

Radiation levels and absorbed doses around copper canisters containing spent LWR fuel

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ASEA-ATOM, Västerås, Sweden, 1982-08-11

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RADIATION LEVELS AND ABSORBED DOSES AROUND COPPER CANISTERS CONTAINING SPENT LWR FUEL

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This report concerns a study which was conducted for SKBF/KBS. The conclusions and viewpoints presented in the report are those of the author(s) and do not necessarily coincide with those of the client.

A list of other reports published in this series during 1982, is attached at the end of this report. Information on KBS technical reports from 1977-1978 (TR 121), 1979 (TR 79-28), 1980 (TR 80-26) and 1981 (TR 81-17) is available through SKBF/KBS.

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Tife!/Title	Radiation levels canisters contair	and absorbed doses ning spent LWR fuel	around copper
Sammanfattning Abstract			
	Spent fuel from S closed in canister final repository. of canisters and water (causing ra ported in KBS TR- to enclose BWR/PW canisters with th term calculations concept for makin All this has give shielding calcula report.	wedish LWRs is pla ers of copper and h The radiation lev the radiation ener diolysis) have pre- 106. However, ther IR fuel with higher inner walls. New r have been made av g canisters has al en rise to a need f tions, which are p	nned to be en- be buried in the rels during handling rey absorbed in eviously been re- re exists a desire adiation source railable and a new so been discussed. For new radiation presented in this
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l Introduction

> Spent fuel from Swedish LWRs is planned to be enclosed in canisters of copper and to be buried in the final repository. Two types of canister have been discussed:

- Rods from spent BWR or PWR fuel are placed in a cylindrical canister of copper. Fuel corresponding to about 1.4 (BWR) or 1.1 (PWR) tonnes of uranium are assumed to fit in one canister. The rest of the space in the canister is filled with lead before sealing.
- Seven BWR or two PWR fuel assemblies are placed in a cylindrical canister of copper. With the help of hot isostatic pressure (HIP) the rest of the space is filled with copper and the canister is sealed.

The above two types of canister are in the following description designated as $\underline{KBS-2}$ and $\underline{KBS-3}$, respectively.

The radiation levels during handling of canisters and the radiation energy absorbed in water (causing radiolysis) have previously been reported (reference 1). A KBS-2 canister with wall thickness 20 cm, average fuel burnup of 30 Mwd/kg U (BWR fuel) and a cooling time of 40 years before putting into canisters were assumed.

There exists a desire to enclose both BWR and PWR fuel with higher burnup and use canisters with thinner walls. New radiation source term calculations have also been made available (reference 2), giving rise to a need for new radiation calculations. Together with KBS it has been decided to perform calculations for the following set of parameters:

Type of canister KBS-2							KBS-3	
Type of fuel	BWR	BWR	BWR	PWR	PWR	PWR	BWR	PWR
Burnup (Mwd/kgU)	33	33	33	33	38	45	33	38
Wall thickness (cm)	1	10	20	10	10	10	6	6

The fuel is assumed to be put into canisters after a cooling period of 40 years but the influence of a shorter period (30 years) has also been examined. The calculatiion of radiation energy absorbed in water outside the canister is performed for the period 40 to 10^6 years after discharge of the fuel from the reactor. Absorbed dose in the event of any water inside the canisters (in direct contact with the uranium) has also been estimated. This is done assuming a decay period of 40 or 10^5 years.

2 Radiation source terms

> The formation of fission products and actinides in LWR fuel during operation and the resulting gamma and neutron source terms have been calculated with the ORIGEN-2 computer code. These calculations are described in reference 2. The photon spectra have been divided into 10 energy groups with fixed mean energies. The neutrons originate from heavy isotopes with relatively short spontaneous fission half-lives or (α, n) reactions in 0-18 and 0-17 present in UO₂fuel. These two types of neutrons emitted are treated separately because of different spectra.

> The upper part of a fuel rod contains springs of stainless steel.An assembly also contains a top tie plate of the same material. These components are neutron irradiated during reactor operation causing induced activity. After some years of operation the dominant gamma emitter is CO-60. This gamma source is small compared with the fuel itself but is important for the radiation levels in the upward axial direction of the canisters and has therefore been calculated using the computer code AKTGAMMA (reference 3). The cobalt content in the stainless steel is assumed to be 0.05 or 0.2 % for BWR or PWR fuel, respectively (In ASEA-ATOM fuel the cobalt content is restricted to 0.05 %.)

When some of the neutrons emitted from the fuel are captured in the different materials around the fuel, hard energy photons will be emitted. The capture rate in the different materials has been calculated together with the neutron transport calculations. These capture rates have been combined with gamma spectra from references 4 and 5 to give gamma source terms.

