plants. At the same plant the power density in an assembly vary during a cycle and between cycles. A lower power density seems to produce a higher k-values in reference 3. In reference 4 the opposite trend is observed. This study is performed with relatively low power densities (23 MW/tU for BWR and 33 MW/tU for PWR). To account for possible periods of operation at different power levels an uncertainty of 0.001 Δk is used for both PWR and BWR.

Axial void- and temperature distribution

Due to the high void content in the top of BWR-cores more Pu239 will be produced in the top of the core than in average. This will lead to non-conservatism if the average burnup is used to reflect the assembly k_{eff} . In the isotopic calculations a density content corresponding to the exit void in a hot assembly was used. This means the curves for BWR in Appendix 2 includes the axial effect caused by the void distribution.

For PWR the temperature distribution will give a similar effect, but smaller. This is covered by using the density corresponding to the exit core temperature.

The axial distribution of isotopics will thus not require any additional uncertainty.

Axial burnup distribution

According to reference 4 an end effect will occur due to the fact that burnup at the end zones of the fuel assembly is very low. This will give a penalty on the reactivity if the average burnup is used to reflect the assembly $k_{\rm eff}$. Therefore an uncertainty factor is suggested: 1% for burnups between 10–20 MWd/kg and 2% for burnups >20 MWd/kg. This is valid for PWR.

The BWR-assemblies normally have low enriched end zones, which will reduce this effect. This is not quantified and the same uncertainty as for PWR is used.

Control rods

During operation control rods are normally not inserted in the core. The effect of inserted control rods has therefore not been evaluated.

Horizontal burnup distribution

In the assembly a horizontal gradient of the burnup could be generated if the assembly is located in an area with a power gradient. This means that one side of the assembly could have lower burnup than the average, which in some cases could give a reactivity increase in the canister. If it is assumed that that the burnup could vary 5% from the average at one side of the assembly the k-value would increase 0.009 Δk for BWR and 0.015 Δk for PWR.

Calculational uncertainty

In reference 4 a number of critical experiments (both UO2 and MOX) were evaluated and reviewed. It was concluded that a 2% uncertainty factor will cover both bias and uncertainty in the Scale calculational methods. This factor could be used for systems with U and Pu.

The Keno result is always associated with a statistical uncertainty, normally 0.0012 Δk . To cover this on the 3 σ -level an uncertainty 0.004 Δk is added.

Manufacturing tolerances

When manufacturing the cast iron insert the measures on the channel width and the centre to centre distance could vary.

The effect of these variations was checked. It shows that a small (5 mm) change in channel size will not significantly change the reactivity. The same is valid for the c-c distance.

However, if eccentric placing of the fuel assemblies and a channel increase or a decrease in the c-c distance is assumed, the reactivity will change.

For a 5 mm change the following increases were calculated:

PWR $0.007 \Delta k_{eff}$

BWR $0.011 \Delta k_{eff}$

Isotopic prediction

There are several sets of radiochemical analyses of irradiated fuel samples that could be used to verify the calculations. Measured data have been calculated with the sequence SAS2H/Origen-S references 5, 6 and 7.

The comparisons, (calculated/measured-1)×100, are shown in Table 7-2.

Table 7-2. Comparison between calculated and measured data.

Nuclide	e ORNL SAS2H 44GROUPNDF5	
	BWR Note 1	PWR Note 2
U234	-0.2	3.3
U235	-2.0	-0.9
U236	-1.2	-1.1
U238	-0.1	-0.3
Pu238	-7	-3.1
PU239	-2.1	-1.3
PU240	-0.9	-0.2
PU241	-4.5	-1.1
PU242	0.5	1.7
Am241	4.1	-11
Am243		-4.3
Np237	-1.1	6.4
Nd143	0.4	0.4
Nd145	0.4	-0.3
Pm147/Sm147		-2.8
Sm149		-35.9
Sm150		-1.5
Sm151/Eu151		28.1
Eu152		20.2
Eu153		5
Eu155/Gd155	-42.6	-24.4
Rh103		?
Tc99	12.1	16.3

Note 1: Assemblies from Cooper, Gundremmingen and JPDR (reference 5)

Note 2: Assemblies from Calvert Cliff, HB Robinson, Obrigheim, Trino and Turkey Point (reference 6 and 7)

The question is how these differences in concentrations will affect the reactivity in the canister. To assess this the calculated isotopics were replaced by the corresponding measured values according to the table above. The calculations gave the following results:

BWR-canister 4.2% 22 MWd/tU

PWR-canister 4.2% 40 MWd/tU

Isotopics set 1 k_{eff} (calc. isotopics)=0.8909

K_{eff} (measured iso)=0.8935

Isotopics set 3 k_{eff} (calc. isotopics)=0.8063

K_{eff} (measured iso)=0.8058

It can be seen that replacing the calculated isotopics with measured the reactivity will not increase the reactivity significantly.

