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Gamma and neutron dose rates on the outer surface of the nuclear waste disposal canisters

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GAMMA AND NEUTRON DOSE RATES ON THE OUTER SURFACE OF THE NUCLEAR WASTE DISPOSAL CANISTERS

Tiivistelmä – Abstract

Gamma and neutron dose rates on the outer surface of the new versions of the canisters designed for the final disposal of the Finnish spent nuclear fuel have been calculated with the MCNP4A code based on the Monte Carlo technique and with the MARMER code, which applies the standard point-kernel method. The gamma dose rates calculated by the programs are in a satisfactory agreement.

Two canister types, one for hexagonal VVER-440 fuel assemblies from the Loviisa nuclear power plant ("IVO canister") and the other for square BWR fuel bundles from the Olkiluoto nuclear power plant ("TVO canister") have been studied. The canister types are quite similar.

The maximum gamma dose rate on the outer surface of the IVO canister designed to contain 11 VVER-440 fuel assemblies is about 250 mSv/h, when the discharge burnup of the spent fuel is 42 MWd/kgU and the cooling time 20 years. For the TVO canister loaded with 11 BWR fuel bundles having the discharge burnup of 45 MWd/kgU and having cooled for 20 years, the corresponding value is about 150 mSv/h. In both cases the average dose rates are about one half of the maximum values.

The neutron dose rates around the canisters are much lower than the gamma dose rates, typically less than 10 mSv/h.

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SÄTEILYTASOT KÄYTETYN YDINPOLTTOAINEEN LOPPUSIJOITUSKAPSELIEN ULKOPINNALLA

Tiivistelmä - Abstract

Säteilytasot suomalaisilta ydinvoimalaitoksilta kertyvän käytetyn ydinpolttoaineen loppusijoituskapselin ulkopuolella on laskettu sekä Monte Carlo -tekniikkaan perustuvalla MCNP4A-ohjelmalla että tavanomaista pisteydinfunktiomenettelyä soveltavalla MARMER-ohjelmalla. Tutkimuksessa on tarkasteltu kahta kapselityyppiä, joiden perusratkaisut, kuten ulkohalkaisija ja materiaalit, ovat samanlaisia. Erot kapselien välillä aiheutuvat niihin sijoitettavaksi aiotun ydinpolttoaineen geometriasta. Toiseen kapseliin (IVO-kapseli) ladataan Loviisan ydinvoimalaitokselta kertyviä kuusikulmaisia VVER-440-elementtejä ja toiseen (TVO-kapseli) Olkiluodon kahdella BWR-yksiköllä käytettäviä neliöllisiä nippuja.

Maksimigamma-annosnopeus IVO-kapselin ulkopuolella on noin 250 mSv/h, kun nippujen poistopalamaksi oletetaan 42 MWd/kgU ja jäähtymisajaksi 20 vuotta. Yhtä kauan varastoiduilla, mutta palamaan 45 MWd/kgU säteilytetyillä BWR-nipuilla ladatun TVO-kapselin ulkopuolella maksimigamma-annosnopeus on noin 150 mSv/h. Atsimuuttikulman yli lasketut keskimääräiset annosnopeudet ovat noin puolet maksimiannosnopeuksista.

Neutroniannosnopeudet ovat huomattavasti pienempiä kuin gamma-annosnopeudet, tutkituissa tapauksissa aina vähemmän kuin 10 mSv/h.

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1 INTRODUCTION

According to the present plans the spent fuel from the Finnish nuclear power reactors (TVO I and II at the Olkiluoto nuclear power plant and Loviisa 1 and 2 at the Loviisa plant) will be placed into copper/iron canisters for final disposal deep in the Finnish bedrock. A canister consists of a copper mantle and a massive nodular cast iron insert. In the cast iron insert there are holes, where up to eleven fuel bundles can be placed. There are two quite similar types of canisters: one for square BWR fuel bundles of the TVO reactors and another for hexagonal VVER-440 (PWR) fuel assemblies of the Loviisa units (later called the TVO and IVO canisters, respectively).

The gamma and neutron radiation field around a spent fuel disposal canister is one of the factors, which must be taken into account, when the thicknesses of the canister walls and the radiation shields of the spent fuel disposal facilities are being decided. In the following, the main results of dose rate calculations of the present canister designs are reported and discussed. The average and maximum dose rates on the outer surface of the canisters have been of special interest. Their dependence on the discharge burnup and the cooling time of the spent nuclear fuel has been studied, too.

Dose rate calculations have been performed with the very sophisticated MCNP4A Monte Carlo code and the quite simple MARMER point kernel code. Results of the two codes are in good agreement.

2 COMPUTER CODES AND THEIR DATA LIBRARIES

2.1 MCNP4A

MCNP4A is according to its User's Manual "a general-purpose, continuous-energy, generalized geometry, time-dependent, coupled neutron-photon-electron Monte Carlo transport code system" (RSIC CCC-200; Briesmeister). A user can apply the code to quite complicated problems almost without any approximations and get accurate results in a reasonable time when having modern workstations or PCs.

The cross section sets of the standard MCNP4A data library based on the ENDF/B-V evaluated data library was used in these calculations.

2.2 MARMER

MARMER is a point-kernel shielding code having many options not normally available in computer codes of this kind (Kloosterman). With the MARS geometry package one can define quite complicated source and shield structures. A flux integration procedure based on the Monte Carlo technique is used to perform integration over energy range and source volume.

The data libraries of MARMER are based on the JEF 1.1 evaluated data library. The buildup factors are, however, based on the library compiled by the ANSI standard 6.4.3 committee (Kloosterman. p. 6; Trubey&Harima).

2.3 Flux-to-dose conversion factors

The radiation transport and shielding codes calculate first the gamma and/or neutron flux distribution in and around a given source volume(s). Then the dose rate distributions are generated by multiplying each flux value with a flux-to-dose conversion factor. The conversion factors depend on the type and energy of the radiation. There are several sets of conversion factors, but the most commonly used set is most probably the American National Standard 6.1.1, which has been updated time to time (ANSI/ANS-6.1.1-1977 ja ANSI/ANS-6.1.1-1991). The dose (rate) calculated according to this standard is the (biological) effective dose equivalent, which is the absorbed dose multiplied by the so-called quality factor.

The ANSI/ANS-6.1.1-1977 conversion factors have been programmed in the MARMER code. With MCNP4A the user can freely choose the data sets. In this study, both ANSI/ANS conversion sets were used. To calculate neutron dose rates some other data sets were given in MCNP4A input files, too.

The flux-to-dose conversion factors can not be very accurate due to difficulties encountered when estimating biological effects of radiation. Regarding gamma radiation, two above mentioned ANSI/ANS standards differ by about 20% in the energy range from one half to one MeV, which was of the most interest for this study, the updated 1991 version predicting lower dose rates. For neutron radiation, the uncertainty may even be larger.

Furthermore, ICRP took already in 1985 the position that the so-called quality factors for neutron radiation should be increased by a factor of two. This recommendation was still under review, when the ANSI/ANS-6.1.1-1991 conversion factors for neutron radiation were approved, and were not taken into account. Afterwards, many countries, including Finland (STUK, Guide ST1.2), have officially accepted the new quality factors.