Gamma and neutron transport calculations

The gamma transport calculations have been carried out with the point kernel codes CYLGAM and CYLGAX (reference 6). The one-dimensional S_n code ANISN (reference 7) has been used for the neutron transport calculations. Conversion factors from reference 8 have been used to obtain dose rates (mSv/h) from neutron fluxes (n/cm², s). The deposition of energy to water outside the canister due to elastic scattering of neutrons was calculated from the neutron fluxes. It was found that more than 90 % of that energy was due to collisions with hydrogen. Together with the neutron fluxes, neutron capture rates in different materials were calculated with ANISN (giving the gamma source terms from neutron capture, see section 2).

The canisters and the source regions have been homogenized, assuming cylindrical geometry. Transport calculations have been carried out both in the mid radial and the upward axial directions. The treated geometries and the homogenized regions are described in table 1 and 2.

All cases except one are assumed to give a radiation field with rotational symmetry. The exception is a KBS-3 canister with 2 PWR assemblies, having higher radiation levels in two directions. The calculated values correspond to these maximum directions and are estimated to be about a factor of two higher than the average value in the radial direction.

Radiation levels during handling of canisters

With the source terms described in section 2 and the calculation methods described in section 3 the radiation levels outside the canisters have been calculated both in the mid radial and upward axial directions. Dose rates at 3 different distances from the canisters (at contact, 1 m from and 2,5 m from) are presented in table 3, 4 and 5.

The handling of canisters is assumed to take place after a decay period of 40 years. A decay period of only 30 years was found to increase the gamma and the neutron dose rates in the radial direction by 42 % and 56 %, respectively. From the results could also be seen, that both a thicker copper shield and a higher burnup level increase the relative importance of the neutrons. No big difference is seen between canisters containing BWR or PWR fuel. Neutron induced activity (i.e. Co-60) in stainless steel was found to give the major contribution to the gamma dose rates in the upward direction.

The axial distribution of the gamma source in the fuel is closely related to the burnup profile. This is considered by using typical burnup profiles for BWR and PWR fuel. Cm-244 is the dominant neutron emitter at a decay time of 40 years. The relation between burnup and Cm-244 concentration is shown in reference 1, which has been used to estimate the axial neutron source profile.

5.

Radiation energy absorbed in water outside the canisters

The canisters are assumed to be surrended by a layer of clay mineral called Bentonite in the final repository. This mineral is assumed to have a dry density of 1.75 g/cm^3 with a typical chemical analysis shown in table 6 (from reference 9). The amount of mechanically held water is assumed to be 20 % or 0.35 g/cm^3 . The radiation that penetrates the surface of the copper canisters causes radiolysis of the water. In order to permit quantitative evaluations of whether the radiolysis products can destroy the integrity of the canisters, a calculation has been made of the radiation energy absorbed (absorbed dose) in the water. This is done for the period 40 to 106 years after discharge of the fuel from the reactor. The absorbed dose is expressed in the SI unit gray (Gy) (1 Gy = 100 rad = = 1 Ws/kg = 6.24 \cdot 10^{15} eV/g).

Dose rates (mGy/h) as a function of time after discharge are presented in figures 1-8. This is done both for positions on the surface and 10 cm from the surface of the canisters and the contributions from gamma and neutrons are shown individually. The increase of gamma dose rates between 10^4 and $2 \cdot 10^5$ years is due to the buildup of Ra-226 in the fuel.

The calculated dose rates have been numerically integrated, giving accumulated doses (Gy) to water outside the canisters. These are presented as a function of time in figures 9-16 and as a function of distance from the surface of the canisters in figures 17-24. It is obvious from the figures that the buildup of Ra-226 causes a significant increase of accumulated dose between $3 \cdot 10^4$ and 10^6 years. The dose due to neutrons decreases somewhat faster with the distance from the surface than the gamma dose.

The total energy depositions (MWs) to water outside the canisters have been obtained by integrating the accumulated doses over the amount of water outside the canisters. The results are presented as a function of time in figures 25-32 and as a function of distance from the surface of the canisters in figures 33-40. 45-75 % of the gamma and 80-85 % of the neutron energy deposition to water take place in the first 10 cm of the Bentonite layer outside the canisters.

Radiation energy absorbed in water inside the canisters

A rough estimate of the radiation energy deposition to water in the event of direct contact with the UO₂ fuel has been made. The basis for calculations has been a KBS-2 canister loaded with BWR (33 MWd/kgU) or PWR (38 and 45 MWd/kg U) fuel and decay periods of 40 or 10^5 years.