Long term reactivity change

Preliminary calculations of the long term change of reactivity was done in reference 8. The results are shown in Diagram 1. It can be seen that the reactivity decreases during the first 100 years, which mainly is due to the decay of fissile Pu241 with a half-life of 14.4 years. For longer times the reactivity will increase due to the decay of Pu239 to U235 and the ingrowth of daughter products.

Compared to the reactivity level at 10 years decay time a small increase in $k_{\rm eff}$ can be seen for burnup 20 MWd/kgU at 10^7 years. For burnup 40 MWd/kgU the reactivity is always lower than the 10-year value. In this study no additional uncertainty is used to account for long term reactivity change.

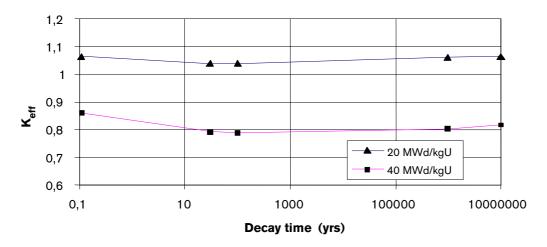


Diagram 1. K_{eff} for Svea64-type in water as a fuction of time.

Summary of the uncertainties

0 0.002
0.002
0.008
0.001
0
0.02
0
9 0015
0
0.007
0.02
0.004
0
2 0.078

In order to safely store the fuel in the canisters the k-value has to be reduced to less than 0.95 in both cases including uncertainties. The total uncertainties are 0.062 for BWR and 0.078 for PWR. This means that the calculated $k_{\rm eff}$ without uncertainties should be <0.888 for BWR and <0.872 for PWR.

7.4 Evaluation of the limit curve

If the burnups for each enrichment and set of isotopics are picked out from the curves in Appendices 1 and 2, the limit curves for k_{eff} values 0.95 for both BWR and PWR including uncertainties could be constructed. In Tables 7-3 and 7-4 the values are shown.

Table 7-3. PWR-Burnups (MWd/kgU) giving $k_{\mbox{\tiny eff}}\!\!=\!\!0.95$ for different enrichments including uncertainties.

Enrichment (%U235)	Isotopic	:s	
	Set 1	Set 2	Set 3
1.8	0.0	0.0	0.0
3.0	25.6	17.5	16.6
3.6	35.3	24.5	23.0
4.2	43.6	31.0	29.6

Table 7-4. BWR-Burnups (MWd/kgU) giving $k_{\mbox{\tiny eff}}\!\!=\!\!0.95$ for different enrichments including uncertainties.

Enrichment (%U235)	Isotopio	s	
	Set 1	Set 2	Set 3
2.8	0.0	0.0	0.0
3.0	4.0	2.2	2.1
3.6	12.4	8.1	7.4
4.2	20.0	14.7	13.8

8 Conclusions

Based on the information in Tables 7-3 and 7-4 limit curves could be constructed. A limit curve combines all the points of initial enrichments and burnups that will produce k_{eff} =0.95 in the canister including uncertainties. The limit curves are shown in Diagrams 2 and 3 for PWR and BWR respectively, where also fuel assemblies stored in CLAB 1998-12-31 are shown.

Limit curves for PWR 4,50 Initial enrichment (%U235) 4,00 3,50 3,00 2,50 2,00 X Set1 1,50 - Set 2 1,00 Set 3 0,50 0,00 0,00 10,00 20,00 30,00 40,00 50,00 60,00 Burnup (MWd/kgU)

Diagram 2. Limit curves for PWR compared to the CLAB-inventory.