For this study, the official ANSI/ANS-6.1.1-1991 conversion factors for neutrons were multiplied by a factor of two and the dose rates reported in Section 4.3 correspond to these doubled flux-to-dose factors. However, it is worth noting that even with the new quality factors the application of the 1991 data set results in smaller (by about 20-25%) dose rates than the 1977 data set.

3 INPUT DATA

3.1 Geometry and material composition of the final disposal canisters

The horizontal cross sections of the TVO and IVO canisters are shown in Fig. 1. The canister versions are in this respect very similar, the biggest differences being the form and size of the holes in the cast iron insert, in which the spent fuel assemblies will be placed. The TVO canister is also longer than the IVO canister. The canisters have room for eleven bundles. The TVO canister can contain ten normal-size bundles and four ABB Atom's SVEA subbundles. One difference worth mentioning is that the eight holes of the IVO canister are placed axisymmetrically, but in the TVO canister there are two different four-hole groups. The canisters will be filled with inert gas.

The following data describe the horizontal layouts of the canisters:

A) Copper mantle:

- Outer radius	49.1 cm *
- Thickness of the mantle	5.0 cm
- Density of copper	8.9 g/cm^3

* In the calculations the outer radius of the mantle was set to be 49.0 cm, because the gap of 0.1 cm between the copper mantle and the cast iron insert was omitted.

B) Iron insert:

- Outer radius	44.0 cm
- Density of nodular cast iron	7.1 g/cm^3
- Composition of cast iron	
- MARMER calculations:	pure iron
- MCNP4A calculations:	iron 92.8wt%; carbon 3.2wt%; magnesium 0.05wt%; silicon 2.15 wt%; manganese 0.8 wt%, nickel 1.0 wt% (Werme & Ericsson 1995)

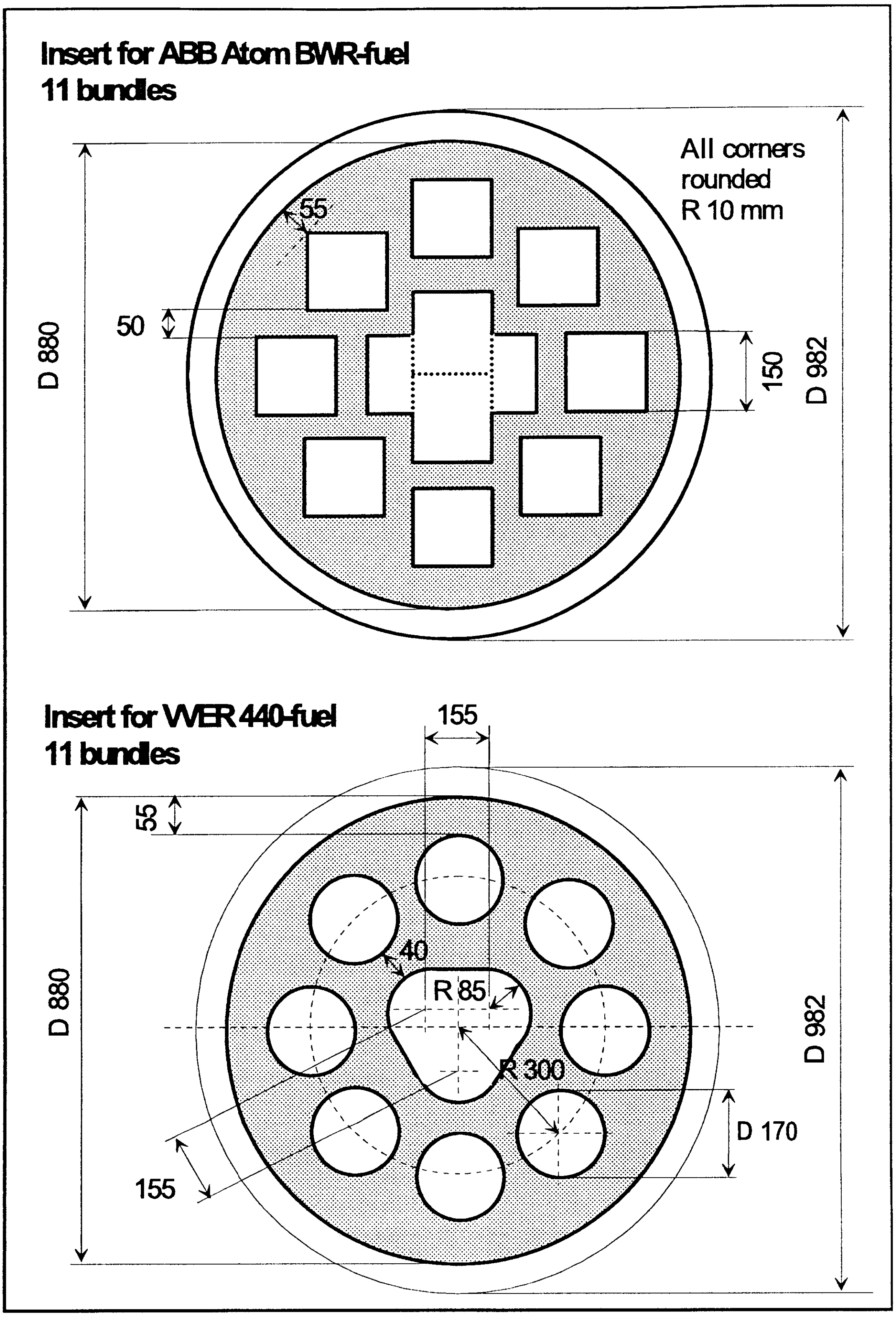

Figure 1. *Radial cross sections of the spent fuel disposal canisters for the TVO (BWR) and IVO (VVER-440) fuel.*

The canisters were assumed to be homogenous in the axial direction, when dose rates on the outer surface of the canisters were calculated. Even then some data concerning the whole canisters and the fuel bundles had to be defined:

	Canister type TVO	IVO
- Bundle geometry	Square	Hexagonal
- Bundles in an assembly	11	11
- Pitch	13.4 cm	14.4 cm
- Flow channel with a bundle	No	Yes
- Length of the fuel rod	3.68 m	2.42 m
- Uranium per bundle	174 kg	120 kg
- Uranium per unit length	5.2 kg/cm	5.5 kg/cm
- Number of fuel rods in a bundle	81**	126

** One water rod replaced by a fuel rod (The TVO fuel bundle was assumed to be of the 9x9-1 type as in Ref. (Anttila 1992)).

The canister and bundle geometry was described almost exactly in the MCNP calculations. The only minor exception was that the gap between the fuel pellet and the clad was homogenized with the clad.

In principle, one could define the canister geometry exactly for MARMER calculations, too. However, it was considered to be too complicated a procedure and a very simple geometric model was constructed for the TVO canister.