The results are divided into contribution from \propto -, β - and γ -radiation (the contribution from neutrons can be neglected) and are presented in table 7. The figures are valid for the case of water in a narrow crack in the UO₂-pellet. The crack width should not exceed the ranges of the \propto - and β -particles in water, which for the energies considered are:

 α -particles: 0.02 - 0.075 mm β -particles: 1 - 20 mm

If, instead, the water is in a thin layer on the outer surface of a pellet, the contribution from \propto and β will be halved while the contribution from γ is unaffected. Water outside the cladding of a fuel rod will be affected mainly by the χ -radiation.

7 Discussion

> The calculation results in this report have been compared with the results in the previous report (reference 1). This comparison is summarized in table 8.

A very good agreement is obtained for the neutron radiation levels outside a 20 cm KBS-2 canister loaded with BWR fuel, especially if a correction is made for the difference in burnup. The difference in gamma radiation level is due to different gamma source term representations in the computer codes used (ORIGEN-2 in this report and BEGAFTP in reference 1). At a decay time of 40 years Cs-137 is the dominant gamma emitter. The fixed mean group energy used in ORIGEN-2 (0.575 MeV instead of the true 0.662 for Cs-137) explains the difference and the gamma radiation levels given in reference 1 are therefore probably more accurate. The difference will be less in the case of a thinner copper shield.

Reasonably good agreement is reached for calculated energy depositions to water outside a 20 cm KBS-2 canister loaded with BWR fuel apart from the contribution from neutrons during the time period $10^4 - 10^6$ years. The very complete neutron source term library in ORIGEN - 2 was not available at the time for the previous calculations. Therefore the results presented in this report should be regarded as more accurate.

The above comparison indicates an uncertainty in presented values of the order of a factor of two. 8 <u>References</u>

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Calculation models for KBS-2 canisters

Radial direction

R (cm)	Composition (g/cm ³)
0 - 2.8	Pb 11.34
2.8 - 3.3	Cu 8.93
3.3 - 18.5	$\begin{array}{c ccccccccccccccccccccccccccccccccccc$
18.5 - x	Cu 8.93

Upward axial direction

Z (CM)	Composition (g/cm ³)
0-365	$\begin{array}{cccccccccccccccccccccccccccccccccccc$
365-390	BWR:Pb4.70PWR:Pb6.23Zr0.90Zr0.69SS0.61SS0.46Cu0.20Cu0.20
390-394	Pb 11.34
394 - x	Cu 8.93

-

Calculation models for KBS-3 canisters

Radial_direction_(7_BWR_assemblies)

R (cm)	Composition	(g/cm ³)
0-24.5	Cu 6.47 UO ₂ 2.05 Zr ² 0.46	
24.5-30.5	Cu 8.93	

Radial direction (1 of 2 PWR assemblies)

R (cm)	Composition	(g/cm ³)
0 - 13.6	Cu 6.05 UO ₂ 2.47 Zr ² 0.49	
13.6 - 19.6	Cu 8.93	

Upward axial direction (7 BWR assemblies)

Z (cm)	Composition	(g/cm ³)
0 - 365	Cu 6.47 UO ₂ .05 Zr ² 0.46	
365 - 390	Cu 6.47 Zr 0.46 SS 0.30	
390 - 409	Cu 8.19 SS 0.31	
409 - 415	Cu 8.93	

(cont.)

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(<u>TABLE 2</u> cont.) Upward axial direction (2 PWR assemblies)
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Z (CM)	Composition (g/cm ³)
0 - 365	Cu 6.05 UO 2.47 Zr ² 0.49
365 - 385	Cu 6.05 Zr 2.47 SS 0.55
385 - 405	Cu 7.62 SS 0.88
405 - 411	Cu 8.93

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TABLE 3

Radiation levels

(mSv/h)

in

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radial

direction outside

		1					1		T
Fuel type		BWR	BWR	BWR	BWR	PWR	PWR	PWR	لم ا
Burnup (MWd/kgU)		33	33	33	33	33	38	45	KBS
Decay period	(years)	40	40	40	30	40	40	40	-2
Copper wall	(cm)	20	10	1	10	10	10	10	canist
At contact	gamma	0.11	38	2.0E4 ¹⁾	54	29	34	40	le r
	neutrons	0.93	4.3	8.1	6.7	2.4	4.5	10	
	total	1.0	42	2.0E4	61	31	39	50	
l m from surface	gamma	0.029	8.1	2500	12	6.2	7.2	8.6	
	neutrons	0.26	0.92	1.0	1.4	0.51	0.96	2.1	
	total	0.29	9.0	2500	13	6.7	8.2	11	
2.5 m from	gamma	0.014	3.5	810	4.9	2.7	3.1	3.7	ţ
Sarrace	neutrons	0.12	0.40	0.33	0.62	0.22	0.41	0.92	
i	total	0.13	3.9	810	5.5	2.9	3.5	4.6	
				1	1	1		1	1