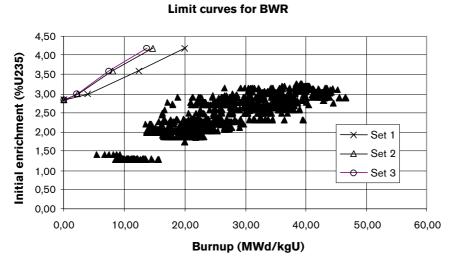


Diagram 3. Limit curves for BWR compared to the CLAB-inventory.

In the diagrams all fuel assemblies (combinations of initial enrichment and burnup) that appear on the right side of the limit curves will result in a $k_{\rm eff}$ <0.95. It can be seen that all BWR-assemblies and all PWR-assemblies stored in CLAB at the end of 1998 could be accepted for storage in canisters for final storage using burnup credit for actinides only (set 1). If additional actinides and selected fission products are included (set 2) substantial margin is obtained. It is noted that the nuclides included in set 3 (Tc99 and Rh103) only give a marginal benefit.

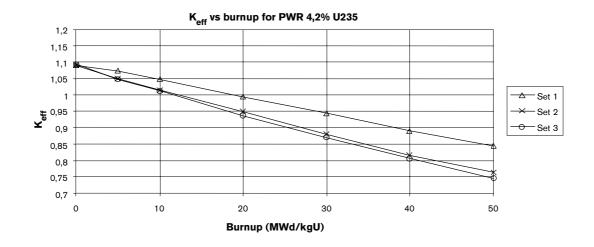
This study, based on state of the art methods and a reasonable assessment of the uncertainties, show that burnup credit is a possible way to demonstrate control of the reactivity in the canisters using a minimum set of nuclides. Additional actinides and selected fission products give more margin.

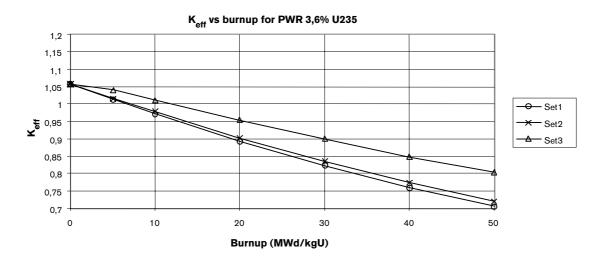
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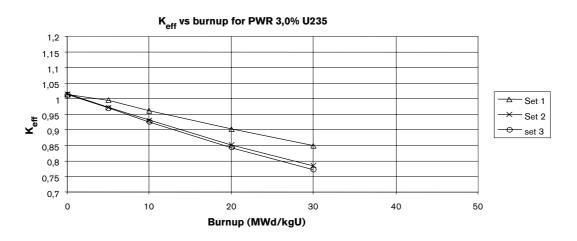
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Appendix 1

$\mathbf{K}_{\mathrm{eff}}$ in a PWR-storage canister vs burnup for different enrichments



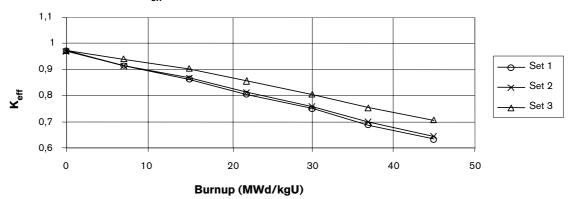




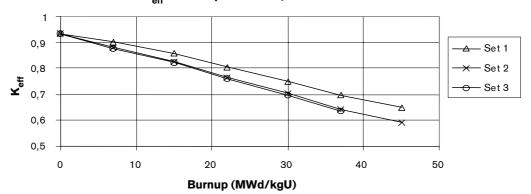
Appendix 2

$\mathbf{K}_{\mathrm{eff}}$ in a BWR-storage canister vs burnup for different enrichments

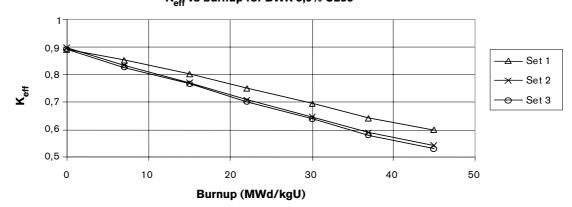




${ m K}_{ m eff}$ vs burnup for BWR 3,6% U235



${ m K}_{ m eff}$ vs burnup for BWR 3,0% U235



Burnup credit – credit for actinides and fission products – selection of isotopes

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1 Introduction

Criticality calculations on final storage canisters have been performed by SKB AB. It appears the burnup credit has to be used to meet the criticality requirements for the suggested canister designs.