The TVO canister was assumed to be loaded with the Siemens 9x9-1 bundles with the flow channel. A fuel rod can then be defined as follows:

- Radius of the fuel pellet	0.455 cm
- Inner radius of the clads	0.4650 cm
- Outer radius of the clads	0.5375 cm
- Pitch of the square pin cell	1.445 cm
- Density of the UO_2	10.2 g/cm^3
- Density of the (Zr) clads	6.55 g/cm^3

All 81 fuel rods (see above) were described for MARMER calculations by a homogenous square, the volume and mass of which were equal of those of all 81 fuel rods. The homogenized square had the following characteristics:

- The side length 8.574 cm
- The material composition:

	Density (g/cm^3)	Number density ($atoms/cm^3$)
- Uranium	6.443	$0.01631 \cdot 10^{24}$
- Oxygen	0.866	$0.03260 \cdot 10^{24}$
- Zirconium	1.648	$0.01088 \cdot 10^{24}$

In all shielding calculations, the fuel was assumed to be fresh uraniumdioxide.

3.2 Photon and neutron sources

3.2.1 Average photon and neutron sources

The photon and neutron sources and their spectra were taken from the ORIGEN 2.1 calculations (Anttila 1992 and Anttila 1995). In the former reference data for the typical spent TVO fuel are given and the latter report contains information of the spent IVO fuel.

The photon production in the IVO and TVO spent fuel for two discharge burnups and four cooling times are given in Table 1. ORIGEN 2.1 calculates the total and nuclide-wise photon source strengths in eighteen gamma energy groups, but the lowest and highest groups contribute negligibly to gamma dose rates outside the IVO and TVO canisters (however, they were included in the MCNP input files). In fact, one nuclide ^{137m}Ba (the short lived daughter of ^{137}Cs) generates more than one half of the total gamma dose rate on the outer surface of the canisters. Its concentration depends almost linearly on the discharge burnup and is almost independent on the fuel type.

The ORIGEN 2.1 program normalizes the photon source terms to the average energy of each group in such a way that the total gamma energy production in each group is preserved. The procedure may not be the best possible for shielding calculations.

Therefore, the source terms of ^{137m}Ba were always renormalized to the correct energy (0.662 MeV). For MCNP calculations the same procedure was also applied to ^{154}Eu.

The neutron production data of the spent IVO and TVO fuel are given in Table 2. Most of the neutrons are produced by spontaneous fissions of ^{244}Cm at the cooling times, which are of interest for this study. Therefore, all the neutrons were assumed to be produced by that reaction, i.e. the energy spectrum was always set to be that of the spontaneous fission of ^{244}Cm. The concentration of ^{244}Cm is strongly dependent of the fuel type and the discharge burnup. The latter dependence is of type (Würz):

$$C = A \cdot BU^B,$$

where C is the curium concentration,
BU is the discharge burnup,
A is a case-dependent constant and
B is a constant varying from 4.5 to 4.8 when calculated according to the ^{244}Cm concentrations of the references (Anttila 1992) and (Anttila 1995).

An increase of burnup by 10% could, therefore, result in an increase of the neutron source strength by 50-60%.

Table 1. Photon source strength (1/s/tU) and spectrum of the spent IVO and TVO fuel at four cooling times according to ORIGEN-2 calculations.

I Spent IVO fuel

a) Discharge burnup 36 MWd/kgU (initial uranium enrichment of 3.6 wt%)

Mean energy of the gamma group (MeV)	Cooling time (a) 10.	20.	30.	40.
0.125	$2.275 \cdot 10^{14}$	$1.449 \cdot 10^{14}$	$1.015 \cdot 10^{14}$	$7.475 \cdot 10^{13}$
0.225	$2.027 \cdot 10^{14}$	$1.490 \cdot 10^{14}$	$1.149 \cdot 10^{14}$	$8.956 \cdot 10^{13}$
0.375	$9.801 \cdot 10^{13}$	$6.270 \cdot 10^{13}$	$4.803 \cdot 10^{13}$	$3.761 \cdot 10^{13}$
0.575	$3.630 \cdot 10^{15}$	$2.618 \cdot 10^{15}$	$2.066 \cdot 10^{15}$	$1.638 \cdot 10^{15}$
0.662 (^{137m}Ba)	$3.234 \cdot 10^{15}$	$2.567 \cdot 10^{15}$	$2.037 \cdot 10^{15}$	$1.617 \cdot 10^{15}$
0.850	$3.133 \cdot 10^{14}$	$5.584 \cdot 10^{13}$	$2.573 \cdot 10^{13}$	$1.455 \cdot 10^{13}$
1.250	$2.213 \cdot 10^{14}$	$7.467 \cdot 10^{13}$	$2.903 \cdot 10^{13}$	$1.241 \cdot 10^{13}$
1.750	$3.227 \cdot 10^{12}$	$1.498 \cdot 10^{12}$	$7.534 \cdot 10^{11}$	$4.030 \cdot 10^{11}$
2.250	$8.062 \cdot 10^{10}$	$2.598 \cdot 10^{8}$	$8.893 \cdot 10^{7}$	$4.610 \cdot 10^{7}$
2.750	$5.265 \cdot 10^{9}$	$4.477 \cdot 10^{8}$	$4.039 \cdot 10^{8}$	$3.642 \cdot 10^{8}$
3.500	$6.553 \cdot 10^{8}$	$2.010 \cdot 10^{7}$	$1.346 \cdot 10^{7}$	$9.378 \cdot 10^{6}$

b) Discharge burnup 42 MWd/kgU (initial uranium enrichment of 3.6 wt%)

Mean energy of the gamma group (MeV)	Cooling time (a) 10.	20.	30.	40.
0.125	$2.758 \cdot 10^{14}$	$1.715 \cdot 10^{14}$	$1.181 \cdot 10^{14}$	$8.600 \cdot 10^{13}$
0.225	$2.328 \cdot 10^{14}$	$1.697 \cdot 10^{14}$	$1.303 \cdot 10^{14}$	$1.014 \cdot 10^{14}$
0.375	$1.125 \cdot 10^{14}$	$7.091 \cdot 10^{13}$	$5.417 \cdot 10^{13}$	$4.237 \cdot 10^{13}$
0.575	$4.301 \cdot 10^{15}$	$3.058 \cdot 10^{15}$	$2.410 \cdot 10^{15}$	$1.911 \cdot 10^{15}$
0.662 (^{137m}Ba)	$3.774 \cdot 10^{15}$	$2.995 \cdot 10^{15}$	$2.377 \cdot 10^{15}$	$1.887 \cdot 10^{15}$
0.850	$4.232 \cdot 10^{14}$	$7.313 \cdot 10^{13}$	$3.285 \cdot 10^{13}$	$1.813 \cdot 10^{13}$
1.250	$2.819 \cdot 10^{14}$	$9.573 \cdot 10^{13}$	$3.743 \cdot 10^{13}$	$1.600 \cdot 10^{13}$
1.750	$4.274 \cdot 10^{12}$	$1.966 \cdot 10^{12}$	$9.728 \cdot 10^{11}$	$5.092 \cdot 10^{11}$
2.250	$9.631 \cdot 10^{10}$	$3.413 \cdot 10^{8}$	$1.254 \cdot 10^{8}$	$6.802 \cdot 10^{7}$
2.750	$6.733 \cdot 10^{9}$	$6.442 \cdot 10^{8}$	$5.796 \cdot 10^{8}$	$5.208 \cdot 10^{8}$
3.500	$8.497 \cdot 10^{8}$	$3.954 \cdot 10^{7}$	$2.672 \cdot 10^{7}$	$1.854 \cdot 10^{7}$

Table 1 (cont.)