1) 2.0 E4 stands for 2.0 \cdot 10⁴

t		······································						
Fuel type		BWR	BWR	BWR	BWR	PWR	PWR	PWR
Burnup (MWd/	kgU)	33	33	33	33	33	38	45
Decay period	(years)	40	40	40	30	40	40	40
Copper wall	(cm)	16	10	2	16	16	16	16
	- 18 - 19 - 19 - 19 - 19 - 19 - 19 - 19							
At contact	gamma	0.010	0.14	5.1	0.034	0.013	0.015	0.019
	neutrons	0.039	0.082	0.19	0.060	0.029	0.054	0.12
	total	0.049	0.22	5.3	0.094	0.042	0.069	0.14
1 m from surface	gamma	0.0012	0.016	0.44	0.0044	0.0017	0.0019	0.0023
	neutrons	0.0072	0.014	0.026	0.011	0.0055	0↓010	0.023
	total	0.0084	0.030	0.47	0.015	0.0072	0.012	0.025
2.5 m from	gamma	0.00026	0.0032	0.081	0.00094	0.00035	0.00041	0.00059
Surrace	neutrons	0.0019	0.0034	0.0062	0.0030	0.0014	0.0027	0.0059
i	total	0.0022	0.0066	0.087	0.0039	0.0018	0.0031	0.0065

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TABLE 4

Radiation levels

(mSv/h)

in the upward axial direction

outside a

KBS-2 canister

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		Mid radial		Upward a:	xial	
Fuel type		BWR	PWR ¹⁾	BWR	PWR	
Burnup (Mw	d/kgU)	33	38	33	38	
Decay peri	od (years)	40	40	40	40	
Copper wall	(cm)	6	6	6	6	
At contact	gamma	410	510	3.0	9.0	
	neutrons	2.1	2.3	0.0030	0.0043	
	total	410	510	3.0	9.0	
l m from surface	gamma	91	80	0.25	0.42	
	neutrons	0.47	0.36	0.00077	0.0013	
	total	91	80	0.25	0.42	
2.5 m from surface	gamma	37	31	0.051	0.076	
i	neutrons	0.13	0.14	0.00022	0.00031	
	total	37	31	0.051	0.076	

1) Not a radiation field with rotational symmetry. The radiation levels in the two maximum directions are given.

|_ upward axial direction outside Radiation levels (mSv/h) i n the a mid mid radial and KBS-3 canister radial

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TABLE 6Typical chemical analysis of clay mineral Bentonite,
moisture free basis (from reference 9)

Percent by Wt. (Varies between)

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Silica (SiO ₂)	58.0	64.0
Alumina (Al ₂ 0 ₃)	18.0	21.0
Ferric Oxide (Fe ₂ 0 ₃)	2.5	2.8
Magnesia (MgO)	2.5	3.2
Lime (CaO)	0.1	1.0
Soda (Na ₂ 0)	1.5	2.7
Potash(K ₂ 0)	0.2	0.4
Ferrous Oxide (FeO <u>)</u>	0.2	0.4
Titanium Oxide(TiO ₂)	0.1	0.2
Other minor consti- tuents	0.5	0.8
Chemically-held water (H ₂ O)		5.64

TABLE 7

Absorbed dose rate (Gy/h) in water in a narrow crack in an UO₂-pellet

Canister Type of fuel Burnup (MWd/k	:gU)	KBS-2 BWR 33	KBS-2 PWR 38	KBS-2 PWR 45
Decay period 40 years	کر ای ک	2000 700 63	2300 800 55	3300 900 65
Decay period 10 ⁵ years	x P X	5.4 1.0 11.10 ⁻³	6.2 1.0 9.6.10 ⁻³	7.5 1.2 12·10 ⁻³

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TABLE 8

Comparison between calculation results in this report and the previous evaluation (reference 1)

Radiation levels (mSv/h) outside a KBS-2 canister

		_	
This	report	Reference	1

Fuel type		BWR	BWR
Burnup (Mwd/kgU)		33	34/30 ¹⁾
Decay period (years)		40	40
Copper wall (cm)		20	20
At contact	gamma	0.1	.1 0.17
	neutrons	0.9	0.40-0.95
	total	1.0	0.57-1.1
1 m from	gamma	0.0	29 0.046
suriace	neutrons	0.2	6 0.11-0.26
	total	0.2	9 0.16-0.31

Energy deposition (MWs) to water outside a KBS-2 canister

Fuel type	BWR	BWR
Burnup (MWd/kgU)	33	34/30 ¹⁾
Copper wall (cm)	20	20
40 - 10 ⁴ years gamma neutrons total	0.073 0.032 0.11	0.13 0.055 0.18
40-10 ⁶ years gamma	4.2	4.6
neutrons	1.5	0.12
total	5.7	4.7

 34 MWd/kgU for the gamma and 30 MWd/kgU for the neutron calculations.



























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