ACTIVITY INVENTORY OF THE BIOLOGICAL SHIELDS OF THE FINNISH NUCLEAR REACTORS

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ABSTRACT

As part of the decommissioning studies of the four Finnish nuclear reactors (two 465 MWe VVER-440 type PWRs at Loviisa and two 735 MWe BWRs at Olkiluoto) the activation products inventories of the biological shields of the reactors have been estimated. First, material data were updated and improved by analyzing samples taken from the bioshields. Then the radial irradiation flux distribution was calculated with the one-dimensional ANISN transport code using cross sections taken from the BUGLE-80 data library. Finally, the activation and the subsequent cooling of the bioshield materials were calculated with the well-known ORIGEN-S program using its original LWR data libraries. The main result was the total radioactive inventory and its nuclide-wise distribution as a function of the cooling time at different radial locations of the bioshields. Due to the different geometries (shielding) the radioactive inventory at the inner surface of the biological shield of a Loviisa unit will be about two orders of magnitude greater than that of an Olkiluoto (TVO) reactor. To some extent the nuclide-wise distributions are different, too. In the case of the Loviisa bioshields H-3, Fe-55, Co-60 and Ni-63 are the most important radionuclides from the point of a prompt dismantling, which at present is the preferred option for the Loviisa reactors. In the long-term the activity will be dominated by Ca-41 and to a smaller extent C-14, Cl-36, Ar-39 (if it remains in the concrete) and Ni-59. In the bioshields of the TVO reactors H-3 and Ar-39 are very dominant and there also Eu-152 is of relatively great importance. Of the nuclides mentioned above only Co-60 and Eu-152 are gamma-ray emitters.

INTRODUCTION

At present, there are four nuclear power reactors in Finland. The Loviisa Nuclear Power Plant consists of two VVER-440 type, 465 MWe PWRs. The units started commercial operation in 1977 and 1981, respectively. The plant is owned and operated by Imatran Voima Oy (IVO). The other two reactors are located at Olkiluoto. These 735 MWe BWRs of Swedish origin are owned and operated by Teollisuuden Voima Oy (TVO). Their commercial operation was started in 1979 and 1982, respectively.

The Finnish nuclear power companies have carried out decommissioning studies since the beginning of the 1980s and they presented their first decommissioning and dismantling plans in 1987. The plans must be updated and revised after a time period set by the authorities.

As part of the decommissioning studies the activation products inventories of the different reactor components and structures have been calculated (1,2,3). In the following, methods used to estimate activity inventories in the biological shields of the Finnish reactors are described. First, measurements performed to update and improve knowledge of the material compositions of the bioshields are described. Then, methods and approximations applied when calculating irradiation flux distributions are discussed. Finally, activity inventory calculations and their main results are described.

UPDATING OF MATERIAL DATA

The main constituents of the fresh bioshield concretes of the Finnish nuclear reactors have been reported quite well in pouring protocols and other documents. However, the concentrations of various trace elements were not measured during the construction. Therefore, as the first stage of this study the concentrations of fourteen elements (H, Li, B, N, Cl, K, Ca, Co, Ni, Cs, Ba, Sm, Eu, U) were measured using various analytical techniques as appropriate (4). The composition of the bioshield material was considered to be uniform. Concrete samples taken from the upper part of the bioshield were ground and homogenized before analysis.

The contents of Li and B in concrete samples were analyzed by atomic absorption spectrophotometry. Nitrogen contents were analyzed by Kjeldahl-method. The concentrations of hydrogen were derived from measurements of water content in concrete using thermogravimetric analysis.

Other elements were analyzed using instrumental neutron activation analysis. A computer program STOAV 84 (5) was used to calculate the results.

Table I shows the chemical analysis results with error limits. When the content of element is below the detection limit of the analytical method, error limits have been omitted.

There were marked differences in the concentrations of H, Ni and K in the concrete samples from the two reactors. The material from Loviisa had a hydrogen content (15200 ppm) a factor of four higher than from TVO (4600 ppm); this was due to the presence of serpentine minerals with chemically bound water in the Loviisa samples.

CALCULATION OF IRRADIATION FLUXES

Programs and Data Libraries

Neutron flux distributions in and around the active cores of the Loviisa and Olkiluoto (TVO) reactors were estimated with the REPVICS program system, which was originally developed for calculations of fast neutron doses in the reactor pressure vessels of the Loviisa reactors (6). More detailed

TABLE I

Contents of Elements in Bioshield Concrete

Element	TVO(BWR) ppm	IVO(PWR) ppm
H	4600±230	15200±2280
Li	23±3	3.6±0.4
B	2060±103	2000±100
N	33±3	33±3
Cl	58	87
K	16900±2800	<400
Ca	75000±9000	76000±7000
Co	22±1	89±3
Ni	<20	1800±100
Cs	4.2±0.5	0.2±0.05
Ba	930±30	320±40
Sm	4.0±0.4	0.6±0.1
Eu	0.74±0.05	<0.05
U	15.6±0.6	6.7±0.9

descriptions of activation flux calculations can be found elsewhere (7,8).