II Spent TVO fuel

a) Discharge burnup 36 MWd/kgU (Initial uranium enrichment of 3.3 wt%)

Mean energy of the gamma group (MeV)	Cooling time (a)			
	10.	20.	30.	40.
0.125	$2.281 \cdot 10^{14}$	$1.427 \cdot 10^{14}$	$9.860 \cdot 10^{13}$	$7.196 \cdot 10^{13}$
0.225	$1.936 \cdot 10^{14}$	$1.419 \cdot 10^{14}$	$1.091 \cdot 10^{14}$	$8.493 \cdot 10^{13}$
0.375	$9.399 \cdot 10^{13}$	$5.939 \cdot 10^{13}$	$4.537 \cdot 10^{13}$	$3.550 \cdot 10^{13}$
0.575	$3.575 \cdot 10^{15}$	$2.574 \cdot 10^{15}$	$2.030 \cdot 10^{15}$	$1.610 \cdot 10^{15}$
0.662 (^{137m}Ba)	$3.179 \cdot 10^{15}$	$2.523 \cdot 10^{15}$	$2.003 \cdot 10^{15}$	$1.590 \cdot 10^{15}$
0.850	$3.200 \cdot 10^{14}$	$5.798 \cdot 10^{13}$	$2.594 \cdot 10^{13}$	$1.393 \cdot 10^{13}$
1.250	$1.505 \cdot 10^{14}$	$5.723 \cdot 10^{13}$	$2.499 \cdot 10^{13}$	$1.159 \cdot 10^{13}$
1.750	$3.461 \cdot 10^{12}$	$1.606 \cdot 10^{12}$	$7.965 \cdot 10^{11}$	$4.184 \cdot 10^{11}$
2.250	$5.938 \cdot 10^{10}$	$1.626 \cdot 10^{8}$	$7.482 \cdot 10^{7}$	$4.901 \cdot 10^{7}$
2.750	$4.552 \cdot 10^{9}$	$5.696 \cdot 10^{8}$	$5.131 \cdot 10^{8}$	$4.613 \cdot 10^{8}$
3.500	$5.664 \cdot 10^{8}$	$3.330 \cdot 10^{7}$	$2.260 \cdot 10^{7}$	$1.566 \cdot 10^{7}$

b) Discharge burnup 45 MWd/kgU (Initial uranium enrichment of 3.8 %)

Mean energy of the gamma group (MeV)	Cooling time (a)			
	10.	20.	30.	40.
0.125	$2.944 \cdot 10^{14}$	$1.816 \cdot 10^{14}$	$1.240 \cdot 10^{14}$	$8.981 \cdot 10^{13}$
0.225	$2.403 \cdot 10^{14}$	$1.759 \cdot 10^{14}$	$1.350 \cdot 10^{14}$	$1.049 \cdot 10^{14}$
0.375	$1.140 \cdot 10^{14}$	$7.311 \cdot 10^{13}$	$5.590 \cdot 10^{13}$	$4.372 \cdot 10^{13}$
0.575	$4.461 \cdot 10^{15}$	$3.182 \cdot 10^{15}$	$2.509 \cdot 10^{15}$	$1.989 \cdot 10^{15}$
0.662 (^{137m}Ba)	$3.928 \cdot 10^{15}$	$3.117 \cdot 10^{15}$	$2.474 \cdot 10^{15}$	$1.964 \cdot 10^{15}$
0.850	$4.396 \cdot 10^{14}$	$7.866 \cdot 10^{13}$	$3.473 \cdot 10^{13}$	$1.839 \cdot 10^{13}$
1.250	$2.006 \cdot 10^{14}$	$7.681 \cdot 10^{13}$	$3.366 \cdot 10^{13}$	$1.559 \cdot 10^{13}$
1.750	$4.709 \cdot 10^{12}$	$2.182 \cdot 10^{12}$	$1.073 \cdot 10^{12}$	$5.561 \cdot 10^{11}$
2.250	$6.228 \cdot 10^{10}$	$2.304 \cdot 10^{8}$	$1.174 \cdot 10^{8}$	$7.817 \cdot 10^{7}$
2.750	$5.274 \cdot 10^{9}$	$9.340 \cdot 10^{8}$	$8.413 \cdot 10^{8}$	$7.553 \cdot 10^{8}$
3.500	$6.528 \cdot 10^{8}$	$6.070 \cdot 10^{7}$	$4.144 \cdot 10^{7}$	$2.870 \cdot 10^{7}$

Table 2. Neutron source strength (n/s/tU) of the IVO and TVO spent fuel at four cooling times.

I IVO spent fuel

a) The IVO spent fuel with the discharge burnup of 36 MWd/kgU (The original uranium enrichment of 3.6 wt%)

	Cooling time (a)			
	10.	20.	30.	40.
(α,n)-reaction	$7.940 \cdot 10^{6}$	$8.076 \cdot 10^{6}$	$8.015 \cdot 10^{6}$	$7.852 \cdot 10^{6}$
Spontaneous fission	$2.740 \cdot 10^{8}$	$1.884 \cdot 10^{8}$	$1.300 \cdot 10^{8}$	$9.015 \cdot 10^{7}$
Total	$2.820 \cdot 10^{8}$	$1.965 \cdot 10^{8}$	$1.380 \cdot 10^{8}$	$9.800 \cdot 10^{7}$
Contribution of ^{244}Cm	$2.713 \cdot 10^{8}$	$1.850 \cdot 10^{8}$	$1.262 \cdot 10^{8}$	$8.607 \cdot 10^{7}$

b) The IVO spent fuel with the discharge burnup of 42 MWd/kgU (The original uranium enrichment of 3.6 wt%)

	Cooling time (a)			
	10.	20.	30.	40.
(α,n)-reaction	$1.181 \cdot 10^{7}$	$1.125 \cdot 10^{7}$	$1.068 \cdot 10^{7}$	$1.014 \cdot 10^{7}$
Spontaneous fission	$5.478 \cdot 10^{8}$	$3.762 \cdot 10^{8}$	$2.591 \cdot 10^{8}$	$1.793 \cdot 10^{8}$
Total	$5.596 \cdot 10^{8}$	$3.874 \cdot 10^{8}$	$2.698 \cdot 10^{8}$	$1.895 \cdot 10^{8}$
Contribution of ^{244}Cm	$5.438 \cdot 10^{8}$	$3.709 \cdot 10^{8}$	$2.529 \cdot 10^{8}$	$1.725 \cdot 10^{8}$

Table 2 (cont.)