It was decided to produce a short report to summarize the international state of the art concerning the knowledge of burnup credit in particular concerning the credit of actinides and fission products.

2 Status of burnup credit

In Sweden burnup credit was studied in 1990–93 (reference A1). The purpose was to evaluate if burnup credit was an acceptable method to increase the spent nuclear fuel storage capacity in CLAB. The result of the evaluation was that burnup credit would give a sufficient reactivity margin. The conclusion of the study was then that burnup credit alone was not a suitable way to control reactivity in CLAB at this time. In this study credit for both fission products and actinides was used. The philosophy was that if the isotopes were stable their reactivity effect could be credited. The nuclear data for the isotopes were considered well known, based on the fact that calculations of core reactivity conditions and power distributions were done in good agreement with measurements at the plants.

This study was also used to estimate the possibility of using burnup credit in the canisters for final storage. The result was that burnup credit certainly would give enough criticality margin in the analysed canister designs (reference A2).

Internationally burnup credit has been used for PWR-storage racks in nuclear plants. The validations of nuclear data are limited to fresh fuel critical experiments and core follow calculations. In US the uncertainties in nuclear data is offset by the requirement that the water in the storage pools should contain 2,000 ppm boron.

For BWR storage racks full burnup credit is not used. Instead credit for burnable poison is used, which requires a burnup of around one cycle where the reactivity peak occurs.

Limited burnup credit in transport casks is approved in France and Germany.

3 Ongoing work on burnup credit

OECD/NEA has initiated a working group of burnup credit issues. The work started in 1991 and was divided in five phases of which three phases are completed. In this project 20 participants from eleven countries are doing benchmark work in order to compare different computer codes, data collections and methods (reference A3 and A4).

IAEA has started an international working group with the purpose to monitor work in the burnup credit field. The work has resulted in a report summarizing the status of the work in burnup credit in 12 countries (reference A5)

NRC has initiated a process called PIRT (Phenomena Identification Rankning Table) with the purpose to identify and rank phenomena important to the burnup credit issue. The work is reviewed in reference A6.

4 Regulatory position I US

In US NRC has issued Interim Staff Guidance 8 (ISG 8, reference A11). The following recommendations are summarized:

Limited partial credit for the reactivity effect of fuel burn up can be taken. Use method in DOE report in reference A7. Assume 50% of the verified and adjusted burnup level from plant records. This applies to intact PWR fuel only. Include actinide effects only.

In practice this guide means that only the following isotopes should be used in burnup credit:

U234, U235, U238, Pu239, Pu240, Pu240, Pu241, Pu242, Am41 (Np237 and U236 not included due to poor validation)

5 Current status concerning data

In the irradiation process around 1,300 isotopes are created in the irradiated fuel.

All isotopes can not be represented in the models and need to establish a limited set to use in burnup credit.

The importance of different isotopes has been assessed in several studies. The reactivity worth of different isotopes vary with fuel design, initial enrichment, operational history and cooling time. The important nuclides seem, however to remain the same.

In reference A8 it is shown that the actinides are contributing with 70% of the absorption at short decay times. This is reduced to 50% at long decay times. The following actinides are included:

U234, U236, U235, U238, Pu239, Pu240, Pu241, Pu242, Am241, Am243 and Np237.

To select which isotopes to credit the following requirements could be established as criteria:

- 1. Knowledge of nuclear data.
- 2. Knowledge about the chemical form, physical form and characteristics, solubility and volatility.
- 3. Verifiability of the isotopic content in the spent fuel.

In reference A6 it is suggested that the following isotopes are candidates to be used in burnup credit:

U234, U236, U235, U238 Pu238, Pu239, Pu240, Pu241, Pu242 Am241, Am243 Np237

Mo95, Tc99, Ru101, Rh103, Ag109, Cs133, Nd143, Nd145 Sm147, Sm149, Sm150, Sm151,Sm152 Eu151, Eu153, Gd155

In nuclear data there are uncertainties in cross-sections. Data on nuclides half-lifes are well known. In reference A7 fresh critical experiments and chemical assay data was used to verify nuclear data and codes.