Because of many other unavoidable approximations and inaccuracies of some input data, one-dimensional flux distributions were considered to be sufficiently accurate for activity inventory calculations. Therefore, the well-known ANISN code was used (9). It is a S_N transport theory program, which can take into account anisotropic scattering. A P_3S_8 approximation was applied in this study.

The cross sections were taken from the BUGLE-80 library, which contains the coupled neutron and gamma constants for many nuclides and elements in 47 neutron and 20 photon groups (10) . Two of the neutron groups are in the thermal range, but upscattering from the lowest group to the 46th group is not taken into account. It may be a rather serious deficiency, when one wants to generate flux data for activation calculations, because the thermal neutron absorption is then the most important nuclide transmutation reaction.

Source Distributions

The activation of the reactor components and structures outside the active core is caused by neutron leakage, which may depend quite strongly on the core loading scheme. It may be difficult and also unreasonable to try and simulate actual irradiation conditions during the whole lifetime of a reactor. A better option is to choose a typical fuel cycle and to use its average neutron source distribution in irradiation flux calculations. This approach was applied in this study. However, the active cores of the Loviisa units were reduced in 1980-81 by replacing 36 fuel assemblies in peripheral core positions by shield assemblies made of steel. Afterwards, the fuel loading scheme was changed from the original Out-In-In scheme to a (very) low-leakage scheme. The effects of these two changes on the activation of the reactor components and structures in the radial direction were studied, too.

Calculation Geometries

The geometries defined for one-dimensional activation flux calculations of the Loviisa and TVO reactors (without some details) are presented in Tables II and III, respectively. There are two facts worth noting. First, in both cases there are air gaps outside the pressure vessel (in and around the thermal shield), which are rather difficult to take accurately into account in one-dimensional calculations. Second, the distance between the active core and the bioshield is much longer in the TVO reactors than in the Loviisa units.

TABLE II

Radial Geometry in the Activity Inventory Calculations of the Loviisa Reactors (Some details ignored)

Component	Radius (cm) Inner	Outer
(Active core	0.	137.0)
Basket	150.5	154.0
Barrel	155.5	161.5
Pressure vessel	177.1	192.0
Thermal shield	223.0	233.0
Biological shield	238.2	305.8
(Common concrete	307.0	590-.0)

TABLE III

Radial Geometry in the Activity Inventory Calculations of the TVO Reactors (Some details ignored)

Component	Radius (cm) Inner	Outer
(Active core	0.	195.0)
Moderator tank	214.5	217.0
Pressure vessel	277.0	291.0
Thermal shield	313.9	334.0
Biological shield	343.0	523.4

Results and Their Accuracy

The main results of the ANISN calculations, one-dimensional 47-group radial neutron flux distributions were condensed to a three-group structure ($0 < E < 0.5\,\text{eV} < E < 1\,\text{MeV} < E < 17.3\,\text{MeV}$). They were also integrated over the separate reactor components or their subzones in order to produce averaged activation fluxes for these regions. The three-group fluxes were used to calculate the three ORIGEN-S flux parameters THERM, RES and FAST (11).

The flux calculation method may not be particularly accurate in the case of the bioshields, which are quite far from the source region. Furthermore, the air gaps between the core regions and the bioshields can cause errors in ANISN-type calculations due to neutron streaming. Thirdly, the BUGLE neutron cross sections are not well-suited to problems, where the thermal energy range is important, as is the case with the activity inventory calculations. However, the degree of uncertainty of neutron fluxes and spectra is very difficult to estimate.

ACTIVITY INVENTORY CALCULATIONS

Radioactive inventory calculations were performed with the ORIGEN-S code of the SCALE-3 program package with its own data libraries (11,12). The LWR data libraries with the three-group cross sections are intended for calculations in or near the reactor core. Their suitability for activation calculations in bioshield conditions may be somewhat doubtful.

The irradiation time was assumed to be 30 years (36 annual cycles with an average load factor of ca. 85%) for the Loviisa reactors and 36 years (40 annual cycles with an average load factor of 90%) for the TVO reactors. The choices were, of course, based on the design lifetimes of the reactors and their actual load factors.

MAIN RESULTS

Total and nuclide-wise activities at different points and regions of the bioshields of the Finnish nuclear reactors as a function of the cooling time were the main results of this study. Figures 1 and 2 are presented here as typical examples of the results. They contain information on the activation products inventory at or near the inner surface of the bioshields.

Due to geometric (shielding) differences the radioactive inventory at the inner surface of the bioshield of a Loviisa unit will be about two orders of magnitude greater than that of a TVO reactor. To some extent the nuclide-wise distributions are different, too. In the Loviisa bioshields H-3, Fe-55, Co-60 and Ni-63 are the most important radionuclides from the point of view of a prompt dismantling, which at present is the preferred option for the Loviisa reactors. In the long-term the activity will be determined by Ca-41 and to a smaller extent C-14, Cl-36, Ar-39 (if it remains in the concrete) and Ni-59. In the bioshields of the TVO reactors H-3 and Ar-39 are very dominant in the successive time periods and there also Eu-152 is of relatively great importance. Of the nuclides mentioned above only Co-60 and Eu-152 are gamma-ray emitters.