II TVO spent fuel

a) The TVO spent fuel with the discharge burnup of 36 MWd/kgU (The original uranium enrichment of 3.3 wt%)

	Cooling time (a) 10.	20.	30.	40.
(α,n)-reaction	$1.063 \cdot 10^7$	$1.029 \cdot 10^7$	$9.876 \cdot 10^6$	$9.451 \cdot 10^6$
Spontaneous fission	$4.634 \cdot 10^8$	$3.181 \cdot 10^8$	$2.190 \cdot 10^8$	$1.514 \cdot 10^8$
Total	$4.740 \cdot 10^8$	$3.284 \cdot 10^8$	$2.289 \cdot 10^8$	$1.609 \cdot 10^8$
Contribution of ^{244}Cm	$4.604 \cdot 10^8$	$3.140 \cdot 10^8$	$2.142 \cdot 10^8$	$1.460 \cdot 10^8$

b) The TVO spent fuel with the discharge burnup of 45 MWd/kgU (The original uranium enrichment of 3.8 wt%)

	Cooling time (a) 10.	20.	30.	40.
(α,n)-reaction	$1.638 \cdot 10^7$	$1.489 \cdot 10^7$	$1.365 \cdot 10^7$	$1.262 \cdot 10^7$
Spontaneous fission	$8.517 \cdot 10^8$	$5.847 \cdot 10^8$	$4.027 \cdot 10^8$	$2.785 \cdot 10^8$
Total	$8.680 \cdot 10^8$	$5.996 \cdot 10^8$	$4.163 \cdot 10^8$	$2.912 \cdot 10^8$
Contribution of ^{244}Cm	$8.458 \cdot 10^8$	$5.768 \cdot 10^8$	$3.933 \cdot 10^8$	$2.682 \cdot 10^8$

3.2.2 Axial and radial variation of source strengths

In the previous section the photon and neutron source strengths were given at some burnup points, which were chosen to represent the average and maximum assemblywise burnups of the spent IVO and TVO fuel. However, in a spent fuel assembly the burnup may vary both axially and radially. An average axial burnup distribution for a recently discharged IVO and TVO fuel batch is given in Tables 3.a and 3.b, respectively.

Table 3.a *The axial burnup distribution of the fuel assembly batch (109 bundles) removed from the Loviisa-1 reactor in Summer 1995 (Antila 1995).*

The axial burnup distribution
(ten axial nodes of equal length)

Axial form factors	Absolute values (MWd/kgU)
.66	22.7
1.00	34.5
1.11	38.1
1.13	39.1
1.14	39.3
1.14	39.2
1.12	38.5
1.07	37.0
.94	32.5
.59	20.5
Average values:	
1.00	34.5

The axial form factor (the ratio between the highest and average node burnup) is about 1.14 for the spent IVO fuel. The radial form factor is ca. 1.12 (Antila 1995). The highest pin burnups of a VVER-440 assembly typically occur in the outermost pin layer. At the axial midpoint of those fuel pins the burnups could be about 28% higher than the average assemblywise burnup.

Table 3.b The axial burnup and void history distribution of the fuel assemblies removed from the TVO I reactor in Spring 1995 (Solala 1995).

The axial distribution
(25 axial nodes of equal length)

Node	Average burnup (MWd/kgU)	Average void history* (%)
1 (top)	9.6	70.2
2	24.0	69.7
3	29.6	79.0
4	33.6	79.2
5	36.5	78.6
6	38.5	77.7
7	39.9	76.0
8	41.0	74.2
9	41.8	72.2
10	42.4	70.2
11	42.9	68.0
12	43.3	65.6
13	43.7	62.9
14	43.9	59.8
15	44.1	56.2
16	44.1	52.1
17	44.1	47.5
18	44.1	42.4
19	43.8	36.9
20	43.3	31.1
21	42.3	24.5
22	40.4	18.0
23	36.3	13.6
24	27.8	13.7
25	6.6	15.6
Average	37.1	

* The void history contains also a contribution of the control rod history.

The lowest and highest nodes represent the so-called axial blankets (reflectors) made of natural uranium. If their contribution is omitted, the average burnup of the batch is 39.6 MWd/kgU. At the axial midplane the typical average burnups are about 44 MWd/kgU.

The radial form factor of a spent TVO fuel bundle might typically be as large as in spent IVO fuel bundles, i.e. about 1.10 and might also be found in the periphery of the assembly.

4 RESULTS

4.1 Introduction

The main goal of this study was to calculate gamma and neutron dose rates on the outer surface of the spent fuel disposal canisters at the axial midplane. Both the maximum and average values were of interest. The dependence of the dose rates on the discharge burnup and the cooling time as well as on the position of the ring of eight holes were studied. With the MCNP4A code an exact comparison between the IVO and TVO canisters could also be carried out. The MARMER computer program was used to calculate maximum dose rates on the surface of the TVO canister in an approximate way.

4.2 Gamma dose rates

4.2.1 Preliminary studies with MCNP4A

The cast iron and copper (as well as the uranium fuel) are rather good materials for gamma radiation shields. Therefore, one can assume that at any point on the outer surface of the canisters studied the gamma dose is determined by the nearest fuel bundle (or at eight specific points by two nearest bundles). The contribution from the bundles at the centre of the canisters should be very small. These assumptions were proven to be correct with a few MCNP4A calculations.

With MCNP4A it was also proven that the orientation of the bundles in an IVO canister has rather small an effect on the surface gamma dose rate and its azimuthal distribution.

4.2.2 Gamma dose rate on the axial midplane of the outer surface of the canisters

In Tables 4.a and 4.b the gamma dose rates at the axial midplane of the outer surface of the IVO and TVO spent fuel disposal canisters are given. In each case the photon source was assumed to be homogeneous both radially and axially. In Table 4.c a typical azimuthal distribution of the gamma dose rate are given for the TVO canister. The same information for both canister types is presented in Fig. 3.

The average surface gamma dose rate of the IVO canister is much higher than that of the TVO canister, if the discharge burnup of the spent fuel is the same in both cases. This results from the geometric differences in the canister designs and to some extent also from the fact in the IVO canister there are a little more uranium per unit height than in the TVO canister.

In both cases, the gamma dose rate depends almost linearly on the discharge burnup.

The ratio between the maximum and average gamma dose rate is about 2.5 for the TVO canister having the nominal geometry. If the ring of the eight holes are moved inward, the ratio decreases. However, based on Table 4.c one can conclude that turning the 'corner' holes by 45 degrees, the maximum gamma dose could decreased by a factor of two.

The same ratio is less than 2.0 (the exact value depends on how narrow an azimuthal sector one is studying) for the IVO canister. In this case there is no other option to reduce the peak gamma dose rate on the outer surface than to move all eight holes of the outer ring inward. Based on Table 4.a it can be estimated that a move by ca. 1.6 cm will decrease the surface dose rate by a factor of about two.

The application of the ANSI/ANS-6.1.1-1991 standard results in gamma dose rates, which are about 20% lower than those predicted by the older data set.