Concerning chemical data the knowledge of which isotopics are stable and will remain in the fuel matrix for very long times are reasonably good.

Concerning the validation of isotopic data the situation is different. The verification is done by comparing isotopic prediction against experimental data (reference A9 and A10). The fission product measurements publicly available seem presently to be limited and predictions for these nuclides could not be considered to be fully validated. This is in fact the major reason for the position in ISG8.

According to reference A12 several experiments are going on that would cover the gaps in the current databases concerning fission products. Examples of experiments are:

Valduc, PIE data, Ceres, Jaeri BUC, Ariannde&Rebus and LWR Proteus Phase II.

Unfortunately results from these experiments are not open but proprietary or insufficiently reported to be used for validation work.

The number of isotopes that are verified with enough data seem to be limited to the following actinides:

U234, U235, U238, Pu238, Pu239, Pu240, Pu241, Pu242, Am241, Am243

6 Calculations

To assess the importance of different nuclides from a reactivity standpoint criticality calculations were done.

Based on the evaluation above and additional information the following sets of nuclides were selected for criticality analysis.

Set 1: U234, U235, U238, Pu239, Pu240, Pu240, Pu241, Pu242, Am241

Set 2: Set 1 + U236

Set 3: Set 2 + Am243, Np237 Nd143, Nd145, Sm147, Sm149, Sm150, Sm151, Sm152, Eu151, Eu153, Gd155

These three sets were recommended above. Additional nuclides were analysed based on the availability of measured data (PNL), references A13–A15.

Set 4: Set 3 + Sr90, Cs135, Cs137

Set 5: Set 4 + Cm243, Cm244

Set 6: Set 5 + I129

First the isotopic content was calculated at different burnups using input data from section 4 of the main report. SAS2H was used to calculate the isotopic concentrations.

The six sets of nuclides were extracted from the isotopics calculation and analysed in the BWR-CSAS25-model for 3.6% initial enrichment and 22 MWd/kgU burnup in canister geometry. The resulting k_{eff} -values are shown in Table 1.

It can be seen that U236 and Am243, Np237 + selected fission products (Sets 2 and 3) give a significant contribution to the reactivity reduction. Sr90, Cs135 and Cs137 (set 4) give a very small reduction. The net effect of Cm243 and Cm244 is zero. The change due to I129 is not significant.

Based on these calculations and in the evaluation above, it was decided to use the following sets of nuclides in the evaluation of burnup credit in the canisters.

Set 1: Actinides only: U234, U235, U238, Pu239, Pu240, Pu241, Pu242, Am241

Set 2: Actinides and selected fission products: Set 1 + U236, Am243, Np237, Nd143, Nd145, Sm147, Sm149, Sm150, Sm151, Sm152, Eu151, Eu153, Gd155

It was also suggested to include a third set consisting of set 2 + Tc99 and Rh103.

Table 1. Criticality results.

Set no	K _{eff}	Diff
1	0.8149	
2	0.8116	-0.0033
3	0.7873	-0.0243
4	0.7858	-0.0015
5	0.7858	0.0000
6	0.7852	-0.0006

7 Recommendations

It is reasonable to require that isotopic prediction in spent nuclear fuel is verified by comparison with experimental data if the nuclides are used in burnup credit. This provides a robust basis for canister design and final storage.

If this position is accepted the following nuclides could presently be used for burnup credit in final storage canisters:

U234, U235, U238, Pu239, Pu240, Pu241, Pu242, Am241

This number of nuclides is more limited than the ones credited in our previous study of burnup credit in the canisters (reference A2).

It is therefore suggested that a criticality calculation is performed crediting only the above limited number of isotopes to assess if burnup credit still is sufficient to control reactivity. If not efforts should focus on validating additional actinides and fission products.

Pending on experimental data and further validation the following nuclides have a potential to be used for burnup credit in the future:

U236, Am243, Np237, Nd143, Nd 145, Sm147, Sm149, Sm150, Sm151,Sm152 Eu151, Eu153, Gd155 Tc99, Rh103

It is recommended that the developments in these areas are followed up in order to include more nuclides in burnup credit for the final storage canisters.

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