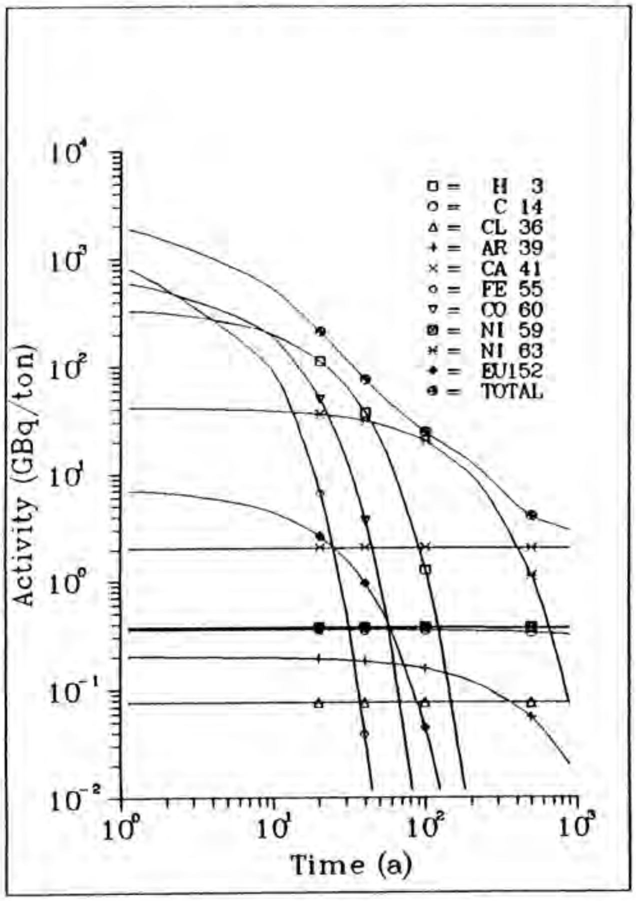

Fig. 1. Inner surface activity in the bioshield (serpentinite concrete) of the Loviisa reactors.

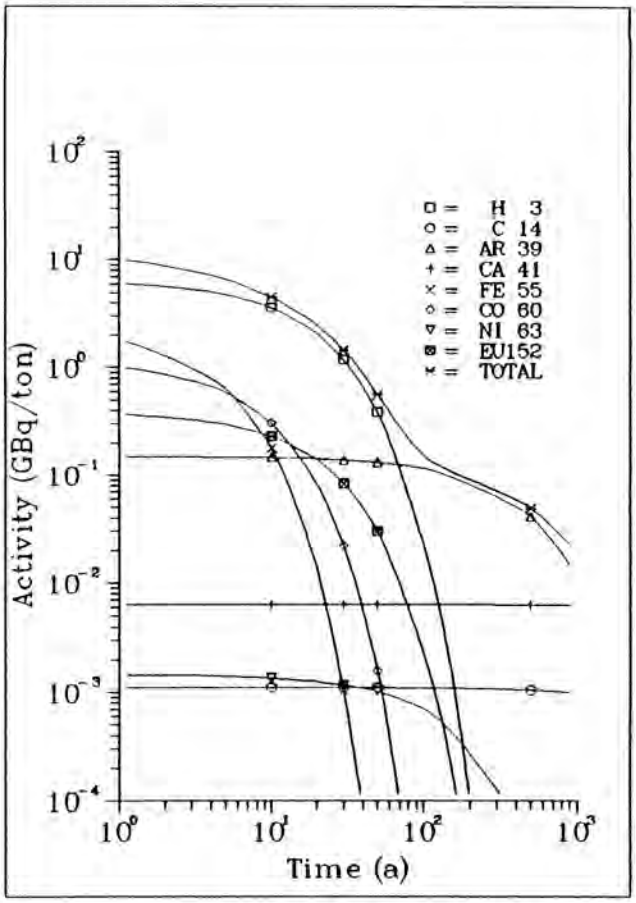

Fig. 2. Inner surface region activity in the bioshields of the TVO reactors.

CONCLUSIONS

The radioactive inventories of the biological shields of the Finnish nuclear reactors have been estimated in a straightforward, but approximate way. The present knowledge on the compositions of the bioshield concretes is satisfactory mainly because of the measurements described in this report. A few checks against other studies indicate that no significant elements have been overlooked. The neutron flux calculations are at present the weakest phase of our procedure. To use a one-dimensional code when trying to estimate neutron flux levels quite far from the source region is certainly not the best possible decision and the BUGLE cross sections may still worsen the situation. Our calculation system will, however, be updated and improved in a near future. The original three-group cross sections of the ORIGEN-S code used in this study have been generated in the active core conditions, which may differ from those in the bioshields. A better option would be to utilize results of accurate neutron flux and spectrum calculations to create a case-specific ORIGEN data library. In conclusion, even if our present calculation system may produce results, which are of the correct order of magnitude, a possibility to verify calculated activities against measured values would be of great importance. In the case of an operating reactor, however, such measurements are difficult, if not infeasible.

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