Table 4.a *Average gamma dose rate at the axial midplane of the outer surface of the IVO final disposal canister according to MCNP4A calculations.*

	Average gamma dose rate (mSv/h)			
	a		b	
Case	1977	1991	1977	1991
1	134	106	163	128
2	88	69	106	84

1 Nominal canister geometry
2 The outer ring of eights holes moved 1 cm radially inward

a Discharge burnup of 36 MWd/kgU; Cooling time of 20 years
b Discharge burnup of 42 MWd/kgU; Cooling time of 20 years

1977 Flux-to-dose conversion factors based on the ANSI/ANS-6.1.1-1977 standard
1991 Flux-to-dose conversion factors based on the ANSI/ANS-6.1.1-1991 standard

Table 4.b Average and maximum gamma dose rate at the axial midplane of the outer surface of the TVO final disposal canister according to MCNP4A calculations.

	Gamma dose rate (mSv/h)			
	Maximum		Average	
Case	1977	1991	1977	1991
1 a	154	121	62	49
b	193	154	79	62
2 a	77	60	36	28
b	100	79	46	36

1 Nominal canister geometry (see Fig. 1)

2 The thickness of the cast iron layer between the centre hole and the eight holes of the outer ring decreased by 1 cm (the center point of the hole moved by 1.41 cm towards the centre of the canister)

a Discharge burnup of 36 MWd/kgU; Cooling time of 20 years

b Discharge burnup of 45 MWd/kgU; Cooling time of 20 years

1977 Flux-to-dose conversion factors based on the ANSI/ANS-6.1.1-1977 standard

1991 Flux-to-dose conversion factors based on the ANSI/ANS-6.1.1-1991 standard

Table 4.c *The azimuthal distribution of the gamma dose rate at the axial midplane of the outer surface of the TVO final disposal canister according to MCNP4A calculations (The ANSI/ANS-6.1.1-1991 flux-to-dose conversion factors, see Table 4.b).*

The midpoint of the azimuthal sector (Degrees)	Average dose rate in the sector (mSv/h) I		II	
	a	b	a	b
0.000	49	62	29	38
5.625	47	59	28	36
11.250	40	51	26	34
16.875	28	36	20	25
22.500	19	24	14	17
28.125	24	31	14	19
33.750	51	65	29	37
39.375	95	121	49	63
45.000	121	154	60	79
Average	49	62	28	36

I Nominal canister geometry (see Fig. 1)
II The thickness of the cast iron layer between the centre hole and the eight holes of the outer ring decreased by 1 cm

a) Discharge burnup of 36 MWd/kgU; Cooling time of 20 years
b) Discharge burnup of 45 MWd/kgU; Cooling time of 20 years

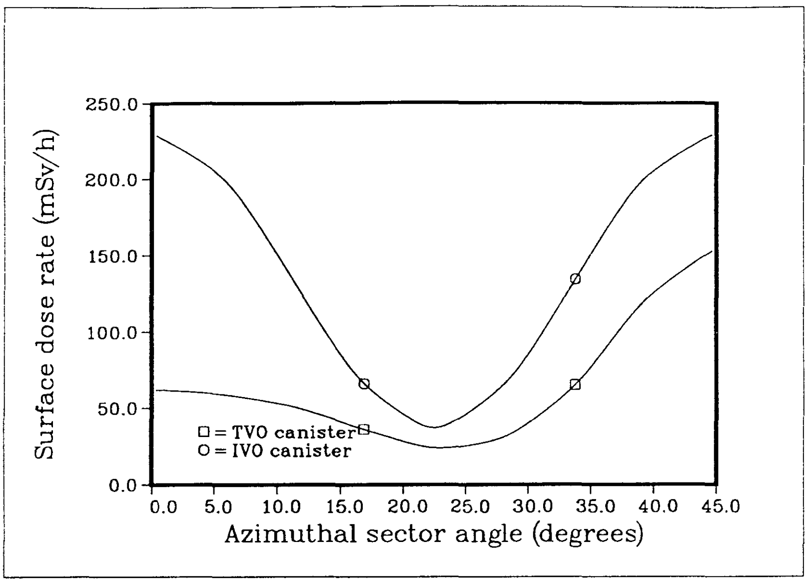

Figure 2. *Gamma dose rate at the axial midplane of the outer surface of the TVO and IVO disposal canisters as a function of the azimuthal angle in a symmetric sector of 45 degrees (TVO fuel: discharge burnup 45 MWd/kgU and cooling time 20 a, IVO fuel: 42 MWd/kgU / 20 a, ANSI/ANS-6.1.1-1991 flux-to-dose conversion factors).*

In Table 5 there are shown the maximum surface gamma dose rates of the TVO canister calculated with MARMER. The agreement between the MARMER and MCNP4A dose rates is very good, especially when taking into account the simple geometric description of the source region applied in MARMER calculations. It indicates that without any filling material in the holes the exact position of the fuel rods of the bundle is of minor importance.

Table 5. Maximum gamma dose rates (mSv/h) on the outer surface of a TVO final disposal canister according to MARMER calculations (Note: In MARMER the ANSI/ANS-6.1.1 flux-to-dose conversion factors are used).

A) Dose rate at the azimuthal angle of 45° (The centre point of the hole at the distance of 27.5 cm from the centre of the canister, see case C below)

Discharge burnup (MWd/kgU)	Cooling time (years)			
	10	20	30	40
36	229	121	82	59
45	282	153	103	76

B Dose rate at the azimuthal angle of 0°

Discharge burnup (MWd/kgU)	Cooling time (years)			
	10	20	30	40
36	107	55	37	27
45	138	70	47	33

C) Dose rate at the azimuthal angle of 45° as a function of the distance of the centre of the hole from the centre of the canister (Discharge burnup of 45 MWd/kgU)

	Cooling time (years)	
Distance	20	30
Nominal geometry (R = 28.28 cm)	201	139
R = 27.89 cm*	171	112
R = 27.50 cm	153	103
R = 26.50 cm**	93	62

* The thickness of the cast iron layer between the bundle hole and the copper wall 5.5 cm

** The cast iron layer between the holes ca. 3.7 cm

In the air the gamma dose rate decreases mainly only due to the geometric factor. In a repository the canister will be surrounded by bentonite. In Table 6 the attenuation of the gamma dose rate around a TVO canister is described in one case (Nominal geometry, discharge burnup of 45 MWd/kgU, cooling time of 20 years). The dose rates are along the radius of the canister extended from the point of the maximum dose rate outward. To halve the gamma dose rate, a 6-7 cm thick layer of bentonite is needed. However, the main part of the gamma energy will be absorbed in the immediate neighbourhood of the canister.

Table 6. Attenuation of the gamma dose rate around the TVO canister surrounded by air or bentonite (Spent fuel: discharge burnup of 45 MWd/kgU and cooling time 20 a).

Distance from the outer surface (cm)	Dose rate (mSv/h)		
	Bentonite		Air
	Dry	Saturated	
0	201	201	201
7.5	61	91	-
10.0	38	61	-
50.0	$6.8 \cdot 10^{-2}$	$2.6 \cdot 10^{-1}$	52
100.0	$6.3 \cdot 10^{-5}$	$6.7 \cdot 10^{-4}$	28

4.2.3 Gamma dose rate above and below the canisters

The IVO and TVO disposal canisters have the similar two-layer lid system at their both ends. The outer lid, the thickness of which is 5 cm, is made of copper and the inner lid with the equal thickness is made of steel.

IVO canister

In a IVO final disposal canister, the so-called bottom and top tail pieces of the flow channels of the inserted fuel assemblies must be assumed to be in contact with the iron lids. Both tail pieces are of almost equal weight (about 9 kg), but the top tail piece is much shorter than the bottom tail piece.

The top and bottom tail pieces of the present IVO fuel assemblies are made of stainless steel. After irradiation in the reactor, the only important photon source in this steel type is ^{60}Co, which emits two photons with relatively high energies (1.173 ja 1.332 MeV) per decay. However, the trace element concentrations of this steel type were not known, when the activity inventory calculations of the IVO spent fuel were carried out. Therefore, also for the cobalt content an assumed value of 350 ppm had to be used (Anttila 1995, Appendix 1). In the activity inventory calculations, the tail pieces were irradiated in the (total) flux, which was half of the average full-power flux in the fuel region. This assumption may lead to an overestimation of the cobalt activity, too.

The real geometry of the top tail piece is rather complicated. Therefore, it was described as a straight tube made of steel (density 7.85 g/cm^3) having the following dimensions:

- length	20 cm,
- inner radius	5 cm and
- outer radius	6.72 cm.

With this definition the weight of the steel tube is about 10 kg.

The gamma dose rates were carried out with the MARMER code. The source density was chosen to correspond to a fuel assembly with the following characteristics:

- initial enrichment of 3.6%,
- discharge burnup of 42 MWd/kgU and
- cooling time of 20 years.

Based on data of Table 6 of Ref. (Anttila 1995) the ^{60}Co activity can be calculated to be $7.52 \cdot 10^8$ Bq/cm^3. The photon source of the MARMER group 19 (1.0 MeV $< E <$ 1.33 MeV; see Appendix 1) was therefore set to $1.50 \cdot 10^8$ photons/cm^3/s.

MARMER calculations gave following gamma dose rates:

- at the centre point of the canister lid	0.28 Sv/h.
- at the centre axis of one fuel assembly in the centre hole	0.27 Sv/h.
- on the outer surface of the canister, at the axial midpoint of the top tail piece	0.18 mSv/h.

The gamma dose rate at the top of the IVO canister is only slightly lower than the maximum dose rate at the axial midplane of the canister (about 0.32-0.33 mSv/h). The importance of the tail pieces increases, if one would fill canisters with fuel assemblies having a shorter cooling time, because the half-life of ^{60}Co is rather short, 5.27 years.

Shortening the cooling period from 20 to 10 years would increase gamma dose rates at both ends of an IVO canister by a factor of about 3.7.

TVO canister

The spent TVO fuel assemblies will be disposed without their flow channels. Therefore, at the bottom of the canister the fuel assemblies stay on the iron lid. In all TVO fuel assemblies there is first a short (ca. 5.4 cm) structural part, which is followed by the so-called axial blanket made of natural or slightly enriched uranium (the fuel assemblies used in the first cycles did not have the axial blankets). The length of the blanket part is about 15 cm. The discharge burnups of the axial blankets are low, typically less than 10 MWd/kgU. The discharge burnusp of the next 15 cm is still lower than the average discharge burnup (see Table 3.b, nodes 1 and 25).

In the upper part of the TVO fuel bundles the distance from the active fuel region to the bundle top is much longer than it is in the lower part. The length of the structural part without any uranium is already ca. 29.5 cm and then there is the handle bar, the height of which may about 10 cm. From point of radiation protection, the lower part of a TVO disposal canister is more important than the upper part.

Gamma dose rates at the bottom of a TVO disposal canister was studied with MARMER code. Due to the rather complicated geometry and material composition, the lower part of the canister was described in a very approximate way. The fuel bundles were homogenized to fill their placement holes. The amounts of materials and the photon source strength per unit length were preserved. The spent fuel was assumed to have a discharge burnup of 45 MWd/kgU and a cooling time of 20 years. The structural part and the axial blanket were omitted. The only variable was the distance from the bottom of fuel region to the lid of the canister. Due to geometry, the maximum gamma dose rate should be at the centre axis of the canister. The dose rate immediately under the canister lid was according to MARMER calculations as follows:

Distance between the fuel region and the lid (cm)	Gamma dose rate (mSv/h) (ANSI/ANS-6.1.1-1977 conversion factors)
15	325
20	260

The dose rates seem to be high, but the calculation model was planned to produce conservative estimates. However, the results indicate that the highest gamma dose rates at the bottom of a TVO canister could be of the same order as those on the axial midplane.

4.3 Neutron dose rates

The neutron dose rates around the copper/iron final disposal canisters were calculated with the Monte Carlo code MCNP4A. The secondary gamma dose rates induced by neutrons were not been taken into account, because they were assumed to be of very minor importance.

The energy spectrum of the source neutrons was assumed to correspond to that of the spontaneous fission of ^{244}Cm (see Ch. 3.21), which according to Ref. (RSIC CCC-200, Appendix H) can be described as follows:

$$f(E)=Ce^{-E/a}\sinh(bE)^{1/2},$$

where C is a normalization factor,
a = 0.906 and
b = 3.848.

The source was defined to be one neutron per second in the canister studied. In a subcritical system the total source is:

$$S_{eff} = S/(1-k),$$

where k is the effective multiplication factor of the system.

MCNP4A was instructed to take automatically into account the effect of neutron multiplication. In the calculations the fuel was assumed to be fresh uraniumdioxide. The multiplication factors of an isolated dry disposal canister studied are then ca. 0.22-0.23 (Anttila 1996). The impact of burnup on the multiplication factors may be rather small. Therefore, the neutron dose rates in and around the canisters are determined mainly by the primary source strengths given in Table 2.

The average neutron dose rates on the outer surface of the axial midplane of the IVO and TVO canisters are given in Table 6 (The dose rates given correspond to the ANSI/ANS-6.1.1.-1991 conversion factors multiplied by two, see Section 2.3).

Table 6. The average neutron dose rates (mSv/h) on the outer surface of the axial mid-plane of the IVO and TVO canisters.

A) IVO canister (enrichment of the fuel 3.6%)

Discharge burnup (MWd/kgU)	Cooling time (years)		
	10	20	30
36	2.0	1.4	1.0
42	4.0	2.8	2.0

B) TVO canister

Discharge burnup (MWd/kgU)	Cooling time (years)		
	10	20	30
36	3.1	2.2	1.5
45	5.7	4.0	2.8

Compared with the dose rates caused by primary gamma radiation, neutron dose rates around a spent fuel disposal canisters are low. The azimuthal variation of the neutron dose rate is also rather small, the maximum value being typically about 10% higher than the average dose rate.

Neutron dose rates outside the canisters are produced by fast neutrons, the energy of which varies from 0.1 to 2 MeV. A few MCNP4A calculations were carried out to study, how effectively concrete or lead glass can attenuate this kind of radiation. A TVO canister was assumed to be surrounded by a 5 cm thick layer of concrete (Wasastjerna 1989) or lead glass, the density of which 3.86 g/cm^3 and the chemical composition of which was Pb_3O_4, 51.23 weight%; SiO_2, 41.53%, K_2O, 7.0 and Ce 0.2%. The MCNP4A calculations gave the following results:

	Relative dose rate
- no extra shield	1.0
- 5 cm concrete	0.52
- 5 cm lead glass	0.67

5 CONCLUSIONS

Gamma and neutron dose rates around the present designs of the canister types for the final disposal of the Finnish spent fuel have been calculated with the MCNP4A code based on the Monte Carlo technique and with the MARMER code, which applies a standard point-kernel method. The gamma dose rates calculated by the programs are in satisfactory agreement.

The maximum gamma dose rate around the IVO canister designed to contain 11 VVER-440 fuel assemblies is about 250 mSv/h, when the discharge burnup of the spent fuel is 42 MWd/kgU and the cooling time 20 years. In case of the TVO canister loaded with 11 BWR fuel bundles having the discharge burnup of 45 MWd/kgU and having cooled for 20 years, the corresponding value is about 150 mSv/h. The average dose rates are about one half of the maximum values in both cases.

The neutron dose rates around the canisters are much lower than the gamma dose rates, always less than 10 mSv/h.

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APPENDIX 1

FLUX-TO-DOSE CONVERSION FACTORS (mSv/hr/(1/cm^2/s)) FOR NEUTRON RADIATION

Note: In the brackets the corresponding quality factors are given.

Neutron energy (MeV)	ICRP-21*		ANSI/ANS-6.1.1 1977*		ANSI/ANS-6.1.1 1991**
$2.5 \cdot 10^{-8}$	$3.85 \cdot 10^{-2}$	(2.3)	$3.67 \cdot 10^{-2}$	(2.0)	$1.44 \cdot 10^{-2}$
$1.0 \cdot 10^{-7}$	$4.17 \cdot 10^{-2}$	(2.0)	$3.67 \cdot 10^{-2}$	(2.0)	$1.58 \cdot 10^{-2}$
$1.0 \cdot 10^{-6}$	$4.55 \cdot 10^{-2}$	(2.0)	$4.46 \cdot 10^{-2}$	(2.0)	$1.74 \cdot 10^{-2}$
$1.0 \cdot 10^{-5}$	$4.35 \cdot 10^{-2}$	(2.0)	$4.54 \cdot 10^{-2}$	(2.0)	$1.60 \cdot 10^{-2}$
$1.0 \cdot 10^{-4}$	$4.17 \cdot 10^{-2}$	(2.0)	$4.18 \cdot 10^{-2}$	(2.0)	$1.49 \cdot 10^{-2}$
$1.0 \cdot 10^{-3}$	$3.70 \cdot 10^{-2}$	(2.0)	$3.76 \cdot 10^{-2}$	(2.0)	$1.38 \cdot 10^{-2}$
$1.0 \cdot 10^{-2}$	$3.57 \cdot 10^{-2}$	(2.0)	$3.56 \cdot 10^{-2}$	(2.5)	$1.63 \cdot 10^{-2}$
$1.0 \cdot 10^{-1}$	$2.08 \cdot 10^{-1}$	(7.4)	$2.17 \cdot 10^{-1}$	(7.5))	$7.13 \cdot 10^{-2}$
$0.5 \cdot 10^{-1}$	$7.14 \cdot 10^{-1}$	(11.0)	$9.26 \cdot 10^{-1}$	(11.0)	$3.13 \cdot 10^{-1}$
1.0	1.18	(10.6)	1.32	(11.0)	$5.15 \cdot 10^{-1}$
2.0	1.43	(9.3)	-		$7.70 \cdot 10^{-1}$
2.5	-	-	1.25	(9.5)	-
5.0	1.47	(7.8)	1.56	(8.0)	1.18
10.0	1.47	(6.8)	1.47	(6.5)	1.48

* Briesmeister, Judith F. (Ed.), MCNP™-A General Monte Carlo N-Particle Transport Code, Version A. Los Alamos National Laboratory, LA-12625-M (November 1993), Appendix H.

** ANSI/ANS-6.1.1-1991, Neutron and gamma-ray fluence-to-dose factors. An American National Standard published by the American Nuclear Society (Approved August 26, 1991)

The quality factors for neutron radiation according to STUK (The Finnish Centre for Nuclear and Radiation Safety), Guide ST1.2 (Application of maximum values for radiation exposures and monitoring of exposures (in Finnish). Second, updated edition, Helsinki 1995, Table A1):

Neutron energy (MeV)	Quality factor
$< 1.0 \cdot 10^{-2}$	5
$1.0 \cdot 10^{-2} < E < 1.0 \cdot 10^{-1}$	10
$1.0 \cdot 10^{-1} < E < 2$	20
$2 < E < 20$	10
> 20	5

APPENDIX 2

GAMMA ENERGY GROUP STRUCTURE OF THE MARMER AND ORIGEN2.1 COMPUTER CODES

MARMER Group no.	MARMER Upper energy (MeV)	ORIGEN2.1 Group no.
14	3.0	14
15	2.5	13
16	2.0	12
17	1.66	"
18	1.50	11
19	1.33	"
20	1.00	10
21	0.80	"
22	0.70	9
23	0.60	"
24	0.512	"
25	0.510	"
26	0.45	8
27	0.40	"
28	0.30	7
29	0.20	"
(30	0.15)	

APPENDIX 3

THE CHEMICAL COMPOSITION OF BENTONITE

(Muurinen 1992, p. 2, Table 1):

Compound	Weight percent
SiO_2	63.0
Al_2O_3	16.1
Fe_2O_3	3.0
CaO	1.1
MgO	1.6
Na_2O	2.2
K_2O	0.48
(sum	87.48)

The density of bentonite was assumed to be 2.78 g/cm^3 . For the MARMER calculations, two kind of bentonite was specified: a 'dry' material, where the water content was set to be 10 vol% and a saturated bentonite with the water content of 43 vol%. Below are the number densities for the 'dry' bentonite:

		Weight percents Other / O	Number densities (10 vol% of water)
SiO_2	63.0	0.4674/0.5326	$1.580 \cdot 10^{22}/3.160 \cdot 10^{22}$
Al_2O_3	16.1	0.5292/0.4708	$4.759 \cdot 10^{21}/7.139 \cdot 10^{21}$
Fe_2O_3	3.0	0.6994/0.3006	$5.662 \cdot 10^{20}/8.494 \cdot 10^{20}$
CaO	1.1	0.7147/0.2853	$2.956 \cdot 10^{20}/2.596 \cdot 10^{20}$
MgO	1.6	0.6031/0.3969	$5.981 \cdot 10^{20}/5.981 \cdot 10^{20}$
Na_2O	2.2	0.7419/0.2581	$1.070 \cdot 10^{21}/5.348 \cdot 10^{20}$
K_2O	0.48	0.8302/0.1698	$1.536 \cdot 10^{20}/7.676 \cdot 10^{19}$

Water (10 vol%) H: $6.68 \cdot 10^{21}$ and O: $3.34 \cdot 10^{21}$; O totally $4.443 \cdot 10^{22}$

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